Abstracts Booklet

Inaugural
Quantum Information Science
Principal Investigators' Meeting

Basic Energy Sciences, Office of Science

June 30, July 1, 2025

Organizer: Dr. Athena Sefat

Location: Hilton Washington DC/Rockville Hotel & Executive Meeting Center

FOREWORD

This is the first Principal Investigators' (PI) meeting for the Quantum Information Science (QIS) program at Office of Basic Energy Sciences (BES) at U.S. Department of Energy (DOE). This PI meeting was sponsored by the Division of Materials Sciences and Engineering (MSE) within BES and held on June 30 and July 1, 2025. This booklet is a collection of abstracts submitted to this PI meeting.

The QIS program focuses on materials sciences and engineering, investigating the fundamental properties of materials critical for developing advanced QIS technologies. Within the BES, this program crosscuts the three MSE Division research areas, namely, Materials Discovery, Design, and Synthesis, Condensed Matter and Materials Physics, and Scattering and Instrumentation Sciences. The research within this QIS program encompasses topics described in BES Roundtable reports: BES Roundtable: Opportunities for Basic Research for Next-Generation Quantum Systems and BES Roundtable on Opportunities for Quantum Computing in Chemical and Materials Sciences. The focus is on advanced materials − both strongly interacting (correlated) and non-interacting (non-correlated) material systems − that support the foundational components for quantum state stability and control. This program aims to create and control entanglement in multi-level quantum systems to enable precise measurement of individual quantum bits while mitigating decoherence – a significant obstacle to reliable quantum computing, quantum networking, and quantum sensing.

This PI meeting brought together close to 100 program-supported researchers, to present their latest scientific breakthroughs, exchange ideas, and foster new collaborations. For BES Program Managers and the participating researchers, the meeting also provided an overview of this program. The meeting format included talks and poster sessions and was organized to include 40 regular presentations and 25 poster presentations.

BES appreciates the contributions of all QIS PIs for presenting their latest research. BES is also grateful for the outstanding support of the Oak Ridge Institute for Science and Education including that of Ms. Stephanie Fox.

Athena S. Sefat, Ph.D.
Program Manager, QIS Program
MSE Dr. Athena Sefat | U.S. DOE Office of Science (SC)

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U.S. Department of Energy (DOE), Office of Science, Basic Energy Science, Quantum Information Science (QIS) Program in Materials Sciences and Engineering (MSE) Division

2025 Quantum Information Sciences Principal Investigators' Meeting

Organizer: Dr. Athena Sefat, Athena.sefat@science.doe.gov
Location: Hilton Washington DC/Rockville Hotel & Executive Meeting Center
Date: June 30 & July 1, 2025

Monday, June 30, 2025

7:30 – 8:25 AM	Registration & *** Breakfast***
8:25 – 8:30 AM	**Welcome to Day 1:** Athena Sefat
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11:00 – 11:20 AM	Stephen Jesse (ORNL), "*Creating and controlling atomic scale quantum systems*"
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2:10 – 2:30 PM	Vlad Pribiag (Univ. of Minnesota), "*Superconductor-semiconductor hybrid quantum devices*"
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4:00 – 4:20 PM	Jennifer Dionne (Stanford Univ.), "*Room-temperature valley-selective emission in Si-MoSe2 heterostructures enabled by high quality-factor chiroptical cavities*"
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Tuesday, July 1, 2025

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11:10 – 11:30 AM	Chris Palmstrom (Univ. of California Santa Barbara), "*Synthesis of improved mobility InAs and InGaAs-quantum wells and of PbSe SAG nanowires*"
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4:00 – 4:20 PM	Archana Kamal (Northwestern University), "*Next-generation parametrically-induced quantum engineering*"
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Monday, June 30, 2025

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3. Shyam Shankar (University of Texas at Austin), "Holographic quantum simulation of strongly correlated electron systems"

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5. Chengyun Hua (ORNL), "Light-matter interaction with quantum light"

6. Joshua Damron (ORNL), "Elucidation of quantum spin defect interactions with quantum materials"

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8. Mark Dean (BNL), "Witnessing quantum entanglement using resonant inelastic X-ray scattering"

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20. Ezekiel Johnston-Halperin (Ohio State Univ.), "Materials development for quantum magnonics"

21. Michael Flatté (University of Iowa), "Entanglement gates enabled by hybrid quantum magnonics"

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25. Arnab Banerjee (Purdue University), "Seeking quasiparticles in perturbed matter from low-energy spin dynamics"

Abstracts

Hybrid Quantum Magnonics for Transduction and Sensing

Award Number: DE-SC0019250

Names of PI, co-PI(s): Gregory Fuchs[1] (PI), Michael Flatté[2] (co-PI), Ezekiel Johnston-Halperin[3] (co-PI)

Affiliations: 1. Cornell University, 2. University of Iowa, 3. Ohio State University
Abstract Title: Entanglement gates enabled by hybrid quantum magnonics

Abstract:Our project focuses on the hybrid quantum magnonics for quantum science and technologies based on the ultra-low loss molecular ferrimagnet vanadium tetracyanoethylene (V[TCNE]$_x$). This molecule-based material has exhibited exceptionally narrow linewidth and magnetic damping coefficient down to $\alpha \approx 4\times10^{-5}$ in thin films at room temperature, which is competitive with any other magnetic material, including the yttrium iron garnet (YIG), the conventional choice. Moreover, V[TCNE]$_x$ can be templated and grown at moderate growth temperatures (~60°C) without changing its loss properties, which is attractive for integration with superconducting quantum technologies. Recent results have demonstrated the integration of patterned magnetic structures into superconducting resonators, yielding strong photon-magnon coupling in the microwave regime in an integrated circuit architecture [1], and the creation of a double disk structure that can be quantitatively read out using nitrogen vacancy (NV) centers in diamond [2]. In this project we have focused on solving the materials, measurement, and theory problems necessary to realize these visions for hybrid quantum magnonic systems.

Here we discuss demonstrations of magnon-qubit coupling following an initial proposal [3] to realize two-qubit entangling gates mediated by virtual excitations of magnons. This was a complementary proposal to a previous qubit-magnon transduction proposal [4]. By studying the self-interaction of a single qubit with magnonic modes of a bar [5] we have validated the theoretical calculations of coupling via fringe fields. Based on the previous proposals this clearly indicates that the coupling strength can be sufficient for realistic configurations and enables qubit entangling gates wherein the qubits are separated by >1 μm.

Shown in Fig. 1 is the surface magnon induced longitudinal relaxation of an NV center. The dramatic increase in the relaxation when the transition is resonant with the surface wave modes provides strong evidence of the effect. The red line is the theoretical calculation, which does not include any adjustments or scaling. Thus we can confidently describe the NV-magnon coupling in a physically realizable geometry. The coupling strongly depends on the characteristics of the spin wave modes and the geometry of the magnetic material. As a result the ability to pattern and template V[TCNE]$_x$ will be essential to realizing highly efficient two-qubit entangling gates using this approach.

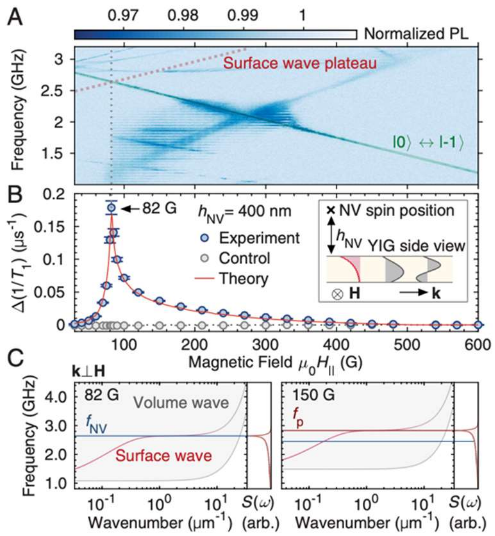

Figure 1. Surface-magnon induced longitudinal relaxation. (A) ODMR of NV centers on magnetic bar. The NV transition and the calculated surface magnon plateau are highlighted by the solid green and the dotted red lines, respectively. Vertical gray line indicates the field where the surface wave plateau is resonant with the NV transition. (B) Longitudinal relaxation rate of NV centers for the 0 to -1 transition. Blue and gray markers represent experiment with and (as control) without magnetic material, red curve is the theoretical calculation. On the vertical axis the Δ indicates a reference of $1/T_1$ to its value at 600 Gauss. NV-magnet distance is 400 nm. Inset shows cross section of the magnet with surface (red) and volume (gray) magnon mode profiles. ***k*** *is the magnon wave vector. (C) Magnon dispersion for* ***k*** *perpendicular to* ***H*** *and noise spectra S(ω) at 82 Gauss (left) and 150 Gauss (right). Surface magnon dispersion is shown with a pink curve with the gray gradient at large k indicating suppression of the surface-wave feature, whereas the gray area shows region of volume modes (see inset of (B)). Sharp peak in S(ω) corresponds to the surface-wave plateau frequency f_p as indicated by horizontal red line. Blue horizontal line indicates NV frequency f_{NV}.*

We will also discuss ongoing efforts to calculate the fringe fields, magnon dynamics, nonlinear features, and other properties of patterned $V[TCNE]_x$, as well as configurations that will enhance magnon-qubit couplings. Studies of the response of $V[TCNE]_x$ to gating are underway and the progress will be outlined.

Project supported References:

[1] Q. Xu *et al.*, Advanced Science **11**, 2310032 (2024). DOI: 10.1002/advs.202310032

[2] B. A. McCullian, M. Chilcote, H. Yusuf, E. Johnston-Halperin, and G. D. Fuchs, In prep. (2025).

[3] M. Fukami, D. R. Candido, D. D. Awschalom, and M. E. Flatté, PRX Quantum **2**, 040314 (2021). DOI: 10.1103/PRXQuantum.2.040314

[4] D. R. Candido, G. D. Fuchs, E. Johnston-Halperin, and M. E. Flatté, Materials for Quantum Technology **1**, 011001 (2020). DOI: 10.1088/2633-4356/ab9a55

[5] M. Fukami *et al.*, Proceedings of the National Academy of Sciences **121**, e2313754120 (2024). DOI: doi:10.1073/pnas.2313754120

Atom-defect Hybrid Quantum Systems
DE-SC0019241

Strongly interacting spin systems in diamond for quantum sensing

__Ania Bleszynski Jayich__[1] **(PI), Kunal Mukherjee**[2] **(co-PI), David Weld**[1] **(co-PI), Norman Yao**[3] **(co-PI),** Lillian Hughes[1], Simon Meynell[1], Weijie Wu[3], **Shreyas Parthasarathy**[1]**, Eveline Postelnicu**[2], Emily Davis[4]

[1]**Department of Physics, University of California Santa Barbara,** [2]**Department of Materials Science and Engineering, Stanford University,** [3]**Department of Physics, Harvard University,** [4]**Department of Physics, New York University**

Abstract: Solid-state quantum sensors are already impacting many arenas, including materials science and biology. With improvements in quantum control and materials engineering, they are poised to make significant leaps forward in their sensitivity, spatial resolution, and applicability in real-world applications in the coming years. Here we use the diamond nitrogen vacancy (NV), one of the most advanced solid-state quantum sensors to date, and demonstrate novel quantum control and materials engineering techniques in spin ensembles with strong dipolar interactions and controlled dimensionality, and we show how these systems can be uniquely leveraged for advances in quantum sensing and simulation.

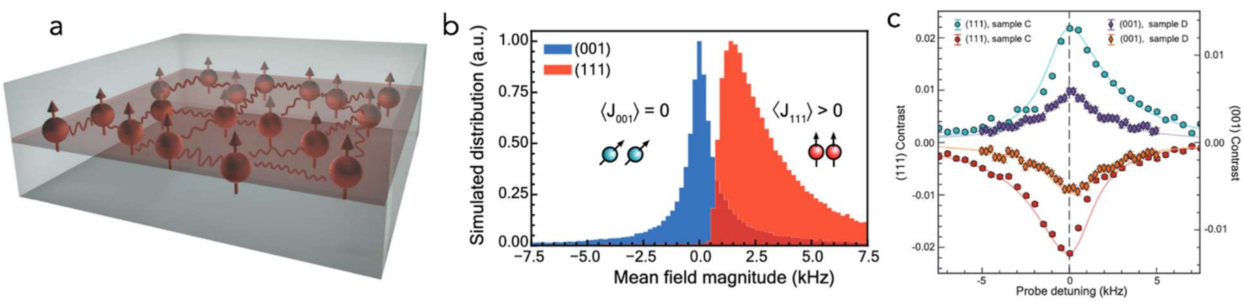

Figure 1: a) schematic of a strongly interacted 2D spin ensemble with spin quantization axis normal to the 2D plane. b) Simulated dipolar interaction-induced mean field distribution for (001) (blue) and (111) (red) NV ensembles, illustrating an asymmetric distribution for (111) due to the uniformly positive dipolar interaction energy. C) ODMR spectra for NV ensembles in (111)-oriented and (001)-oriented sample, where the asymmetric distribution of mean fields is reflected in the asymmetry of the 111-data (and not in the 100-data).

We first show the creation of strong dipolar interactions in a two-dimensional ensemble of nitrogen-vacancy (NV) centers generated via PECVD on (111)-oriented diamond substrates [1]. We find that diamond growth on the (111) plane yields high incorporation of spins, both nitrogen and NV centers, where the density of the latter is tunable via the miscut of the diamond substrate. Our process allows us to form dense, preferentially aligned, 2D NV ensembles with volume-normalized ac sensitivity down to η_{AC} =810 pT $\mu m^{3/2}$ $Hz^{-1/2}$. Importantly, we show that (111) affords maximally positive dipolar interactions among a 2D NV ensemble, which is crucial for leveraging dipolar-driven entanglement schemes and exploring new interacting spin physics. With these ensembles, we next demonstrate the ability to generate and characterize metrologically useful many-body entangled states (spin-squeezed states) [2].

Looking forward, we aim to fruitfully integrate these entanglement-enhanced sensors into close proximity with sensing targets. To that end, we discuss the challenges of bringing these sensors near a surface (such as increased decoherence) and our progress in attacking these challenges [3].

[1] **"A strongly interacting, two-dimensional, dipolar spin ensemble in (111)-oriented diamond",** L. B. Hughes, S. A. Meynell, W. Wu, S. Parthasarathy, L. Chen, Z. Zhang, Z. Wang, E. J. Davis, K. Mukherjee, N. Y. Yao, A. C. B. Jayich, *Physical Review X* **15**, 021035 (2025). https://doi.org/10.1103/PhysRevX.15.021035
[2] **"Spin squeezing in an ensemble of nitrogen-vacancy centers in diamond",** W. Wu**,** E. J. Davis, L. B. Hughes, B. Ye, Z. Wang, D. Kufel, T. Ono, S. A. Meynell, M. Block, C. Liu, H. Yang, A. C. B. Jayich, N. Y. Yao, *arXiv:2503.14585* (2025). https://doi.org/10.48550/arXiv.2503.14585
[3] **"Role of Oxygen in Laser Induced Contamination at Diamond-Vacuum Interfaces", S.** Parthasarathy, M. Joos, L. B. Hughes, S. A. Meynell, T. A. Morrison, J. D. Risner-Jamtgaard, D. M. Weld, K. Mukherjee, A. C. B. Jayich, *Phys. Rev. Appl.* **22**, 024067 (2024). https://doi.org/10.1103/PhysRevApplied.22.024067

Creating and Interfacing Designer Chemical Qubits
DE-SC0025176

Molecular color centers for quantum sensing

PI: **Danna Freedman** | Massachusetts Institute of Technology Co-PIs **James Rondinelli** | Northwestern University; **David Awschalom** | University of Chicago; **Mark Hersam** | Northwestern University; **Jeffrey Long** | University of California Berkeley; **Michael Wasielewski** | Northwestern University
Participants and collaborators: Mike Park, Alessandro Pereyra, Dane Johnson, Grant Smith, Dr. Noah Mendelson, Dr. Leah Weiss, Dr. Ryan Murphy, Dr. Bahman Golesorkhi, Priya Patel, José Méndez, Dr. Jens Niklas, Dr. Oleg Poluektov

Abstract:A bottom-up molecular approach enables the design and synthesis of tailored materials for quantum information science (QIS). As articulated in the Report of the Basic Energy Sciences Roundtable on *Opportunities for Basic Research for Next-Generation Quantum Systems*, creating and understanding new materials for QIS is crucial to realize quantum computing and quantum sensing.[3–5] The impact of an integrated chemical approach to the creation of designer systems was further reinforced in the recent National Academy of Sciences report on *Advancing Chemistry and Quantum Information Science*, which highlighted the essential role of chemistry in the creation of next generation quantum systems. Towards that end, our team is harnessing molecular our toolkit to *execute quantum sensing experiments to measure quantum materials*. Our approach uniquely situates molecular quantum sensors with atomic precision in van der Waals contact or covalent linkage with an analyte, thereby providing unparalleled sensitivity and enabling distance-dependent studies at unprecedentedly short length scales.[9]

Here, we will describe two recent accomplishments (1) the development of thin films for quantum material sensing and (2) the first molecular Er qubit. To integrate molecular qubits with 2D materials, **Hersam** deposited films onto monolayer graphene. Electron paramagnetic resonance (EPR) (**Freedman**) and magneto-photoluminescence (magneto-PL) (**Awschalom**) measurements confirm that Cr(*o*-tolyl)$_4$ remains microwave addressable and optically active in the thin film environment (**Figure 1**). **Hersam**, **Freedman**, and **Awschalom** have shown that the spin-state can be initialized and read-out optically for thin films grown on sapphire. Ongoing work is focused on characterizing the spin-optical interface for thin films grown on 2D materials. In parallel, **Hersam** has been expanding the thin-film library with additional combinations of molecular qubits and host matrices. For instance, films of Cr(o-tolyl)$_4$/Sn(4-fluoro-2-methylphenyl)$_4$ and Cr(2,3-dimethylphenyl)$_4$/Sn(2,3-dimethylphenyl)$_4$ have been grown on monolayer graphene. This growing list of thin film compositions is expanding the quantum sensing options for optically addressable molecular qubits.

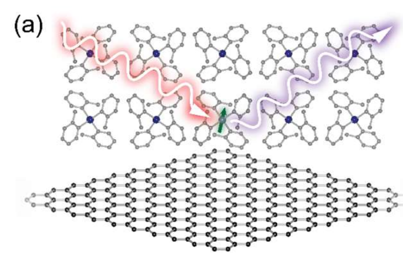

Figure 1 | (a) Schematic illustration of a CrSn(*o*-tolyl)$_4$ molecular qubit thin film on monolayer graphene.

In a second area **Awschalom** and **Long** have demonstrated a proof-of-principle optically addressable molecular qubit, $Cs[Er(hfac)_4]$ (hfac = hexafluoroacetylacetone), highlighting the benefits of Kramer's ion molecules as a complementary platform for quantum sensing. As highlighted in **Figure 2**, the qubit features the requisite level structure and optical cycling (**Figure 2**) that has now enabled demonstration of optical initialization and readout (**Figure 2c,d**) as well as coherent ground state qubit control.[i] We have further demonstrated optical spin-spectroscopy making use of the narrow homogeneous (~MHz) and inhomogeneous linewidths (~GHz), key features for the performance of this platform in future sensing applications at 1.53 μm (within the NIR-III transparency for biological materials). Note, this collaborative work leverages **Long**'s orthogonal BES-funded efforts to synthesize lanthanide-based molecular qubits, supported at Lawrence Berkeley National Laboratory under contract no. DE-AC02-05CH11231.

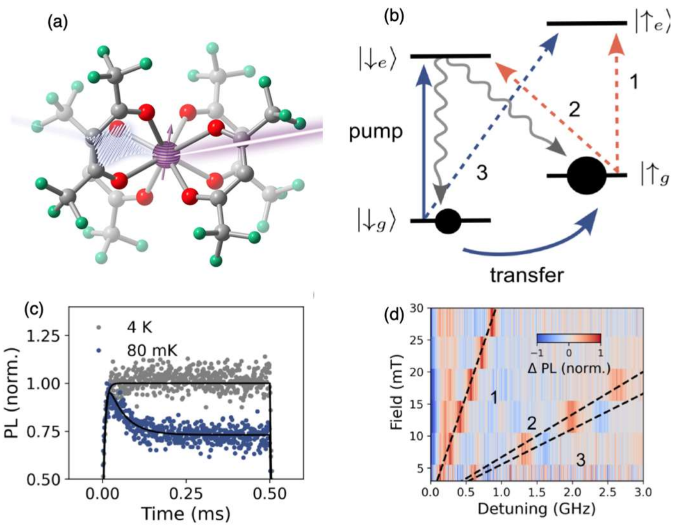

Figure 2 | Optically addressable lanthanide molecular qubits. (a) Depiction of $Cs[Er(hfac)_4]$. Red, grey, green, and purple correspond to oxygen, carbon, fluorine, and erbium, respectively. Cs and H atoms omitted for clarity. (b) Level diagram and spin-dependent optical transitions of $Cs[Er(hfac)_4]$. (c) Temperature-dependent depletion of population due to optical spin pumping at millikelvin temperatures. (d) Differential photoluminescence as a function of applied magnetic field and optical detuning (via electro-optic modulation) which features enhanced photoluminescence along transitions 1,2 as labeled in (b) and diminished photoluminescence at zero detuning and 3 as labeled in (b) consistent with optical spin pumping into the electronic spin sublevel.

Phonon Control for Next-Generation Superconducting Systems and Sensors

KCAS23

Alp Sipahigil, Haoxin Zhou, Kangdi Yu, Zi-Huai Zhang, Yashwanth Balaji, Mythili Surendran, **Shaul Aloni, Sinéad Griffin**, **Adam Schwartzberg**

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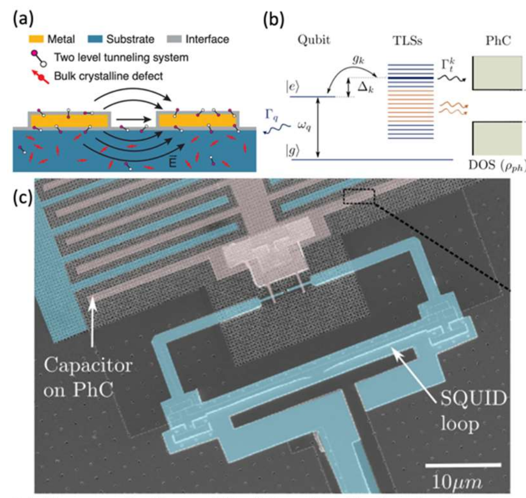
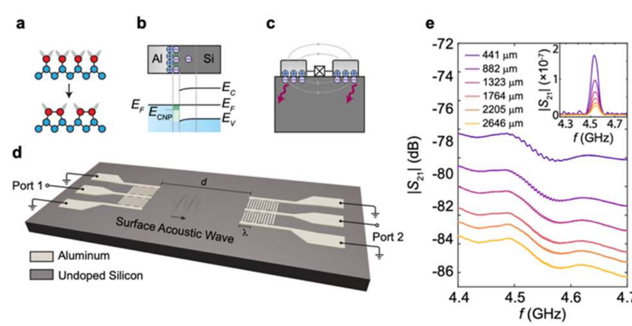
Abstract: Our project aims to advance superconducting qubits and quantum sensors by understanding and controlling their interactions with phonons. Piezoelectricity results in decoherence in superconducting qubits by converting electrical energy of the qubit into phonons in the substrate. Traditional qubit substrates, such as sapphire and silicon, are centrosymmetric crystals that lack piezoelectricity. However, in real-world quantum devices, symmetry is broken by defects and interfaces that can lead to a weak piezoelectric response and qubit decoherence by phonon radiation. In this presentation, I will discuss three recent results by our team on (i) identifying microscopic origins of superconducting qubit decoherence due to defect-induced piezoelectricity in silicon [1], (ii) suppressing phonon radiation in superconducting qubits via artificial materials with phononic bandgaps [2], and (iii) the first observation of interface piezoelectricity at the aluminum-silicon interface as a decoherence mechanism for superconducting qubits [3].



(a) Superconducting qubits interact with two-level system defects at interfaces and bulk crystalline substrates [1]. (b) Qubit-TLS interactions lead to decoherence by phonon radiation, which can be suppressed using a phononic bandgap crystal (PhC). (c) Image of a superconducting qubit embedded in an artificial, phononic bandgap material. The qubit showed slowed relaxation to the environment [2].

I will begin by presenting the first microscopic identification of two-level system defects (TLSs) in crystalline substrates (Figure 1, [1]). In this work, we used the electronic structure of acceptor defects in silicon to predict their behavior as TLSs for superconducting qubits. Our findings show that even ultralow concentrations of boron in silicon at 10^{12}/cm^{-3} are sufficient to limit superconducting qubit performance, pointing to the need for higher-purity silicon substrates to enable next-generation superconducting qubits. Next, I will discuss a complementary approach that suppresses TLS-induced phonon radiation from superconducting qubits by utilizing artificial phononic bandgap materials. I will discuss the emergence of non-Markovian qubit dynamics inside the phononic bandgap [2] and discuss ongoing work to extend qubit lifetimes using this approach. Finally, I will present our discovery of interface piezoelectricity occurring at aluminum-silicon junctions at cryogenic temperatures (Figure 2, [3]). Breaking of centrosymmetry at material interfaces leads to a dipole density and an effective piezoelectric response. While interface piezoelectricity has been predicted as a loss mechanism for superconducting qubits two-decades ago, it has not been experimentally observed. I will present our observation of this effect using



(a,b) Surface reconstruction or work function mismatch can lead to a (c) polar interface with piezoelectric response. (d) Device geometry to probe interface piezoelectricity using surface acoustic waves (SAWs). (e) Observation interface piezoelectricity using microwave transmission measurements surface acoustic wave transducers from room temperature down to 10 mK. I will discuss temperature, material processing, and bias dependence of this effect and discuss possible origins. I will conclude by discussing the consequences of interface piezoelectricity on superconducting qubits, and present ongoing work where we are directly probing this effect with a superconducting qubit.

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Scalable quantum photonic materials based on precision doped diamond nanostructures
Award Number: DE-SC0025295

High quality quantum photonic interface for tin-vacancy color centers in thin-film diamond

Jelena Vuckovic[1] (PI), Hope Lee[1], Abigail Stein[1], Hannah Kleidermacher[1], **Michael Titze**[2] (co-PI), Shei Su[2], Luca Basso[2], Ania Jayich [3] (collaborator), Lillian Hughes[3], Hyunseok Oh[3], Kyuhyun Kim[3]

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Abstract: The tin-vacancy (SnV) in diamond has been identified as one of the most promising color centers for implementation of quantum networks, facilitating their scaling beyond the present record of 3 quantum nodes. As opposed to the silicon vacancy (SiV) in diamond, the SnV provides the same properties (long coherence, robustness to interfaces), but in naturally strained diamond and at elevated temperatures of 1.7K (as opposed to mK). Our team demonstrated single shot readout for the SnV-'s electron spin with record fidelity of 87.4% [1], but this must be improved for use in quantum technologies. The fidelity is limited by low photon collection efficiency from diamond (total efficiency of ~0.1% in the mesa type structure employed in this work). For this reason, in this project we have been focusing on development of high quality quantum photonic interface for SnV in diamond, and specifically in diamond thin films which are more suitable for high quality cavity fabrication.

Diamond thin films were fabricated in collaboration with Prof. Ania Jayich's group at UC Santa Barbara via smart cut methods. We then characterized the optical properties of SnV- centers implanted in these samples via photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy. From the PL confocal scans, bright color centers, with a density suitable for photonic crystal fabrication, were identified. From PLE spectroscopy, we probed the temporal stability of implanted color centers and determined an average linewidth of ~500 MHz for emitters.

We subsequently fabricated arrays of high quality photonic crystal nanobeam cavities together with microwave delivery electrodes and wire bonding pads surrounding the thin film diamond membrane (see Figure 1).

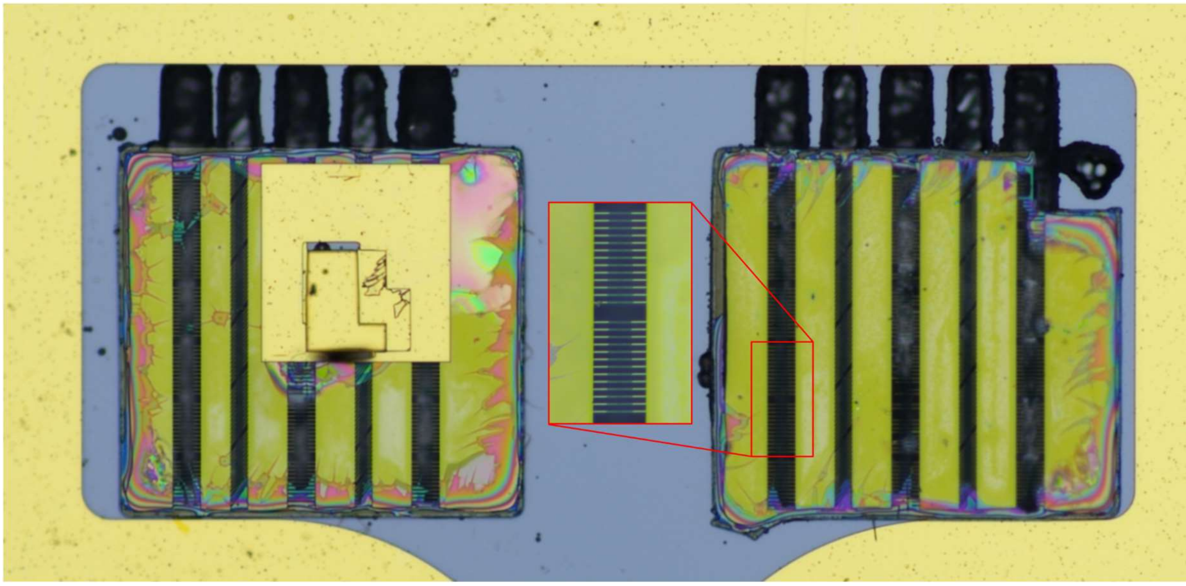

Figure 1. *Optical microscope image of completed photonic crystal devices in diamond thin films. The films are surrounded by Au microwave delivery electrodes. A small extra Au patch is used to anchor the left side film that has a hole from the bonding process. The inset shows details of the photonic crystal nanobeams, with the darker region in the center indicating the presence of periodic etched holes*.

We characterized these cavities via cross-polarized reflectance. For this configuration, the sample is mounted with cavity mode aligned to the horizontal polarization (H). A vertically polarized (V) broadband light source serves as the input, and the reflected spectrum is collected in H; the two paths are mixed with a polarizing beamsplitter. A half-wave plate oriented at ~22 degrees allows for partial excitation and collection of the cavity mode. From fitting spectrum peaks, we observe quality factors up to ~6000 at wavelengths as low as 612 nm, limited by spectrometer resolution. These resonances can then be red shifted via controlled Ar gas condensation to interact with coupled SnV- color centers, with preliminary demonstrations showing a factor of 12 PL enhancement when the cavity is on resonance with the SnV- zero phonon line.

We are currently working on measuring spontaneous emission rate enhancement (Purcell enhancement) of individual SnV's in these cavities, and will then perform spin control and single shot readout on SnV's embedded in cavities. We expect that single shot readout fidelity of an electron spin should be significantly approved, approaching 100%.

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2. org.stanford.idm.oclc.org/10.1103/PhysRevX.14.041008

Creating and Controlling Atomic Scale Quantum Systems

Award Number: ERKCK47
Abstract Title: Atom-by-Atom Fabrication for Quantum Information Sciences

Names of PI, coPI(s): Stephen Jesse, Ondrej Dyck, Chris Nelson, Mina Yoon, Andy Lupini

Affiliation: Oak Ridge National Laboratory

Abstract: Atomic-scale fabrication is pivotal for advancing quantum information sciences (QIS),

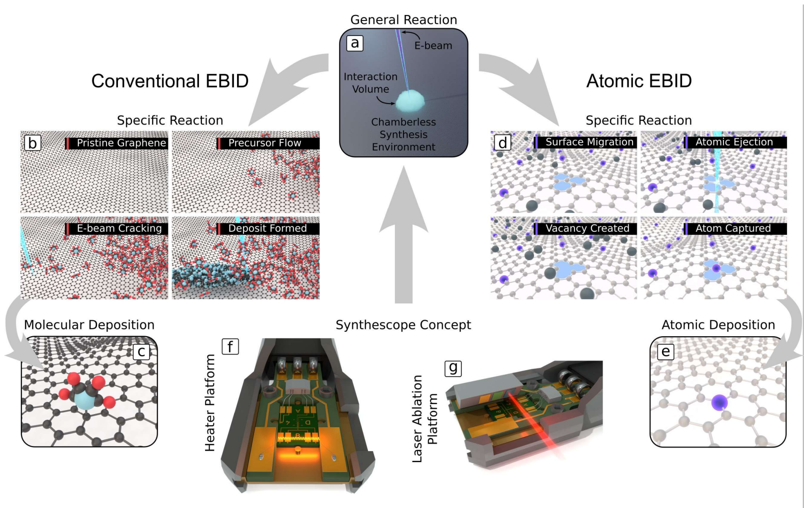

Figure 1: Synthescope concept. (a) Depiction of a general reaction. (b) Illustration of the conventional EBID process. (c) Fundamental limit of conventional EBID. (d) Illustration of an atomic EBIC process. (e) Fundamental limit of atomic EBID. (f) Schematic of heater deposition platform. (g) Schematic of laser deposition platform.

enabling the creation of tailored quantum devices such as qubits and emitters. We have developed scanning transmission electron microscopy (STEM)-based techniques to achieve atomic manipulation, directly addressing QIS needs. This abstract outlines technical advancements in electron beam-induced fabrication, focusing on “the synthescope”, and applications to quantum technologies. Our approach builds on the deterministic placement of single atoms, initially demonstrated with silicon impurities in graphene. By creating chemically reactive defect sites and leveraging electron beams to position atoms, we achieved precise doping with elements like tin, chromium, and platinum. A key innovation is the synthescope, which transforms the STEM into a synthesis platform—the beam-sample interaction

volume conceived of as a chamberless synthesis environment (CSE) (Fig. 1a). Unlike traditional electron beam-induced deposition (EBID) (Fig. 1b-c), which is limited to molecular deposition, atomic EBID creates vacancies that capture single atoms, achieving the ultimate in precision at the single atom atom level (Fig. 1d-e).[1] The abstraction of these two approaches is one where a general reaction is sought within the CSE without specifying what that reaction should be. To discover and control these reactions

we must furnish the STEM with additional capabilities aimed at environmental control. The first target is the supply of atoms. The platforms illustrated in Fig. 1f-g illustrate a heater and laser strategy. By heating the substrate to stimulate thermal migration, we enabled long-range patterning of atomic clusters, as demonstrated with copper on twisted bilayer graphene.[2] The filament-based heater (Fig. 2a-c) builds on this idea to evaporate source materials like tin in situ, growing nanoflakes on graphene (Fig. 2d).[3] An alternative laser ablation approach will further expand versatility and control (Fig. 2e-f).[4]

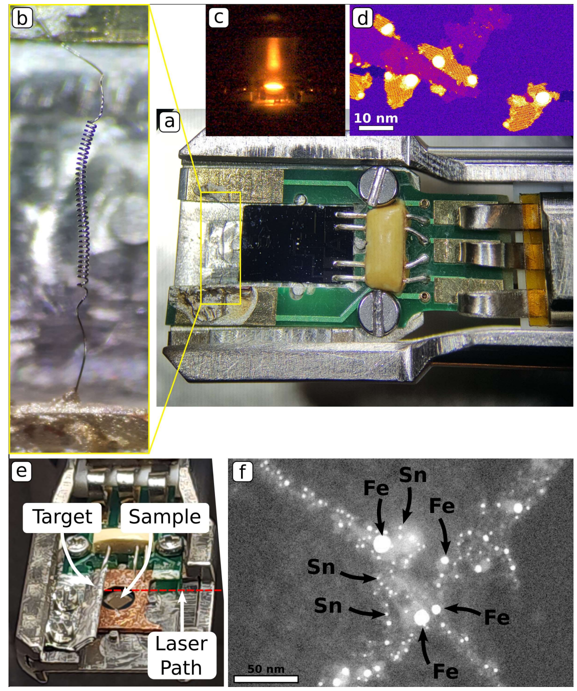

Figure 2. (a) Heater deposition platform. (b) Magnified view of W heater filament. (c) Heater operating in microscope stage. (d) Sn nanoflakes grown in situ. (e) Laser deposition platform. (f) Example Sn nanoparticles deposited in situ.

To enhance device fabrication, we are developing a quantum lab-on-a-chip in collaboration with MIT and KAUST, integrating e.g. electrical biasing, gating, magnetic fields, amplifiers, heaters, and photon detectors into a MEMS chip at the synthescope's core (black chip in Fig.2a). These added capabilities will further enhance control over the sample environment and provide access to new information channels for experiments (transport, photon detection, electron density measurements). Our atomic fabrication techniques offer a promising approach to QIS. By enabling precise control over atomic structures, we pave the way for prototype quantum devices and the implementation of experiments thought to be impossible to perform due to fabrication limitations.

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Modeling, probing, and controlling quantum coherence in materials
DE-SC0020313, FWP: SCW1797 (LLNL)

Scalable parallel quantum sensing with individual nitrogen-vacancy centers in diamond

Victor Brar[1], Matthew Cambria[2], Saroj Chand[2], **Jennifer Choy**[1], Ádam Gali[3], **Shimon Kolkowitz**[2], **Alex Levchenko**[1], **Vincenzo Lordi**[4], Jeronimo Maze[5], **Robert McDermott**[1], **Keith Ray**[4], Caitlin Reiter [2]

[1]University of Wisconsin, Madison, WI, USA. [2]University of California, Berkeley, CA, USA. [3]Wigner Research Centre for Physics, Budapest, Hungary. [4] Lawrence Livermore National Laboratory, Livermore, CA, USA. [5]Pontificia Universidad Católica de Chile, Santiago, Chile.

Abstract: In this collaborative QIS project our central aim is to apply a detailed understanding of decoherence to develop next-generation quantum sensors and high-fidelity qubits that are amenable to scaling. Our team of theorists and experimentalists are working closely together to achieve the following aims: (i) Refine quantum probes and theoretical models to determine the microscopic origins of noise from surfaces, interfaces, and the bulk across a range of quantum platforms. (ii) Integrate the quantum probes and theoretical models we have developed to cross-validate experimental and theoretical results and conclusions, and to gain deeper insights into coherence in materials. (iii) Harness our newfound knowledge and understanding to inform system-level design of quantum devices in order to mitigate and eliminate materials-induced decoherence. The improvements in materials properties and quantum device design enabled by our efforts will open a wider range of parameters and geometries for viable qubits and quantum sensors, improving the prospects for scaling solid-state qubits toward fully error-corrected fault-tolerant arrays, and enhancing the spatial resolution and sensitivity of quantum sensors.

One focus of our efforts has been on understanding the microscopic origins of decoherence in nitrogen vacancy (NV) centers in diamond. Most applications of NV centers take advantage of their long electronic spin coherence times. Spin-phonon relaxation places an ultimate limit on the achievable coherence times, but surprisingly the limits as a function of temperature had not previously been fully characterized. In addition, the microscopic origins of spin-phonon coupling were not fully understood. We characterized the phonon-limited three-level relaxation rates in multiple samples with differing defect concentrations over a broad range of experimentally relevant temperatures. We developed a self-consistent microscopic model that fully explains the observed relaxation rates, and that clarifies and corrects prior longstanding misunderstandings in the field. These results were published in Physical Review Letters [1]. In the process, we also discovered a physically motivated analytical expression for the temperature dependence of the zero-field splitting of the NV center electronic spin, which is widely used for nanoscale thermometry [2].

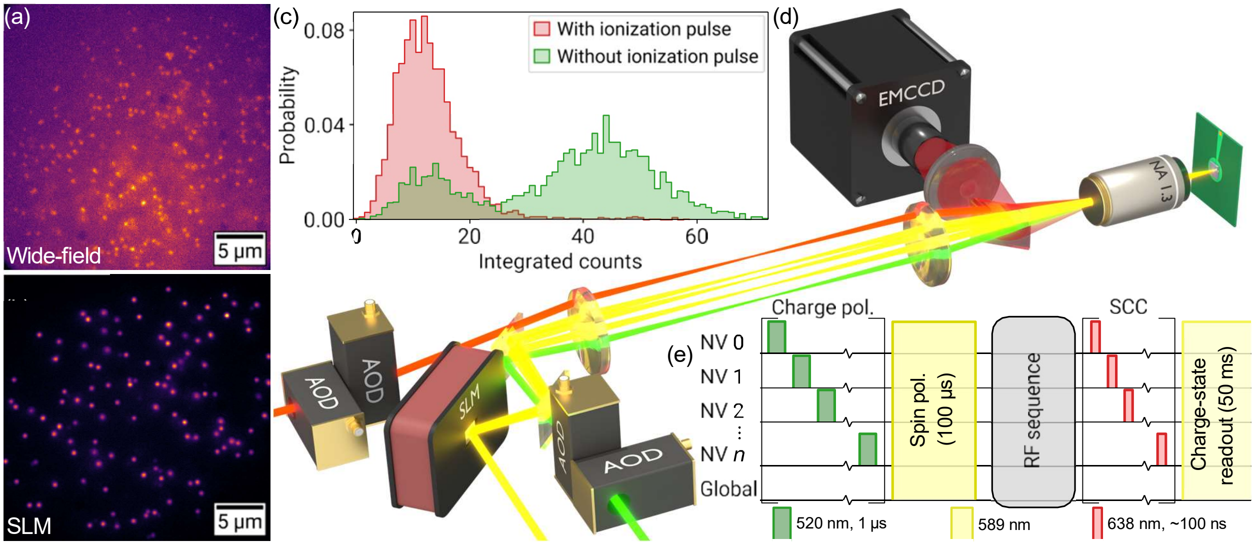

Figure 1: (a) Image of NV centers in diamond under wide-field 520-nm illumination. (b) Same region under 589-nm illumination patterned with a spatial light modulator (SLM). 108 charge-stable single NV centers are targeted, exhibiting consistent ODMR signals. (c) NV center charge-state manipulation and readout. (d) Experimental apparatus for multi-NV center quantum sensing. (e) Experimental sequence for parallel spin measurements. Figure adapted from Ref. [3]

Another focus of our efforts has been on leveraging our improved understanding of the physics of the NV center to scale up nanoscale quantum sensing. Typically, quantum sensing experiments using NV centers are performed either with a single isolated NV center or with an unresolved ensemble of many NV centers, resulting in a trade-off between measurement speed and spatial resolution or control over individual defects. We developed a new experimental platform with the capability to coherently manipulate and measure over 100 NV centers in parallel. These results have been accepted for publication in *Physical Review X* [3]. We are currently in the process of integrating our novel multiplexed quantum sensing technique with a cryostat in order to study the origins of flux noise at superconductor interfaces.

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Novel Probes of Topological Superconductivity for Next-Generation Quantum Systems

DE-SC0020305

Leon Balents (co-PI), Stephen Wilson (co-PI), Ganesh Pokharel, Stephen J. Gomez Alvarado, **Eric Cui, Taka Park, Arman Rashidi, Jiashu Wang, Andrea Young (co-PI),** **Susanne Stemmer (PI)**

University of California, Santa Barbara

Abstract: Topological superconductors host quasiparticles that are of great interest for generating quantum entangled states that are inherently and robustly protected against decoherence. Although several promising candidates have been identified, experimental techniques that unequivocally diagnose topological superconductivity are currently lacking. In this collaborative project between three experimental groups (Wilson, Young, Stemmer) and one theory group (Balents), we utilize topological superconductor candidates developed by our team to advance, test, and refine new techniques for probing the physics of topological superconductors. We aim at advancing local detection of signatures of their unique quasiparticles via their influence on observables like vortices, currents, or temperature, as well as more global measurements, such as x-ray scattering and transport, including thermoelectric and thermal signatures. To this end, the project is currently focusing on two main materials platforms: (i) kagome superconductors and (ii) synthetic structures that interface a superconductor with a topological material.

The kagome family offers a tunable platform for identifying signatures of topological superconductivity through phenomena, such as composite pairing, and through the interplay with charge correlations, such as charge density waves (CDWs), short-range charge correlations [1], and nematicity [2]. For example, theoretical studies of electronic instabilities in kagome metals in this project found superconductors, charge, orbital moment, and spin density waves when the Fermi energy was close to saddle points [3]. Experimentally, it was found that the addition of holes lifts the Van Hove points above the Fermi level and rapidly suppresses the CDW instability, resulting in "double-dome" superconductivity (Fig. 1). Recent experiments focused on gaining a better understanding of the interplay between charge correlations and superconductivity. To this end, the Wilson and Young groups imaged vortex lattices via nano-SQUID on-a-tip (nSOT) measurements and determined charge correlations via synchrotron x-ray diffraction. An example is shown in Fig. 1. The nSOT measurements show the vortex lattice in the kagome superconductor CsV_3Sb_5. This vortex lattice persists across the entirety of the electronic phase diagram—across both superconducting domes, signifying 2e pairing both in the presence and absence of CDW order. Complementary x-ray measurements showed that the charge correlations abruptly vanish in the second superconducting dome. This is distinct from prior measurements of hole-doped CsV_3Sb_5. We are currently preparing a manuscript reporting on these studies.

In parallel with the experimental work, we are developing theoretical models of the mechanisms that can lead to unconventional superconductivity in these materials. In the limit of a perfectly flat band, the behavior is dictated by quantum geometry. In this project, we aim at understanding how quantum geometry influences the selection of emergent orders, such as intrinsic topological superconductivity. To this end, the Balents group has developed a formalism to determine the maximum susceptibility within flat bands, for both charge/spin density and superconducting orders. For repulsive interactions we found dominant antiferromagnetic order, while for attractive interactions we predicted unconventional superconducting states. We performed numerically exact determinantal Monte Carlo simulations, which confirmed the theoretical predictions [4].

The second major platform in this project are synthetic topological superconducting candidates, consisting of hybrid interfaces, either within a single materials platform [5] or by combining superconductors with topological or Chern insulators. To this end, we have focused on developing junctions that are compatible

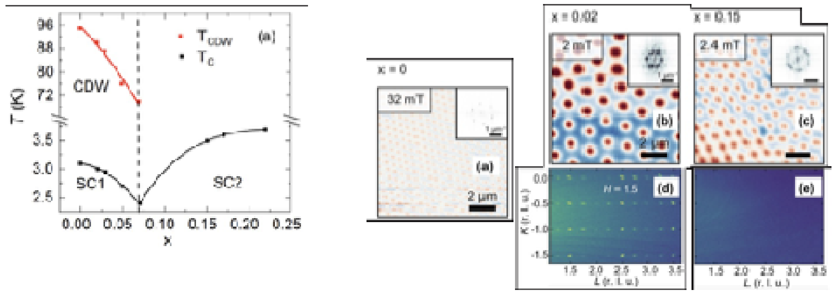

Fig. 1: (Left) CDW order and superconductivity in $CsV_{3-x}Ti_xSb_5$ as a function of hole-doping x. (Right) Evolution of the vortex lattice and charge correlations in $CsV_{3-x}Ti_xSb_5$: (a-c) vortex lattice (d) CDW order (e) Vanishing charge correlations in the second SC dome.

with nSOT imaging and that allow for new types of transport probes, including thermoelectric measurements. We found phenomena, such as diode effects, in the hybrid junctions. A model that incorporates the self-field induced by the superconducting current in the junctions describes some of the observations. A manuscript has been submitted. We have also fabricated superconductor/quantum Hall hybrid junctions and are currently analyzing the data.

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Imaging Emergent Phenomena in Two-Dimensional Magnets Using Single-Spin Quantum Microscope

DE-SC0024177

Brian Zhou[1], **Yu-Xuan Wang**[1], Thomas Graham[1], Xin-Yue Zhang[1], Zhong Lin[2], Claire Besson[2], Md Islam[2], Rafael Gontijo[2], Ganesh Tiwari[2], Tibendra Adhikari[2], Elton Santos[3], Ricardo Rama-Eiroa[3], Mohammad Badarneh[3], Kenji Watanabe[4], Takashi Taniguchi[4]

1) Boston College, Chestnut Hill, MA
2) Binghamton University, Binghamton, NY
3) University of Edinburgh, Edinburgh, United Kingdom
4) National Institute for Materials Science, Namiki, Tsukuba, Japan

Abstract:

This project aims to leverage and enhance the exceptional capabilities of quantum sensors based on the single electron spin of the nitrogen-vacancy (NV) center in diamond to deepen our understanding and drive the device integration of two-dimensional magnetic materials. A key focus is to transcend basic static characterizations by investigating dynamical phenomena and novel control mechanisms that may emerge under optical, electrical or magnetic perturbations, as well as via interfaces between heterogeneous systems. Our efforts, which rely on precise quantum control to isolate weak target signals from the noise, have the potential to unlock next-generation magnetic memories that overcome the energy efficiency bottleneck in current high-performance quantum-classical computing systems.

As a recent breakthrough, we have visualized for the first time the antiphase states in even-layer regions of an A-type antiferromagnet (AFM) with scanning NV magnetometry. Previously, it was believed that even-layer A-type AFMs have perfectly compensated magnetizations; however, by improving the stray field sensitivity of the quantum sensor, we were able to resolve a minute, uncompensated surface magnetization. This surface magnetization allowed us to visualize the nominally degenerate antiphase states (e.g., ↑↓ vs ↓↑ in bilayer; see Fig. 1a) and crucially the domain walls between them [1]. Moreover, by directly configuring the antiphase states of even-layer regions surrounding an odd-layer region, we revealed a strong *lateral* exchange bias (Fig. 1b) and elucidated its microscopic domain wall origin [1]. This lateral exchange bias, stemming from the intrinsic intralayer exchange interaction within a pristine single material, offers superior robustness and predictability compared to conventional exchange biases realized in vertical ferromagnet-AFM bilayers, where the buried vertical interfaces are prone to disorder.

Our upcoming efforts will target the integration of these 2D AFMs into deployable devices, such as in magnetic tunnel junctions, where our novel exchange bias can perform an essential role in stabilizing the magnetization of the reference layer. Moreover, we will pursue customized dynamical sensing schemes with scanning NV magnetometry, requiring more advanced experimental synchronization, to broaden the range of dynamics, signals and samples that can be studied.

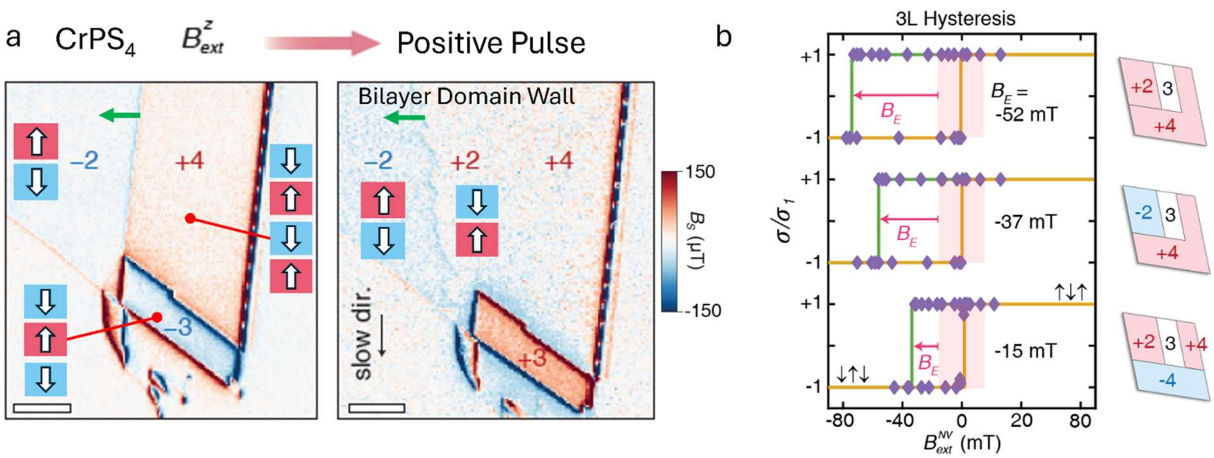

Figure 1 Antiferromagnet domains and lateral exchange bias in atomically thin $CrPS_4$ visualized by single spin quantum sensing.

[1] Y.-X. Wang, T. K. M. Graham, R. Rama-Eiroa, A. Islam, M. H. Badarneh, R. N. Gontijo, G. P. Tiwari, T. Adhikari, X. Zhang, K. Watanabe, C. Besson, E. J. G. Santos, Z. Lin, and B. B. Zhou, Configurable antiferromagnetic domains and lateral exchange bias in atomically thin $CrPS_4$, to appear in *Nature Materials* at https://doi.org/10.1038/s41563-025-02259-x (2025).

Probing Two-Dimensional Quantum Materials with Flying Electron Qubits
Award Number: DE-SC0025542

Dafei Jin

University of Notre Dame

Abstract:

We utilize our prior accomplishment in QIS – the long-coherence high-fidelity electron-on-solid-neon (eNe) qubit platform [1,2] – to develop a quantum-enabled SPM. The eNe-SPM is ideally suited to probe the electronic structures and phase transitions in two-dimensional (2D) quantum materials, i.e., exfoliated few-layer van der Waals (vdW) materials, at near-zero (~10mK) temperatures. (See **Fig. 1.**) With this technique, 2D quantum materials under investigation are transferred onto a superconducting quantum circuit and coated with a ~3nm thick noninvasive spacer – solid Ne. A single-electron qubit flying above the solid Ne at a ~1nm distance is transported, trapped, controlled, and read out by the quantum circuit. The spin states of the qubit (sensor) electron strongly interact with the spin or current states of the material (target) electrons and shed light on the local properties (e.g., spin texture and supercurrent) of the materials. Position scanning can be achieved by continuously sweeping the sensor electron above the materials with electric fields free of mechanical vibration or heat generation. The spatial resolution can reach ~1nm, which is limited only by the sharpness of the electron wavefunction and the precision of control electronics. The ultralong spin coherence time (~1s) [3] and charge coherence time (~0.1ms) [2] versus the ultrashort transport time (~10ns) and spin-to-charge conversion time (~5ns) in this system guarantee ultrahigh sensing fidelity. In the end, we intend to implement this new technique to study magnetic and topological materials, unconventional superconductors, quantum spin liquids, etc.

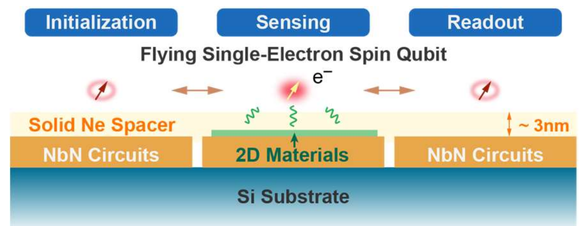

Fig. 1: Schematic of the electron-on-solid-neon (eNe) qubit enabled scanning probe microscopy (SPM).

Last year, we made several crucial breakthroughs for this project. **This award critically supported the growth and characterization of solid neon, as the key to all subsequent developments.**

- We measured the charge noise spectrum of eNe qubits and confirmed their order-of-magnitude quietness compared to semiconducting quantum-dot qubits. [4] (See **Fig. 2.**)
- We promoted the operation temperature of eNe qubits to above 100mK and observed their much slower performance downgrade compared to superconducting transmon qubits. [4]
- We achieved the first set of entangled two-qubit gates in the eNe system, which paves the way toward multi-qubit distributed quantum sensing in the longer term. [5]
- We built a new experimental setup to accurately measure the electron mobility on solid neon, which will calibrate the electron spin shuttling in the proposed scanning probe technique.

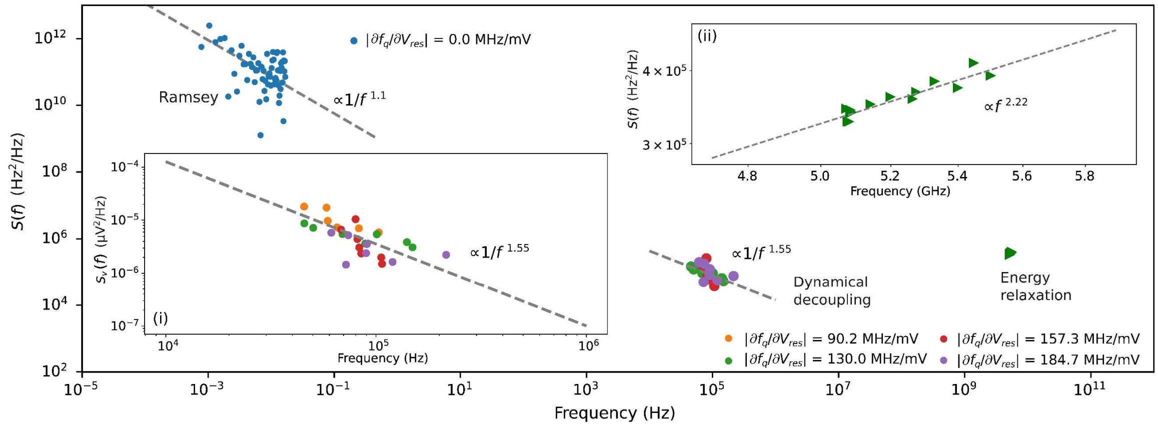

Fig. 2: Noise spectrum of an eNe qubit. Color dots in the main plot for 0.01–1MHz: total noise density derived from dynamical decoupling data at different qubit bias points. Blue dots in the main plot for 10^{-3}–10^{-1}Hz: calculated total noise density from long-term Ramsey measurements when biased at charge sweet-spot. Green triangles in the main plot near 5GHz: transverse noise of the eNe qubit. Inset (i): equivalent charge (voltage) noise on the resonator electrode. Inset (ii): zoom-in of transverse noise. Gray dashed lines: power-law fits of frequency-dependent noise. [4]

Next year, we have several tasks to complete. **This award is the primary support for all these efforts.**

- We will use a 4K cryogenic atomic force microscope (AFM) to *in situ* study the surface morphology of solid neon grown on a 2D material, specifically graphene and hexagonal boron nitride (hBN).
- We will attempt to trap a layer of 2D electrons atop a solid-Ne-coated 2D material and move the electrons around by surrounding electrodes.
- We will fabricate a prototype spin-charge conversion device containing a superconducting resonator and a micromagnet.

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Hybrid Magnon Systems
DE-SC0022060

Magnetoelastic Coupling to Magnons

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Abstract

The inherent time-reversal breaking of magnetic materials makes them ideally suited for non-reciprocal phenomena. Indeed. non-reciprocal microwave propagation, such as in isolators and circulators, are often engineered by careful mode matching with magnetic excitations. However, the direct conversion of microwave photons to magnons suffers often from inefficiencies that are hard to overcome. Therefore, our goal is to boost magnon non-reciprocity and magnon-photon coupling via magnetoelastic interactions by combining surface acoustic wave devices with magnetoelastic materials.

Towards the goal of utilizing magnetoelastic coupling, we have systematically studied the magnetostriction and temperature dependent magnetic damping in B-doped $Fe_{80}Ga_{20}$ thin films grown by magnetron sputtering [1]. We observed that the incorporation of boron induced a phase transition from poly-crystalline to amorphous, confirmed by X-ray diffraction and high-resolution transmission electron microscopy. We identified that 10% boron doping, which was in the mixed phase regime, was optimal for simultaneously realizing large magnetostriction of 50 ppm and low magnetic damping of 6×10^{-3}, see Fig. 1. Furthermore, we have conducted temperature dependent magnetic damping studies and observed a peak in

magnetic damping at around 40 K, which we attributed to magnetoelastic coupling. We found the magnetic damping at 5 K was lower than room temperature damping, indicating potentially further suppressed damping in quantum regimes at mK. Subsequently, $(Fe_{80}Ga_{20})_{90}B_{10}$ films were integrated into surface acoustic wave (SAW) devices prepared on both $LiNbO_3$ and AlN to study magnon phonon coupling, including acoustic wave driven ferromagnetic resonance.

Furthermore, we are working with bulk acoustic wave (BAW) based coherent magnon-phonon coupling in a single-crystal YIG substrate. We have successfully deposited high-quality piezoelectric AlN thin film on the YIG substrate, and fabricated top and bottom electrodes with a Pt/AlN/Pt vertical structure. The sample will allow us to excite perpendicular BAW across the YIG thickness, which will couple to perpendicular standing spin waves with matching frequency and wavelength to provide coherent magnon-phonon coupling.

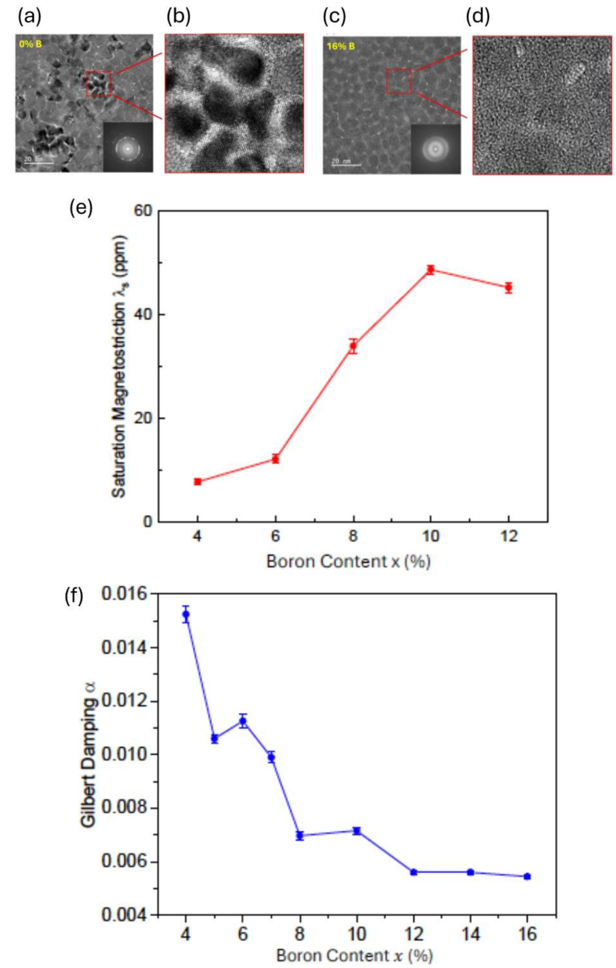

Fig. 1: Transmission electron microscopy of (a,b) $Fe_{80}Ga_{20}$ and (c,d) $(Fe_{80}Ga_{20})_{84}B_{16}$. Boron-doping dependence of (e) magnetostriction and (f) magnetic damping in $(Fe_{80}Ga_{20})_{1x}B_x$.

Building on these results we will use SAW devices to study magnon-phonon coupling as a function of $(Fe_{80}Ga_{20})_{1-x}B_x$ composition. Furthermore, we will use these devices to study the temperature dependence of magnetostriction, which will allow us to determine whether magnetoelastic SAW devices can be used for non-reciprocal coupling at cryogenic temperatures for quantum operations. In addition, we will compare to first-principles total energy simulations to gain detailed understanding of magnetostriction in these compounds and to distinguish energy changes induced by composition and strain. In parallel we will also explore magnon-phonon coupling in high quality YIG. We will explore both SAW coupling with YIG thin films and perpendicular acoustic wave coupled to standing spin wave in bulk YIG disks. Due to the strict substrate requirements for high-quality YIG growth, we plan to fabricate piezoelectric AlN layers on top of YIG for SAW propagation. We will also tune the lateral profile of the AlN layer and compare the coupling efficiency between AlN layer with uniform thickness and AlN layer with a thickness gradient right on top of YIG.

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Project Title: Multi-component Cavity Polaritons for Tunable Intermolecular Entanglement and Controlled Photon-to-Electron Quantum Transduction
Award: DE-SC0022134

Abstract Title: Molecular Interactions Towards Light-Induced Quantum Entanglement Generation

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Abstract: Given their ability to be modified structurally, molecules present unique opportunities as components in quantum information science and technology (QIST). By modifying the identities or positions of specific functional groups, synthetic chemists can change the energies of light molecules absorb, control their physical dispersion into different media, and modulate the lifetimes of excited states driven by photoexcitation. Despite this promise, it remains unclear how these amenable changes to their structure affect the ability of molecules to produce quantum information such as intermolecular quantum entanglement. Moreover, there remains few methods through which decoherence mechanisms that destroy quantum entanglement can be minimized to enhance the applicability of molecular systems in QIST. We have undertaken a combined theoretical and experimental approach to assess these types of issues.

Our theoretical efforts have been broadly made along three separate thrusts. I will highlight the accomplishments our group members have made establishing the mechanisms of quantum entanglement generation via the driving of model material systems in simple cavities by incident classical light fields. Our group has defined methods to compute a naïve estimate of the quantum concurrence, which estimates the maximum level of entanglement between two systems [1]. These calculations indicate that one can generate quantum correlations between an incident, classical light field and a material system embedded within an electromagnetic (EM) resonator. These calculations predict how the naïve concurrence will evolve in time as a function of different light-matter coupling conditions. These results indicate the ability to generate quantum correlations related to entanglement through the strong coupling of light and matter within an EM resonator.

The experimental efforts have been divided into two general thrusts. First, we have designed, fabricated, and characterized different types of hybrid states known as cavity polaritons through strong light-matter coupling within EM resonators. The efforts in this thrust were then distributed into separate areas based on the types of materials resonances used to couple strongly to cavity photons. In one area, group members utilized the electronic transitions of porphyrin-based molecules to form cavity polaritons in nano-scale EM resonators to assess how cavity polariton formation affects molecular dynamics central to maintain quantum entanglement. Precision spectroscopic studies on these systems enabled determination of the coupling conditions needed to suppress energetic disorder in molecular light absorbers due to vibrational dynamics. which represents a fundamental impediment to entanglement maintenance [2]. Additional studies leveraged a newly developed spectroscopic method, angle-resolved photoluminescence excitation spectroscopy, to determine the cavity polariton constituent that limits the hybrid light-matter state lifetime. These studies indicate that the coherence time between light and matter can be extended by over an order of magnitude when compared to the bare molecular state, which improves this performance metric for maintaining quantum entanglement in molecular systems [3].

In the second area of the first experimental thrust, group members have used vibrational resonances of molecular systems to form cavity polaritons in micro-scale, mid-infrared EM resonators. Using 2-dimensional infrared (2DIR) spectroscopy, these studies have enabled our group to assess how coherence can be controlled with resonator design. By creating trenches lithographically in Fabry-Perot micro-resonators, we demonstrate cavity polariton formation in 3-dimensionally confined spatial regions whose energies do not disperse with incidence angle conventionally [4]. Additionally, group members have

designed and applied a 2DIR hyperspectral imaging apparatus to assess how coherences induced by incident laser pulses propagate in these resonator systems spatially.

In the second experimental thrust area, group members have been assessing electron spins in porphyrin-based molecular materials towards light-mediated entanglement generation in these systems. Combined time-resolved electron paramagnetic resonance (EPR) and optical spectroscopic studies have shown that singlet and triplet molecular states do not delocalize their respective densities in correlated ways [5]. While no spin delocalization was found for photo-excited Zn-porphyrins, time-resolved EPR spectra at low temperatures suggest that adjacent electron spins in H-aggregates of paramagnetic Cu-porphyrin molecules can delocalize.

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Integrated Materials Platforms for Topological Quantum Computing Devices
DE-SC0019274

Abstract Title: Superconductor-semiconductor hybrid quantum devices

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Abstract: A significant challenge towards developing robust quantum computing systems with large numbers of logical qubits is rapid decoherence. A promising approach to addressing this challenge is based on the concept of fault-tolerant quantum computing architectures that leverage topological quantum states such as Majorana zero-modes (MZM). Substantial progress toward realizing and detecting topological states has been enabled by quantum devices which combine semiconductors with superconductors due to the combination of processing-friendly materials that offer high spin-orbit coupling, large g-factors, ballistic transport, and the ability to create transparent interfaces with various superconductors. However, further breakthroughs leading to topological quantum computation require next-generation quantum nanostructures that offer substantial improvement in the electronic purity together with optimal coupling across the superconductor-semiconductor interface. *Here we discuss our recent results under this project, focusing on superconductor-semiconductor quantum devices.* This work is a subset of our larger effort supported by this grant, which combines quantum device fabrication and characterization with materials synthesis and data interpretation informed by ab initio simulations and machine learning to develop robust, tunable and scalable platforms for the manipulation of quantum states.

(i) Few-mode quantum transport, Josephson junctions and π-shifted quartets using PbTe nanowires–toward topological states: We have used MBE-grown PbTe selective-area grown (SAG) nanowires to create quantum devices and study their normal and superconducting transport properties. In our recently-published work [1] we have seen very encouraging initial results in these nanowires, including well-defined conductance quantization plateaus (Fig. 1). This is superior to typical results reported to date in InSb or InAs nanowires, on which the community has previously focused. It could be indicative of low disorder effects in transport, which could have a major impact on developing MZM experimental platforms. We have also shown that these PbTe nanowires can be used to make superconducting devices, such as Josephson junctions, using Al contacts (Fig 1). A second major breakthrough from this work has been the observation of full multi-terminal superconducting correlations (π-shifted Cooper quartets) using nanowires with three branches, each connected to a superconductor. These results establish PbTe SAG nanowires as a promising

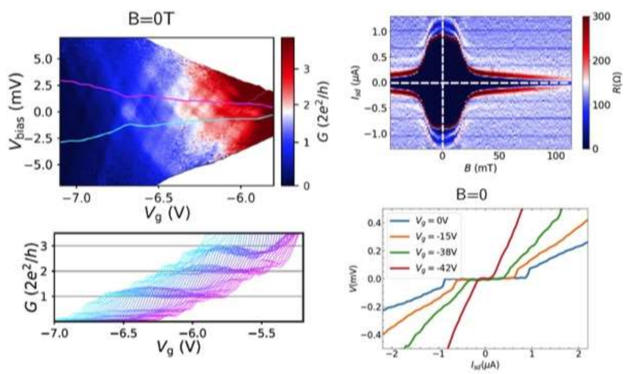

Fig. 1: Quantum transport results on PbTe SAG nanowire quantum devices. **(top-left)** Conductance in the few-quantum-modes regime, showing plateaus and phase-coherent transport. **(bottom-left)** Line cuts showing conductance plateaus. A slight modulation with bias is also seen, possibly attributable to a nearby quantum dot. **(top-right)** and **(bottom-right)** Transport in the superconducting regime (adapted from our Ref. [1]).

platform for pursuing MZMs and advancing the emerging field of multi-terminal superconducting devices. We are currently exploring ways to improve the superconductor-semiconductor interface and further reduce disorder. We are also exploring super-semi devices with PbSe, for which our preliminary first-principles calculations have identified promising interfaces with several superconductors.

(ii) Reproducible Quantum Devices: A significant goal for the project is to put materials discovery for quantum computing on firm footing. Here we are navigating a landscape where not all claims of advancement in topological effects have historically been backed by solid data. To this end, we have fabricated InAs/Sn and InSb/Al superconductor-semiconductor devices and explored tunneling zero-bias peaks previously reported as evidence of MZM (hysteretic supercurrents and subgap bound state spectra, and zero-bias peaks at zero external magnetic field) [2–4]. However, in this case the team has determined that they originate from Andreev bound states and are not of topological origin. The results also show the possibility to control superconductivity in a thin shell of Sn or Al superconductor, as well as induced superconductivity in an InAs or InSb nanowire using remote but nearby micromagnets.

(iii) Gate-tunable three-terminal superconducting diode: We have demonstrated a gate-tunable three-terminal superconducting diode that can rectify superconducting signals, with the polarity and efficiency of rectification being tunable by electrostatic gating, and operating in ambient magnetic field [5]. The multi-terminal nature of our diode device allows for integration and simultaneous rectification of multiple signals. The devices display a high degree of reproducibility. These results are very timely due to the rapidly-growing interest in superconducting diodes. Such devices could be relevant as a basis for cryogenic on-chip control circuits for superconducting qubits. Among of the most intriguing implications of this work are the potential to use such devices for topologically-stabilized superconducting circuits and for hybrid neuromorphic circuits for AI.

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Modeling, probing, and controlling quantum coherence in materials
DE-SC0020313, FWP: SCW1674

Engineering diamond surfaces for efficient NV emission

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Abstract: Spin defects in diamond, such as the negatively charged nitrogen-vacancy (NV) centers, can be highly sensitive to local (nanoscale) changes in magnetic field, temperature, and strain, which make them useful quantum sensors to measure spatially varying magnetic and electric fields in nearby materials, including superconductors and 2D materials. However, near-surface quantum defects within 10 nm from the surface tend to exhibit spin decoherence. Additionally, in a high-refractive-index environment such as diamond, most of the nonclassical light emitted is trapped within the bulk crystal due total internal reflection at the interface.

In this collaboration, we strive to better-understand and control interfaces in diamond, towards generating brighter and more-coherent near-surface emitters. One theme of this work is to use materials analysis tools, such as X-ray Photoelectron Spectroscopy (XPS), to provide molecular and atomistic insights on diamond surfaces. We conducted detailed investigations of diamond surfaces with various terminations and ligands to optimize the performance of near-surface NV centers. Using angle-resolved XPS, our work has revealed the presence of sub-nanometer-thick native sp2 carbon on oxidized diamond surfaces (Figure 1) [1], which has implications for the stability and coherence of near-surface NV centers used in quantum sensing. We also studied the effects of molecular contamination and sp^2 carbon on the oxidation of diamond surfaces [2], from which we gained insights into the layered heterogeneity of diamond surfaces, allowing us to optimize surface treatments. Our experimental efforts are performed alongside density functional theory calculations of surface energies and electron affinity for different types of carbon-oxygen bonding and surface coverage. By analyzing the electron affinity for partial coverage and mixed binding cases, we can model the complex surfaces present after realistic experimental conditions, as well as inform treatment methods for positive electron affinity surfaces with low charge noise.

In parallel, we address the issue of NV emission extraction by developing a framework for analyzing the room-temperature emission rates of near-surface NV centers of various orientations and diamond crystal cuts [3], which is helpful for understanding the depth-dependent radiative behavior of near-surface NV centers. This analysis is crucial for any sensing measurements relying on radiative lifetime determination (such as FRET) and for designing photonic devices that modify color center emission. We then designed and implemented crystalline silicon antennas to enhance light extraction from near-surface NV centers [4], demonstrating an order-of-magnitude improvement in single-photon emission in comparison to a bare diamond surface. Notably, the silicon antennas are formed by membrane transfer of crystalline silicon from a silicon-on-insulator wafer and avoids etching of the diamond surface to prevent degradation emitter's

quantum properties. In our collaboration, we will apply the aforementioned techniques to prepare bright and coherent NV centers to probe charge noise at interfaces of superconductors and dielectrics.

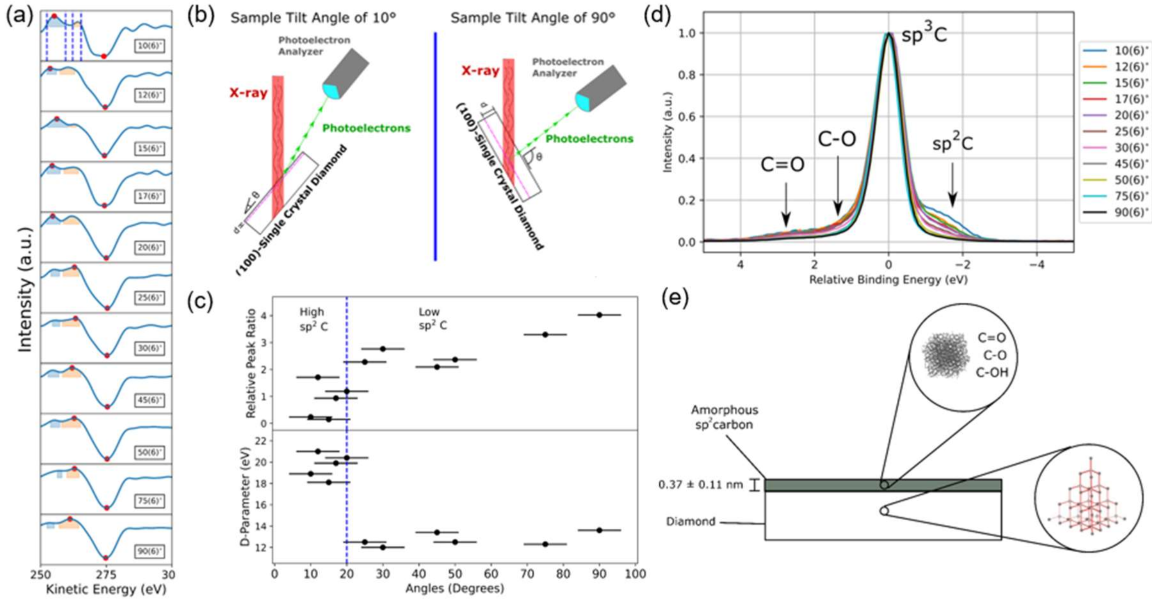

Figure 1: (a) Angle-resolved auger spectra for oxygen-terminated diamond. (b) Experimental configuration. (c) (Top) Relative peak ratio and (Bottom) Calculated D-parameter values plotted as a function of sample tilt angles, along with the regimes of high sp^2 C and low sp^2 C. (d) Angle-dependent C1s spectra. (e) Model of oxidized diamond surfaces, based on ARXPS data.

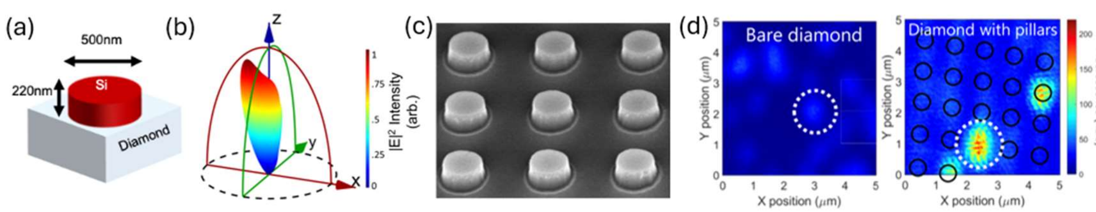

Figure 2: (a)-(c) Schematic, simulation, and fabrication of silicon nanoscale light extractor for near-surface NVs in diamond. (d) (Left) NV fluorescence from bare diamond and (Right) fluorescence enhanced by silicon antennas.

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Phonon Control for Next-Generation Superconducting Systems and Sensors
KCAS23

Material growth and fabrication of asymmetric tunnel junctions for phonon and quasiparticle transport suppression

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Abstract: One of the main goals of the PhononNext project is the suppression of quasiparticle interactions in superconducting quantum devices. In superconducting qubits, suppressing effects due to phonon interactions is critical for improving coherence times and reducing error rates, particularly during energetic impact events. In this project, we are developing materials and fabrication techniques to fabricate asymmetric Josephson junctions (JJs) composed of two superconducting materials with different superconducting gap energies. This approach with a large gap asymmetry will reduce quasiparticle tunneling across the junction which produces correlated errors and decreases qubit coherence time. A recent work has demonstrated the effectiveness of this technique, controlling the superconducting gap by changing JJ layer thicknesses using a single material.[1] However, the amount of gap engineering that can be achieved with a single material is limited. Here, we combine aluminum with the superconducting transition metal nitride niobium nitride (NbN), which has a comparatively large superconducting gap energy, to demonstrate a significantly asymmetric JJ and the potential available when other materials can be used.

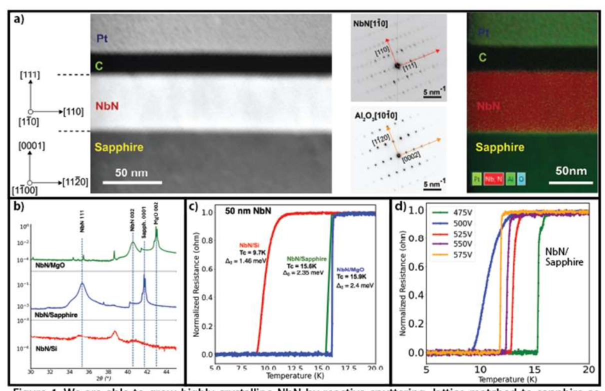

Figure 1. We are able to grow highly crystalline NbN by reactive sputtering, lattice matched to sapphire as shown in TEM cross section, electron diffraction and EDS (a). As the lattice match of the substrate decreases, so does the crystalline phase purity and the critical temperature/superconducting gap of the deposited material, as shown by x-ray diffraction (b) and low temperature transport (c). We also find that we can tune the superconducting gap by changing growth parameters, in this case substrate bias, enableing another route for superconducting gap control (d).

In the first part of the work we studied how NbN growth affects crystallinity and superconducting gap.[2] Films are grown with reactive sputtering which enables epitaxial growth on sapphire (fig 1a). The NbN film crystal morphology and transition temperature/superconducting gap are sensitive to the lattice of the substrate onto which it is grown, resulting in reduced order when growing on silicon, and increased order on MgO, which has a superior lattice match to NbN (fig 1b and 1c respectively). Because growth conditions are somewhat limited for fabrication processes, this is critical information to understand how the superconducting gap will be affected. It also enables some degree of superconducting gap tunability. However, we found that by carefully controlling growth parameters we were able to more precisely tune the superconducting gap, which will be useful for more precise gap engineering in our devices (fig 1d).

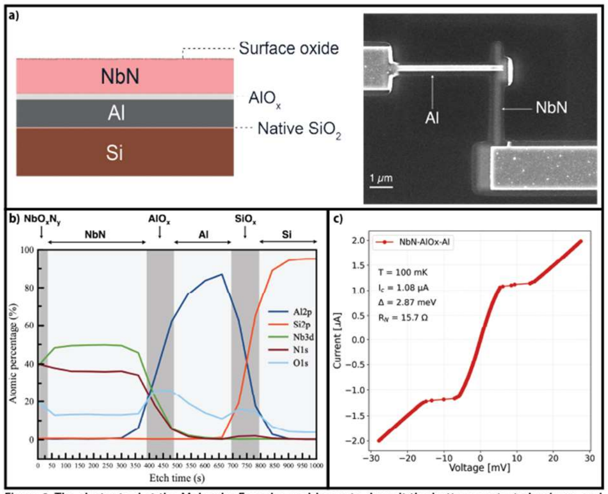

Figure 2. The cluster tool at the Molecular Foundry enables us to deposit the bottom contact aluminum and aluminum via conventional e-beam evaporation and oxidation, followed by reactive sputtering of NbN in a separate chamber without breaking vacuum (a). XPS depth profiling shows excellent elemental control over NbN, alumina, and aluminum (b). Ic measurements of these devices show non-ideal supercurrent behavior that is likely due to the NbN layer being too thin (c).

In the second part of our work we have focused on fabrication and testing of the asymmetric JJs.[3] Taking advantage of the high degree of control available through the quantum fabrication cluster tool at the Molecular Foundry, we were able to fabricate asymmetric Al/AlOx/NbN junctions. After the first evaporation and oxidation step, we transfer the wafer through vacuum to the reactive sputtering chamber and carry out a specialized, long distance sputtering process that enables better growth directionality (fig 2a). We tested the composition of each layer of this film to confirm that there was limited or no intermixing using XPS depth profiling (fig 2b). Finally, we carried out transport measurements at operational temperatures (100 mK) to observe the junction behavior. While we do find supercurrent, as expected, the slope is not as steep as it should be (fig 2c). Additionally, we find that the room temperature resistance of these asymmetric junctions to be two times higher than expected. We believe that both effects may be due to the NbN film being too thin, approximately 10 nm, which is currently set somewhat by fabrication limits. We are now developing improved fabrication methods to reach our targeted thickness of 30-50 nm, enabling us to determine the nature of the anomalous tunnel current shape and leading to improved device performance.

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Hybrid Magnon Systems
DE-SC0022060

Coherent Control of Magnons

Yi Li[1], Moojune Song[1,2], Tomas Polakovic[3], **Jinho Lim**[4,5], **Xingzhi Wang**[4,5], **Pratap Kumar Pal**[1,4,5], Thomas W. Cecil[6], John Pearson[1,7], Ralu Divan[7], Wai-Kwong Kwok[1], Ulrich Welp[1], Wei Zhang[8], Dmytro A. Bozhko[9], Kab-Jin Kim[2], **Jian-Min Zuo**[4,5], **Axel Hoffmann**[4,5,10], and **Valentine Novosad**[1]

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Abstract

Magnons, the quanta of collective spin excitations in magnetically ordered media, exhibit unique features for microwave quantum engineering, with examples of non-reciprocity in the form of propagating spin wave, highly desired for noise-isolated qubit operations with on-chip integration, and magneto-optical coupling for microwave-to-optic transduction, highly desired for implementing quantum transduction and building quantum interconnects. For either purpose, the capability of coherent magnon control in the time domain is crucial for designing coherent magnon gate operations and extend the dynamic control into the quantum regime.

Towards the objective of coherent control of propagating magnons, we demonstrate single-shot magnon pulse interference between two remotely coupled $Y_3Fe_5O_{12}$ (YIG) spheres embedded in a superconducting resonator [1] (see Fig. 1). Using a pair of vertical antennas to selectively couple to the two YIG spheres with minimal cross-talk to the in-plane superconducting resonator, we realize real-time constructive and destructive magnon interference by precisely controlling the frequency and time delay of two consecutive microwave pulses applied to one YIG sphere. In addition, we show a diffraction-grating-like interference pattern using up to four consecutive magnon pulses, where the interference peak becomes increasingly enhanced and sharpened with the number of pulses. Our results reveal that magnons can preserve full coherence, while being transferred between distant magnetic resonators. This is a

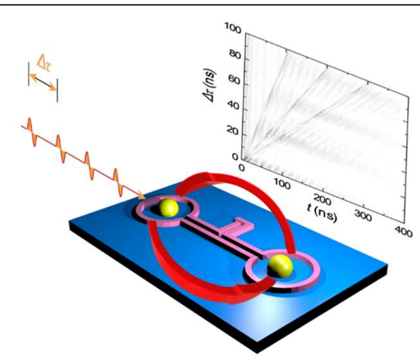

Fig. 1: Magnon interference: Real-time magnon interference is demonstrated between two remotely coupled yttrium iron garnet (YIG) spheres on a compact magnon-superconducting-resonator hybrid circuit [1].

critical requirement for implementing coherent magnon gate operations in real time. Moreover, the experiments were conducted on a chip-integrated cryogenic superconducting circuit platform along with high-fidelity interference performance. This work establishes a robust foundation for integrating magnonics with cQED and developing magnon-based quantum information processing and quantum sensing.

For future development of coherent control of propagating magnons, we are developing on-chip magnon-superconducting resonator hybrid system that couples to spatially nonuniform spin wave modes in a YIG thin film. We have fabricated quarter-wavelength NbN superconducting resonators directly on top of epitaxial YIG films grown on $Y_3Sc_2Ga_3O_{12}$ (YSGG) substrates. High Q-factor up to 100,000 is measured for NbN resonators fabricated directly on YSGG, and a high Q-factor of 3,000 is measured for NbN resonators fabricated on the YIG/YSGG film at 1.3 K. Strong coupling and mode anticrossing are observed between the superconducting resonator mode and the Damon-Eshbach (DE) spin wave mode of the YIG film. Our results introduce the first all-on-chip superconducting hybrid magnonic circuit base on low-damping YIG thin films, which will become the next-generation circuit platform for exploring propagating-magnon-based quantum information science. We will further use our recently developed single-shot electrical detection technique of propagating spin waves [2] to explore time-domain propagating magnon control.

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Optoelectronic Properties of Artificially Tailored Quantum Materials

DE-SC0024486

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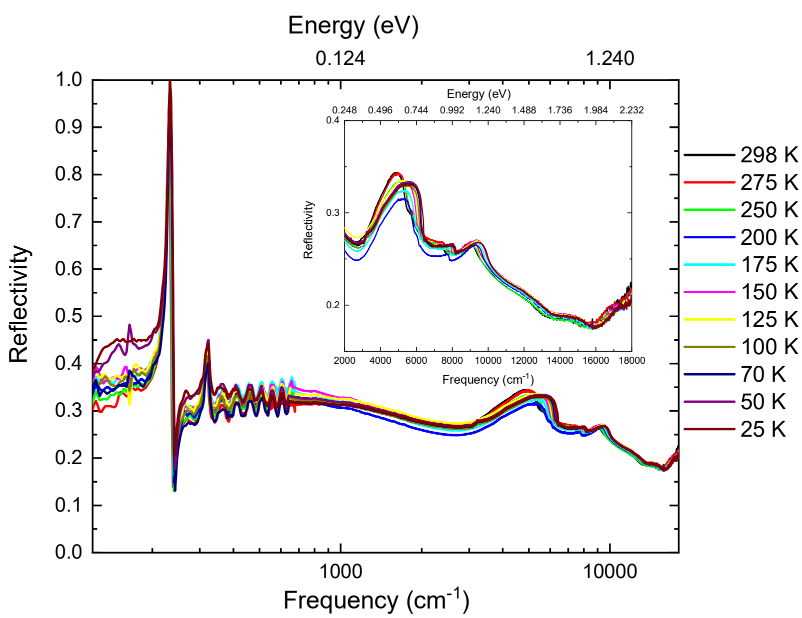
Abstract:

We present our recent results from broadband optical spectroscopic investigations on Moiré superlattices (MSL) based on 2D electron gas quantum wells. To create MSLs using IL, two separate, rotated AG patterns are created in the photoresist prior to development. By carefully controlling the angles between the four exposures defining the AG patterns, the twist angle between the two AG lattices can be defined without requiring post-processing alignment. The first set of parallel lines for AG1 is formed in the resist followed by a second set of parallel lines for AG2 with a twist angle of 5 degree. This is followed by the third set of lines at a twist angle of 60 degrees with respective to the first set. Finally, a fourth set of parallel lines is defined at a twist angle of 60 degree with respective to the second set. The laser intensity and expose time for each set of parallel lines are the same as in AG pattern. The first and third sets of lines create the first AG1 pattern. The second and forth sets of lines create the second AG2 pattern with a twist angle of 5 degrees. After post-exposure bake, photoresist development and RIE, holes are etched through the 2DEG, and MSLs are generated [1].

To study the electronic structure of the MSL devices over a broad energy range, we employed broadband optical spectroscopic techniques of Fourier transform infrared (FTIR) spectroscopy and ellipsometry. The infrared measurements were performed using the FTIR spectrometer Bruker IFS80v coupled to a Hyperion 2000 microscope. The FTIR microscope can cover a broad frequency range from 100 to 18000 cm^{-1} i.e., far-infrared (FIR) to visible frequencies. It employs three different light sources for the different ranges in the infrared frequencies. Depending on the frequency required for the measurement, different combinations of source, beamsplitter and the detector are chosen. The silicon-based bolometer used in the FIR frequency range is a cryogenic detector operated at liquid helium temperature, while mercury cadmium telluride (MCT) and InSb detectors used at mid-infrared (MIR) and near-infrared (NIR) frequencies, respectively, are operated at liquid nitrogen temperature. A silicon diode detector was used for measurements in the visible range. The variable temperature



environment was achieved using an optical cold finger cryostat coupled to the Hyperion microscope.

Broadband reflectivity measurements were performed for four samples: MSL 300/300/5, MSL 300/250/5, AG 300 and the substrate sample in the temperature range from 25 K to 300 K. Furthermore, higher frequency data from 18000 cm^{-1} to 47000 cm^{-1} was also obtained for the four studied samples using room temperature ellipsometry. The results obtained for MSL 300/300/5 are shown in Figure as an example. All four samples exhibit minimum change in the overall reflectivity with decreasing temperature. However, the phonons at FIR frequency range and the electronic transitions in the NIR frequency show temperature-dependent red shift (inset of Figure). These changes in the reflectivity spectra correspond to the evolution of band structure of this MSL device close to Fermi level [2]. Therefore, by comparing data from the MSL devices with the optical response of the substrate and single AG layer, we will be able to reveal the mechanism of the band structure evolution.

Our plan is to perform Kramers-Kronig transformation of the temperature dependent reflectivity of the samples to obtain optical conductivity. The optical conductivity being an additive optical constant will allow for deconvolution of contributions to the optical response from various components of the MSL device. In addition, we will continue to obtain more information using Raman scattering and photoluminescence on these samples to obtain comprehensive understanding of the quantum phenomena in these artificially tailored materials.

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Multimodal Quantum Material Control Monitored with Ultrafast Coherent X-Rays
Award Number DE-SC0019126

THz Control of Magnetization and Polarization Through Magnon and Phonon Coherences

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Abstract: The primary project goals are advances in our ability to control, monitor, and theoretically model novel quantum phases of matter. We use multiple dynamic stimuli to control multiple coupled modes including optical and acoustic phonons, magnons, and low-frequency electronic excitations. THz and long-wavelength IR excitation fields are used to drive optical phonons, magnons, and electronic responses; optical pulses to excite selected acoustic/shock waves that deliver specified in-plane or through-plane uniaxial strains; and crossed X-ray pulses to drive material responses with specified nanoscale spatial modulations that may reach length scales comparable to those that form spontaneously in many quantum phases. We probe the dynamic material responses with ultrashort coherent X-ray probe pulses at XFELs, and in THz through visible spectral regions using tabletop ultrafast laser systems. In some cases, we induce transformations of matter into new states with altered structural, electronic, and/or magnetic characteristics. X-ray diffraction and diffuse scattering, X-ray absorption, and resonant X-ray scattering measurements may be conducted to monitor the formation and duration of these states including their nanoscale textures.

Two recent experimental efforts will be reviewed. First, nonlinear THz excitation of magnon coherences in $YFeO_3$ and $ErFeO_3$ canted antiferromagnetic crystals will be illustrated. 2D THz EPR spectroscopy of the induced magnon responses has revealed nonlinear coupling of two orthogonally polarized magnon modes [1-3]. The spectra reveal sum and difference frequency generation; up-conversion through which excitation of one magnon mode generates a coherence of the other; and parametric amplification of the lower-frequency mode by the higher-frequency one. These nonlinear interactions could provide the fundamental underpinnings for nonlinear magnonic signal processing with ultrahigh bandwidths and extremely low power consumption. The measurements were conducted by focusing free-space THz fields onto the samples. However, we have very recently conducted 2D THz spectroscopy of magnons using an integrated solid-state platform in which the THz fields are generated optically and interacted with a sample to produce new nonlinear THz signals that are measured optically, all with no free-space THz field propagation. The system is compatible with samples in a magnet bore or an XFEL facility. Finally, we have recently used enhanced THz field amplitudes to generate large-amplitude magnon responses, revealed through multiple-quantum (up to 7-quantum) coherences observed in 2D THz spectra. We are hopeful that versatile switching of macroscopic magnetization will be possible.

We have used THz fields to induce a transient ferroelectric (FE) configuration in $SrTiO_3$, starting in its low-temperature quantum paraelectric phase [4]. Since we used a spatially uniform THz driving field, we initially believed that the induced FE ordering, monitored using optical SHG and birefringence measurements, was also spatially uniform. But in more recent XFEL diffuse scattering measurements, the FE dynamics were observed not at a Bragg diffraction peak but off-Bragg by a wavevector magnitude that indicated FE regions of ~ 10 nm dimensions [5]. The results strongly suggested that pre-existing, randomly oriented polar nanoregions were driven into a highly oriented collective configuration. The soft optical phonon and transverse acoustic phonon dynamics are consistent with theoretical predictions that a spatially modulated polarization-density wave configuration can be formed, stabilized by flexoelectric effects. We hope to combine x-ray transient grating and THz excitation to induce a potentially metastable configuration with long-range spatially periodic ordering.

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Quantum Entangled X-ray Beams

DE-SC0023176

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Abstract: Photons play key roles in many aspects of Quantum Information Science (QIS), especially as entangled pairs. The Hong-Ou-Mandel (HOM) effect is the most famous example, where pairs of indistinguishable photons are combined into maximally entangled N00N states with N=2 (biphotons). Although x-ray synchrotrons are now among the brightest sources of photons in existence, efforts to apply quantum optics to x-rays have been quite limited. This QIS project aims to bring quantum optics to x-ray synchrotrons by using HOM interference to generate pairs of entangled x-rays. Preliminary results from recent beam time at the APS-U x-ray synchrotron (Argonne) show the defining property of HOM for a pair of indistinguishable photons entering the two input ports of a beam splitter: instead of one photon exiting from each of the two exit ports, some of these pairs bunch together into one or the other exit port. Quantum mechanics forbids such photons from the classically allowed choice of exiting on separate paths. Boson statistics requires them to bunch together. The result is an entangled two-photon state.

Reproducing the HOM effect with a synchrotron source requires first splitting the beam so that the two beams can be recombined at a second (HOM) beamsplitter. This required the development of a new Mach-Zehnder interferometer (Figure 1). Obtaining two indistinguishable photons from the same x-ray bunch requires a very high brightness source to increase the photon degeneracy factor. To utilize the enhanced brightness of the APS-U it was essential to preserve brightness throughout, so we developed a new silicon Bragg beam splitter to minimize losses.[1] Furthermore, a synchrotron HOM measurement needs to record and analyze the exit photons from each synchrotron pulse, which in the APS-U brightness mode has a rate of 13 MHz. For this we developed a novel scheme involving two APD detectors and an FPGA-based signal analysis system.[2]

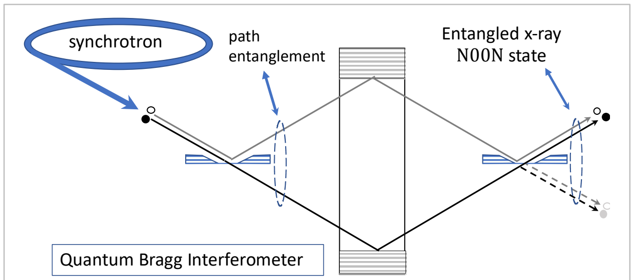

Figure 1: Overview the interferometer that splits the synchrotron beam and recombines them at the HOM beamsplitter, creating a two-photon maximally entangled N00N state.

Data from over 10^8 individual measurements using 8 keV x-rays at the APS-U synchrotron (Argonne) were recorded as the HOM beamsplitter was translated through the intersection point where the incident photons can be indistinguishable. As shown in Figure 2, the rate of coincident pairs (1-1) dropped while the rate of two photons together (2-0 and 0-2) increased by a similar amount. The difference shown corresponds to well over 10^4 entangled x-ray pairs per second. Further experimental improvements and beamline optimization should increase this rate substantially over these preliminary results.

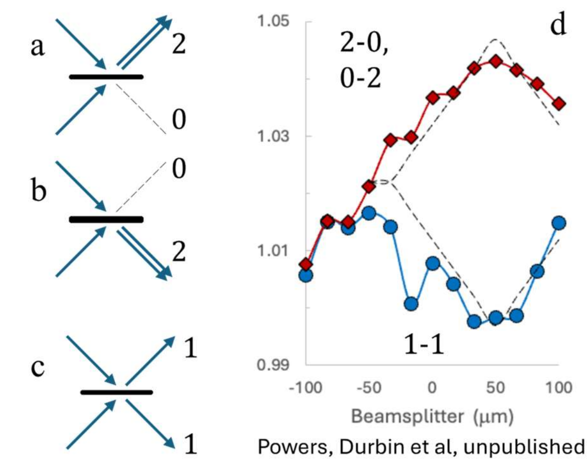

Figure 2: a) and b) show the bosonic bunching of exit photons that combine to form a N00N state; c) the two photons taking separate exit paths (classically allowed but quantum forbidden); d) data with 8 keV x-rays (APS-U) showing the HOM dip in coincident pairs and corresponding increase in biphoton bunching.

The goal of creating quantum entangled x-ray beams appears to be in reach. The overall mission is to use these beams as probes for detecting entanglement in quantum materials. Such an effort would begin with resonance enhanced x-ray scattering (REXS) in the 7-12 keV energy range. A number of interesting quantum magnets are available containing elements with L_{III} absorption edges in this range, including Ho, Er, Yb, Hf, Os, and Ir.

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Project Title: Understanding the Many-body Properties of Rare Earth-Based Materials Relevant to Next Generation QIS Applications

Award Number: PM-062
Abstract Title: Detecting Multipartite Entanglement in Real Materials

Names of PI, (co-PI): **R. Konik[1] (PI), A. Weichselbaum[1](co-PI), T.C. Wei[2] (co-PI), Rajesh Malla (Postdoc)**

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Abstract: We have developed a new means to quantify multipartite entanglement in a quantum material. Multipartite entanglement is relevant to a range of quantum technologies including quantum communication, enhanced quantum metrology, quantum sensing, quantum machine learning, quantum imaging, and quantum computation. In our work we have shown how to turn the spectroscopic information measured in single electron probe techniques such as scanning tunneling microscopy (STM) and angle resolved photoemission (ARPES) into a lower bound on a material's multipartite entanglement. We have done this in the framework of the quantum Fisher information (QFI).

The quantum Fisher information (QFI), a concept from quantum metrology, provides a way to connect bounds on the multipartite entanglement with integrated spectral information that is obtained in a spectroscopic experiment. This connection had been established previously in neutron scattering experiments measuring the dynamical spin response of materials [1]. We have shown in this work how to use the spectroscopies measured in an ARPES or STM experiment to infer a value of the quantum Fisher information (QFI) and then use this value to place a bound on the multipartite entanglement in the material [2]. We have done so considering the symmetries of the material, in particular particle number conservation. The bounds that the QFI must exceed in order to infer a certain level of entanglement when symmetries are present requires a non-trivial computation. These bounds are described in Fig. 1 when particle number conservation is present. While computing these bounds does require additional work, in doing so we are able to extract more fine-grained information about the entanglement in a system. We are not only able to tell whether a block of entangled n-sites is present in the material's wavefunction, but we also obtain information on the presence of patterns of entangled blocks.

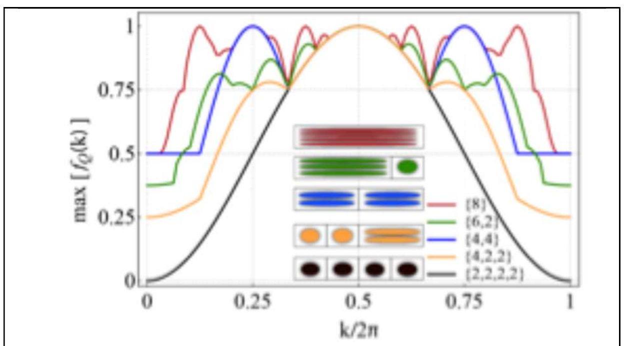

Fig. 1: The maximal QFI for various entanglement patterns is plotted as a function of k for an eight-site spinless fermionic system at half-filling. The entanglement patterns are shown schematically in the inset. The system is split up into independent blocks where electrons are confined within a block as visualized by the colored clouds. The smallest block size here contains two sites and the bars separating individual sites have been omitted within a block.

As future work in this space, we intend to use symmetries to refine the QFI bounds that are employed in neutron scattering spectroscopy to show that magnetic materials are entangled. In preliminary work, we have shown that neutron spectroscopy can infer more entanglement in a spin chain with SU(2) invariance than previously thought.

We also plan to investigate the role that degrees of freedom not directly probed by a spectroscopy affect the level of entanglement that can be detected. As an important example, phonons are typically present in quantum materials. However, in Ref. [2], our investigation supposed that only itinerant charge degrees of freedom were present. We thus plan to investigate how phonons alter the QFI bounds that must be exceeded for entanglement to be detected. More generally we will investigate the performance of the QFI as a means to detect entanglement using spectroscopies when there are multiple degrees of freedom present in a material.

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Non-Equilibrium Physics at the Nanoscale
DE-FG02-05ER46225

Fault-Tolerant Majorana-Based Quantum Algorithms in Topological Superconductors

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Abstract

Advances in the quantum engineering of complex systems at the atomic level, in combination with the ability to probe and manipulate them at nanoscopic length and time scales, have not only paved the way for next generation quantum materials, but have also opened unprecedented opportunities for advancing quantum information science and the realization of quantum computing. The PI's research program has therefore identified a critical need to develop a theoretical framework to investigate and visualize non-equilibrium processes over varying time and length scales that are key to the implementation of quantum algorithms. To this end, the PI's program has focused on two-dimensional (2D) topological superconductors that host Majorana zero modes (MZMs) and Majorana edge modes (MEMs) whose non-Abelian braiding statistics and robustness against disorder and decoherence has provided a new framework for the realization of fault-tolerant quantum computing. The PI's research program has made significant advances toward these goals by developing novel theoretical methods to (i) simulate the full many-body dynamics of fault-tolerant quantum algorithms for unprecedented system sizes up to 2500 sites and on time scales ranging from a few femtoseconds to nanoseconds [1,2], and (ii) visualize quantum process such as MZM braiding in space and time via the non-equilibrium local density of states, N_{neq}, which is proportional to the differential conductance measured in scanning tunneling spectroscopy [1]. In particular, the PI's group demonstrated the first, successful simulation of a Majorana-based, fault-tolerant quantum algorithm to solve the **Bernstein-Vazirani problem** in 2D magnet-superconductor hybrid (MSH) structures from initialization to read-out of the final many-body state [3]. Using the Majorana zero modes' topological properties, we introduced an optimized braiding protocol and a scalable architecture for the implementation of the algorithm with an arbitrary number of qubits. We visualized the algorithm's execution in real time and space by computing the zero-energy non-equilibrium local density of states N_{neq} [see Fig.1(a)], allowing us to represent the quantum code via Majorana world lines [see Fig.1(b)]. We showed that the algorithm's final qubit state can be read out using capacitive charge measurements, allowing us to even identify faulty implementations of the algorithm.

We subsequently demonstrated that a crucial component of quantum algorithms -- **the quantum memory** – can be realized using Majorana edge modes [4]. To this end, we proposed a novel material platform in which trivial domains are embedded in a 2D topological superconductor (see Fig. 2). We showed that while MEMs can store qubit states, MZMs located in vortex cores [5] can be employed to execute quantum gates. This execution is achieved by moving vortices in real time through the edges of topological phases, converting delocalized MEMs into localized vortex core MZMs (see Fig. 2). Computing the full many-body dynamics of quantum gate processes, we demonstrated that all Clifford gates can be successfully realized in this novel material platform, opening the path for the future implementation of complex, scalable quantum algorithms.

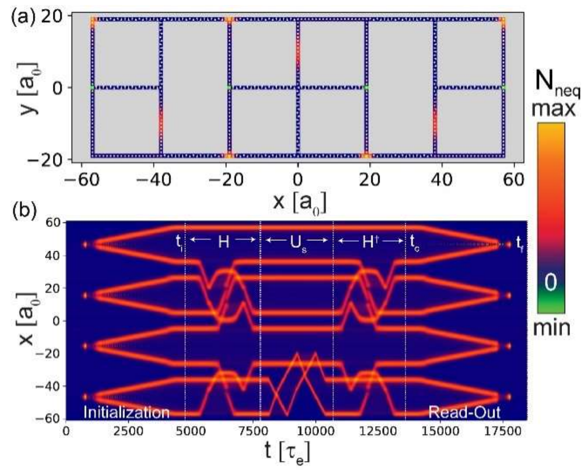

Figure 2 (a) N_{neq} during a Hadamard gate. (b) MZM world lines of the BV algorithm – the quantum code.

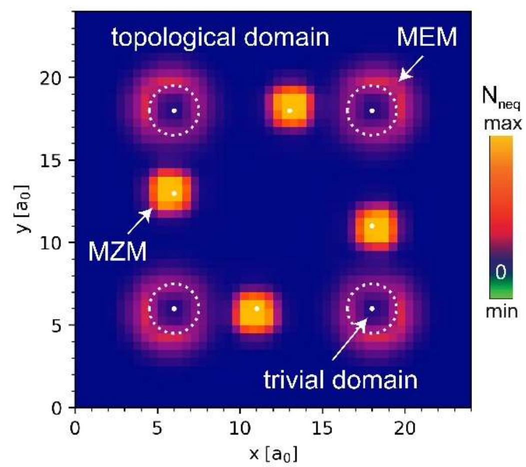

Figure 3: N_{neq} during a Hadamard gate with MEMs and vortex core MZMs.

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Mapping the genome of coherent quantum defects for Quantum Information Science
DE-SC0022289

High-throughput computational discovery of new quantum defects in silicon

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Abstract: Color centers in semiconductors are important building blocks of quantum networks and sensors. A series of well-established quantum defects exist especially in diamond (e.g., the NV center and the silicon divacancy) and have shown promises in demonstrating entanglement at long distance or quantum sensing. However, these defects have all limitations coming from their host (e.g., diamond is difficult to process) or from the defect in itself (e.g., the NV shows low optical coherence). Ultimately the performances of quantum networks and quantum sensing device will depend on the properties of these defects and there is a growing need to discover and develop new quantum defects sometimes in emerging hosts and/or understand their properties.

We will present our approach to systematically search for new quantum defects with attractive properties using first principles high-throughput computational screening and focusing on silicon. Silicon is an emerging host for quantum defects with tremendous advantages in terms of ease of process and nanofabrication. Color centers in silicon have recently started to be used in quantum applications (e.g., T center, G center, …) including showing indistinguishable photons.[1] We will show how a database of first principles computed data on 20,000 charged defects in silicon can be built and used to analyze the challenges and opportunities for designing quantum defects in such a small band gap material.[2,3] A special focus will be on the recently discovered T center-like defects which could form attractive spin qubits.[4] We will as well touch on how our approach can be extended to diamond, the most common host in for quantum defects proposing new defect with higher optical coherence.[5] In these different hosts, the comparison between theory and experiment will be highlighted as well as the challenges in theoretical screening and how close work with experiments can direct the necessary development efforts in first principles methods.

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Topological Superconducting Heterostructures for Quantum Sensing
DE-SC0022245

Tunable Josephson Diode Effect for Qubit Noise Shielding

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Abstract

We present our recent work aimed at enhancing the quality of planar Josephson junctions based on SC-SM heterostructures[1,2], and designing and fabricating SQUIDs exhibiting a field-free JDE[3,4,5]. The enhancement of the quality of planar Josephson junctions based on SC-SM heterostructures is essential for the broader goal of the project to fabricate quantum sensors, and fault-tolerant qubits, based on the unique properties of the topological superconducting states that can be realized in such junctions. The JDE allows the realization of ideal, dissipationless, diodes and is therefore key toward the project's goal to realize noise-free, compact, quantum devices to shield qubits and quantum sensors from the noise inevitably introduced by the read-out electronics.

We have made progress toward the epitaxial growth of Al on antimonide heterostructures[1], an important step toward the realization of novel SC-SM heterostructures that should allow the realization of topological superconducting states without the need of external magnetic fields. We have fabricated Josephson junctions with periodic hole structures on the superconducting contact leads (Fig. 1(a)) on InAs heterostructures with epitaxial superconducting Al. By depleting the chemical potential inside the holes' region with a top gate (Fig. 1(b)), we observed an enhancement of the junction' supercurrent, (Fig. 1(c)).

We have developed the theory for the a.c. JDE in asymmetric SQUIDs[3] formed by Josephson junctions with different current-phase relations. We find that in such SQUIDs a strong a.c. JDE can be realized, with strength, and polarity, that can be tuned also by varying the a.c. power. For SQUIDs formed by one topologically trivial junction, and one topological junction, (Fig.2 (a)) we found that the JDE is particularly strong, tunable (Fig. 2 (b)), and could be used to help detect the topological nature of planar Josephson junctions.

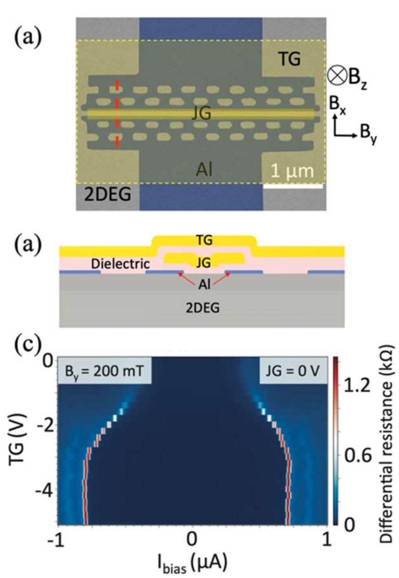

Figure 1. Planar Josephson junction based on SC-SM

We have studied SQUIDs based on heterostructures formed by the Dirac semimetal Cd_3As_2 and Al (Fig. 2(c)). A careful analysis of the a.c. response of such SQUIDs[4] revealed the presence of anomalous collective modes, Leggett modes, that strongly suggests the establishment of an uncommon superconducting state that spontaneously breaks time reversal symmetry. Such state should lead to a JDE *even in the absence of any external magnetic field.* We have observed such JDE[5] (Fig. 2(d)). This result is potentially transformative for the shielding from read-out noise of qubits and quantum sensors because it allows the realization of ideal circulators and isolators without the need of magnetic fields, a fact that greatly reduces their size making them extremely compact and potentially viable for on-chip integration.

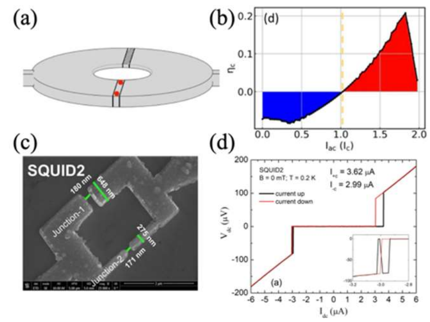

Figure 2. Results for JDE

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Topological Superconducting Heterostructures for Quantum Sensing

DE-SC0022245
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Robust Superconducting Diode Effect in MBE Grown Dirac Semimetal Cd_3As_2

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Abstract

We present our results of robust and symmetric-in-magnetic-field SDE in asymmetric superconducting quantum interference devices (SQUIDs) [1] on Dirac semimetal Cd_3As_2 thin films [2]. Molecular-beam epitaxy (MBE) is utilized to grow our high mobility Cd_3As_2 thin film of 240 nm thickness. Details about growth can be found in Ref. [2]. Electron beam lithography is used to define SQUID structures. The bottom inset of Figure 1a shows an SEM image of this SQUID. The dimension of the SQUID is ~ 1.3 × 1.0 μm^2. Electron beam evaporation is used to deposit 5 nm of titanium (to promote metal adhesion to the substrate), 230 nm of aluminum (Al), and 10 nm of gold (to mitigate oxide formation on the Al). Fig. 1a shows the quasi-four-terminal sample resistance R_{xx} as a function of temperature (T). The resistance is nearly constant and reaches R_{xx} ~ 6 Ω at high temperatures. It drops to zero at T_c ~ 1.2 K signaling a superconducting transition in the device. The top inset of Fig. 1a shows R_{xx} as a function of magnetic (B) field measured at 0.3 K. In the low field regime |B| < 50 mT, the device is in the supercurrent regime and R_{xx} = 0 Ω. Beyond a critical B field of ~ 53.5 mT, the supercurrent state is destroyed, and the device enters the normal state regime. Figure 1b shows the I-V characteristics in two current sweep directions (0 → P and 0 → N), measured at zero B field. A magnetic-field-free SDE is clearly observed. Furthermore, as shown in Figure 1c, the difference between the switching currents $I_{\pm c}$ is independent of the strength and polarity of the out-plane magnetic fields in the range of -10 mT and 10 mT.

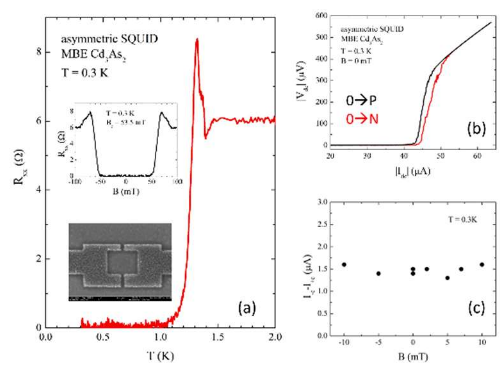

Figure 1: Superconducting diode effect in MBE grown Cd_3As_2. (a) R_{xx} *vs* T trace shows a superconducting transition at T ~ 1.2K. (b) Current-voltage (I-V) for two current sweep directions. (c) The difference of critical currents as a function of B field.

The observation of a zero-field SDE is consistent with our previous work on mechanically exfoliated Cd_3As_2 thin flake [3], in which we show that the coupling between the surface and bulk superconducting channels in a Dirac semimetal can provide an intrinsic TRS breaking mechanism and, consequently, give rise to a field-free SDE. The same mechanism is applied here for the observed field-free SDE in the MBE grown Cd_3As_2 thin films. To understand the origin of field resilience in our devices, we refer to recent work on SDE in multiferroic Josephson junctions [4] and speculate that an antiferromagnetic spin-spiral order (helimagnetism) might be responsible for this resilient symmetric-in-field SDE behavior. Indeed, an antiferromagnetic phase has been predicted in Dirac semimetals like Cd_3As_2 in the presence of interactions [5]. This could serve as a possible explanation for the observed robust SDE.

We point out that a robust and symmetric SDE in SQUIDs using MBE grown Cd_3As_2 thin films should allow future superconducting diode applications to take advantage of state-of-the-art semiconductor synthesis and processing.

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Quantum entanglement between a semiconductor donor and a trapped ion

DE-SC0020378

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UW Undergrad students: Sam D'Ambrosia (undergrad)
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Abstract: The central goal is to connect trapped ion qubits to single defects in semiconductors through a photonic link. This will be the first demonstration of entangling disparate matter qubit systems (aside from neutral atoms/ions of the same species) and mark progress toward scalable hybrid quantum systems. The immediate focus on the project is to generate indistinguishable single photons from the two different quantum emitters, a ytterbium trapped ion and a donor qubit in ZnO (either indium or tin-lithium). The two systems emit at nearly identical frequencies which can be bridged via Stark tuning and/or detuned Raman emission. The latter also enables temporal matching of the emitted photons [1].

On the trapped ion side, the Blinov group has performed theoretical analysis and numerical simulations of single photon pulse shaping to achieve >99% overlap of the emitted photons with the desired pulse shape, using both traditional computational techniques, such as genetic learning algorithms, and machine learning techniques for fast feedback. We have predicted that reversing the phase of the driving field will allow for photon shapes shorter than the limit set by the natural lifetime of the ion excited state, which could be useful given the shorter ZnO donor-bound-exciton lifetime.

On the ZnO donor side, the Fu group has realized indium donor formation via implantation and annealing [2] as well as isolation of single donors [3]. Most recently, in a related project, we have realized the synthesis of a new donor, a Sn-Li complex, which is even closer in wavelength to the Yb transition. In the Sn-Li complex we have resolved the hyperfine interaction to the longer-lived nuclear spin memory. Moving forward we may utilize the Sn-Li for this project in place of the donor.

The next step on the trapped ion side is to verify pulse shaping experimentally using a single trapped Yb^{+} ion quantum emitter, quantified by Hong-Ou-Mandel interference with an attenuated coherent source. Trapping has been a struggle over the past year in the Blinov lab. In recent weeks, we completely rebuilt the parabolic mirror trap. It is currently back on the optical table, being set up for operation. We also constructed a backup linear trap which should be more robust, albeit with a lower photon collection efficiency (about 2% vs. 40% for the parabolic mirror trap), to mitigate risk.

In parallel, on the donor side, we are focusing our effort on ZnO photonic device fabrication for enhanced photon collection efficiency. Device fabrication in ZnO is a relatively immature area. Process development is near completion. While prior results have included ZnO donor formation via implantation and also single donor isolation, both have not been achieved simultaneously. Device integration will enable deterministically created single donors as well as the photon collection efficiency needed for hybrid entanglement schemes. The Raman excitation scheme used to match the donor photon with the ion will be informed by our recent fundamental work on the optical properties of donors [4].

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Integrated Materials Platforms for Topological Quantum Computing Devices
DE-SC0019274

Abstract Title: Synthesis of improved mobility InAs and InGaAs-quantum wells and of PbSe SAG nanowires

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Abstract: A significant challenge towards developing robust quantum computing systems with large numbers of logical qubits is rapid decoherence. A promising approach to addressing this challenge is based on the concept of fault-tolerant quantum computing architectures that leverage topological quantum states such as Majorana zero-modes (MZM). Substantial progress toward realizing and detecting topological states has been enabled by quantum devices which combine semiconductors with superconductors due to the combination of processing-friendly materials that offer high spin-orbit coupling, large g-factors, ballistic transport, and the ability to create transparent interfaces with various superconductors. However, further breakthroughs leading to topological quantum computation require next-generation quantum nanostructures that offer substantial improvement in the electronic purity together with optimal coupling across the superconductor-semiconductor interface. *Here we discuss our recent results under this project, focusing on the synthesis of higher purity III-V and IV-VI materials platforms.* This work is a subset of our larger effort supported by this grant, which combines quantum device fabrication and characterization with materials synthesis and data interpretation informed by ab initio simulations and machine learning to develop robust, tunable and scalable platforms for the manipulation of quantum states

(i) Enhancing mobility in InGaAs and InAs quantum wells on InP(001) substrates: Critical to the superconductor/semiconductor multiterminal Josephson junction device effort is to increase the carrier mean free path (mfp) in the semiconductor. Although very high mobility InAs quantum wells can be grown on nearly lattice matched GaSb utilizing AlGaSb barriers, parallel and side wall conduction and the processing challenges of Sb-based semiconductors, makes semi-insulating InP substrates a highly desirable choice. High In content (≥75%) InGaAs quantum wells grown on InP substrates rely on InGaAs cladding layers and InAlAs barrier layers to confine electrons within a thin well. The highest mobility QWs are limited by interfacial roughness scattering and alloy disorder scattering in the quantum well, cladding and buffer layers. Efforts at UCSB have focused on increasing the electron mobility and thus the mfp in MBE-grown quantum wells (Fig. 1). The approaches taken involved investigating the use of i) digital alloys (superlattices of monolayer thicknesses of InAs and InGaAs) to reduce the alloy disorder scattering within the quantum well while achieving the same average composition as a high In (≥75%) concentration random InGaAs alloy and ii) strain compensation techniques during the cladding and barrier layer growth. The mobility of 545,000 cm^2/Vs achieved for the digital alloy InGaAs quantum well is higher than that highest

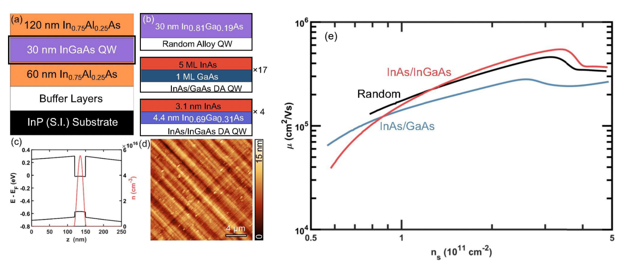

Fig. 1: (a) Sample structure schematic. (b) Different samples in the study. (c) Self-consistent Schrödinger-Poisson simulation of the sample structure. (d) Representative AFM micrograph of InAlAs top barrier; crosshatching typical of metamorphic growth is observed. (e) Gated Hall bar measurements of mobility vs gated carrier density.

previously reported for the equivalent random alloy ~430,000 cm^2/Vs [1]. By utilizing strain compensation during the cladding and barrier layer growth, the critical thickness of the InAs can be increased before relaxation occurs and a substantially wider quantum well with increased mobility can be grown [2]. Quantum point contact devices showed 1D conductance quantization ($2e^2/h$) up to 17 plateaus [3].

(ii) Synthesis of PbSe semiconductor-superconductor materials platforms: We are also developing procedures for PbSe nanowire networks via selective area growth (SAG) using molecular beam epitaxy (MBE) (Fig. 2), which leverage our prior work [4]. We are particularly sensitive to growth conditions that yield continuous and smooth films, with minimal non-selective growth. At the outset, we have identified a growth window of 360-375 °C for PbSe to grow selectively on exposed crystalline GaAs. Here, the sticking of PbSe on adjacent dielectric regions is reduced to zero. This has allowed us to make smooth nanowires. The tendency for PbSe to congruently sublimate makes the use of compound sources convenient for MBE. However, we are learning that the source stoichiometry is crucial in obtaining good SAG synthesis. We show results with two batches of source material, Pb-rich PbSe and stoichiometric PbSe. We find that stoichiometric PbSe yields continuous nanowires. Pb-rich PbSe requires the addition of Se flux to improve morphology. The background carrier concentration is another important synthesis target, in addition to the surface morphology. We have discovered that PbSe is sensitive to atmospheric oxygen, and exposure at room temperature converts the films to p-type in the range of $10^{18}/cm^3$. We are showing that in-situ capping of the samples with wider bandgap GeSe (~1.1 eV bandgap) can sufficiently prevent oxygen diffusion into the PbSe to preserve the unintentional (n-type) doping of the as-grown PbSe.

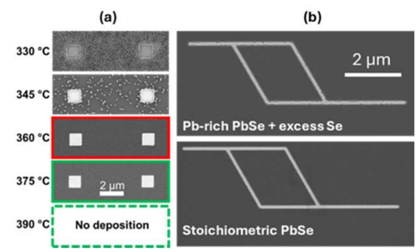

Fig. 2: Synthesis of PbSe SAG nanowires. (a) Growth selectivity of PbSe is enhanced at 360-375 °C, just at the limit of sticking. Polycrystalline nuclei are seen at temperatures of 330-345 °C. (b) A stoichiometric PbSe source results in more continuous nanowires.

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Abstract Title Parametric interactions in superconducting multi-qubit+cavity architectures

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Abstract:
In the PIQUE (Parametrically-Induced QUantum Engineering) Program, we are developing a multi-element architecture that can cancel static couplings between multiple circuit-QED elements, like qubits and cavities, but still allow strong and tunable parametric interactions. As a demonstration, we have utilized a galvanic SQUID inductive coupler that is shared by two qubits and a cavity (see Fig. 1). Here, the inductive coupling cancels the static capacitive coupling between any pair of elements at a static operation flux. Cancellation minimizes both qubit cross-talk, unintended interactions, shot-noise qubit dephasing, and energy decay of the qubits. If we modulate the flux in the SQUID coupler, we can parametrically produce all-to-all interactions between all three elements. In our experimental investigation of this system [1], we verified minimal coupling between the qubits and cavity at the cancellation flux. With applied parametric drives to the coupler, we developed a new tunable dispersive readout technique [2].

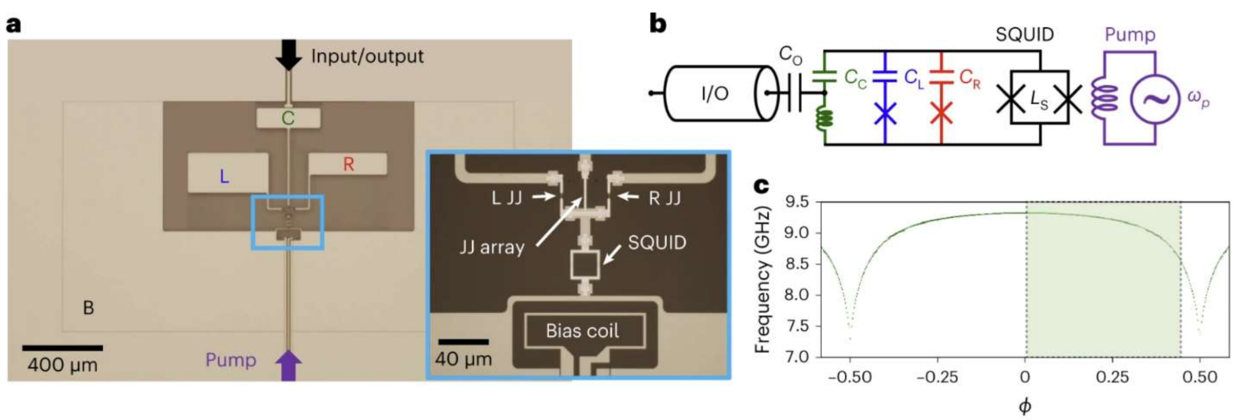

Fig. 1: a, Optical micrograph of a tunable circuit–QED system, where JJ stands for Josephson junction.**b**, Schematic of the system including three elements: cavity mode (C), left transmon (L) and right transmon (R). The parametric flux drive is applied through a shared SQUID coupler. **c**, Periodic cavity spectrum, where green shading denotes flux operation region.

We have shown experimentally that we can create tunable dispersive shifts between either or both qubits and the cavity and this dispersive interaction grows with the size of the parametric drive strength. In addition, we can also access both positive and negative dispersive shifts depending on the pump frequency (See Fig. 2). This demonstration of tunable qubit readout is versatile. It is possible to cancel residual dispersive interactions and tune joint two-qubit readout to distinguish all four readout states or to make odd or even parity states indistinguishable, which is useful for generating two-qubit entanglement or error correction.

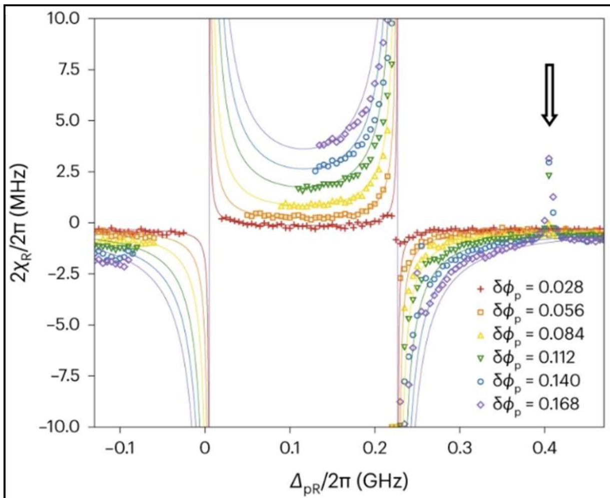

Fig. 2: Total dispersive shift $2\chi_R/2\pi$ as a function of $\Delta_{pR}/2\pi$.

In next-generation PIQUE program, we have extended this architecture to develop a two-cavity + qubit system where the long-lived "readout cavity" is isolated and the "buffer cavity" provides photon access into and out of the system. With this device we are developing a sequentially pulsed parametric dispersive readout with integrated *in-situ* amplification. This extends our previous work by providing a phase gate on the readout cavity to capture qubit state information which can be amplified and accessed through the buffer cavity. The pulsed nature of the readout protocol protects the qubit and readout cavity throughout the measurement process.

The techniques we have developed for these architectures can be scaled to larger numbers of qubits and cavities. For instance, in next-generation PIQUE program, we have developed a 3-qubit + cavity system with no static interactions, but all-to-all parametric interactions mediated via a single coupler. This general increase in connectivity is not "hard-wired" and so that various Hamiltonians become software programmable via frequency-selective pumping. In an attempt to scale to larger numbers of elements we have developed compact, high quality capacitors that can be used in coherent superconducting qubits [3].

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Understanding the relationship between surface lattice and single photon emission dynamics in strongly confined cesium lead bromide perovskite quantum dots
DE-SC0024441

Towards non-blinking and photostable perovskite quantum dots

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Abstract: Lead halide perovskite quantum dots (QDs) have emerged as promising single-photon-emitting materials due to their uniquely bright triplet excitonic emissions. However, their insufficient single-photon emission performance and stability, often broadly attributed to fragile crystal lattices, remain poorly understood. Our QIS project aims to explore the effect of surface lattice rigidity on the single-photon emission dynamics in individual perovskite QDs. To reduce selection bias in single QD spectroscopic studies, we first sought to precisely control the size and surface morphology of perovskite QDs during synthesis by unraveling their growth mechanisms. To understand the impact of surface lattice on the stability of single-photon emission, we then embedded single QDs into a molecular crystal matrix (**Figure 1a**). The attractive intermolecular interactions between low-steric ligand tails in the crystallized matrix, such as π-π stacking, can promote the formation of a nearly epitaxial ligand layer, significantly reducing the QD's surface energy. Here, we demonstrate that single $CsPbBr_3$ QDs covered by stacked phenethylammonium (PEA) ligands exhibit nearly non-blinking single-photon emission with high purity (~ 98%) and extraordinary photostability (12 hours of continuous operation and saturated excitations), allowing for the determination of size-dependent exciton radiative rates and emission linewidths of $CsPbBr_3$ QDs at the single-particle level (**Figure 1b-1g**). Additionally, benefiting from the material design, we have investigated the impact of various surface defects on the purity of single-photon emission from $CsPbBr_3$ QDs.

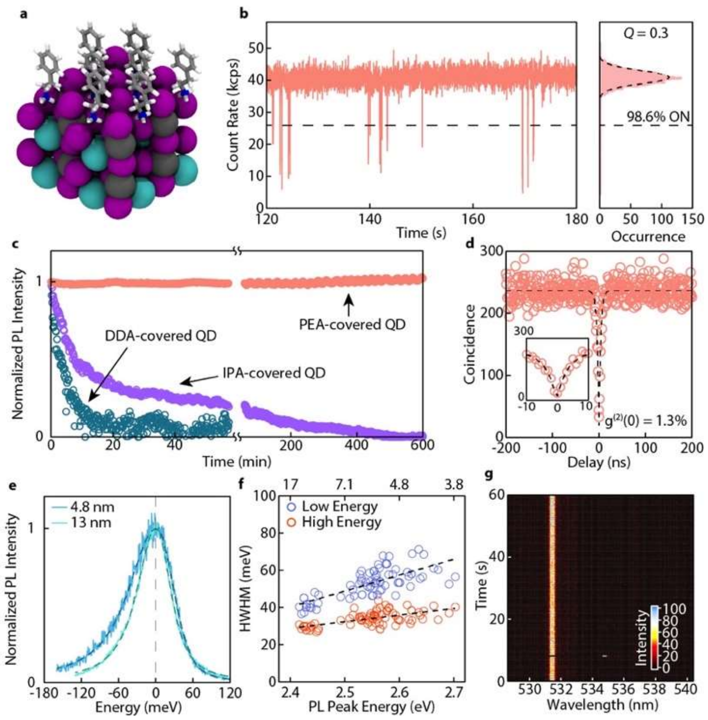

Figure 1. (a) A model showing molecules (PEA) epitaxially aligned on the surface of perovskite QDs. (b) PL blinking traces and intensity distribution histograms of a 4.5 nm $CsPbBr_3$ QD embedded in PEA matrix. (c) Normalized PL intensity of a collection of single PEA-covered $CsPbBr_3$ QDs (red) under 10 h of continuous laser excitation. In contrast, traditional ligand-covered QDs experienced severe photodarkening after a few minutes. (d) Second-order correlation ($g^{(2)}$) function of the QD in (b). (e) PL spectra of two single $CsPbBr_3$ QDs. Dashed curves are fits, each using two Voigt functions with a common peak position for the asymmetric spectra. (f) PL linewidths (half-width half-maximum, HWHM) extracted from the high and low energy sides of PL spectra for 81 QDs. (g) PL spectra-time trace of a single $CsPbBr_3$ QD monitored at 3 K exhibiting undetectable spectral jumps/diffusions. ((b-f), adapted from Reference 2, under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International License)

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Project title: Color centers in noise-free hosts for quantum sensing and communication applications

Award Number: DE-SC0025486

Authors: Kusal Abeywickrama[1], Pralay Paul[1], Melissa Artola[1], Sreehari Purayil[1], Sumit Gaswami[1], Dhiman Biswas[1], Casey Kerr[1], Mritunjaya Parashar, Mohin Sharma[3], Darshpreet Kaur Saini[3], Bibhudutta Rout[3], Thirumalai Venkatesan[1], and **Alisa Javadi**[1,2]

Affiliation: [1]Department of Physics and Astronomy, The University of Oklahoma, [2]School of Electrical and Computer Engineering, The University of Oklahoma, [3] Department of Physics, The University of North Texas

Abstract: The spin state of an electron in quantum emitters is pivotal in quantum technologies such as quantum communication and quantum sensing. For both types of quantum emitters, color centers, and rare-earth dopants, and for the materials with the highest quality, the hyperfine interaction between the nuclei of the host crystal and the spin of the electron gives rise to fluctuating electron energy levels, which in turn cause decoherence of the spin state. Suppressing this noise calls for isotopically purified samples, which is an expensive process and poses a problem for scaling technologies based on these materials.

Interestingly, a limited number of elements (oxygen, calcium, and cerium) are free of magnetic nuclei in their naturally abundant isotopes. Given the sparsity of magnetic nuclei, color centers in naturally occurring CeO_2 are projected to have remarkable coherence times as long as 47ms [1]. This characteristic significantly enhances the material's potential for quantum sensing and communication applications.

CeO_2 benefits from several other advantages as a host material. Its relatively large bandgap (3.5 eV) allows it to accommodate color centers ranging from visible frequenciesto the mid-IR range. CeO_2 is compatible with silicon [2] and, as a lanthanide-based host, it is adept at supporting lanthanide emitters, which are significant due to the long coherence of 4f energy levels and the long coherence of their nuclei. In addition

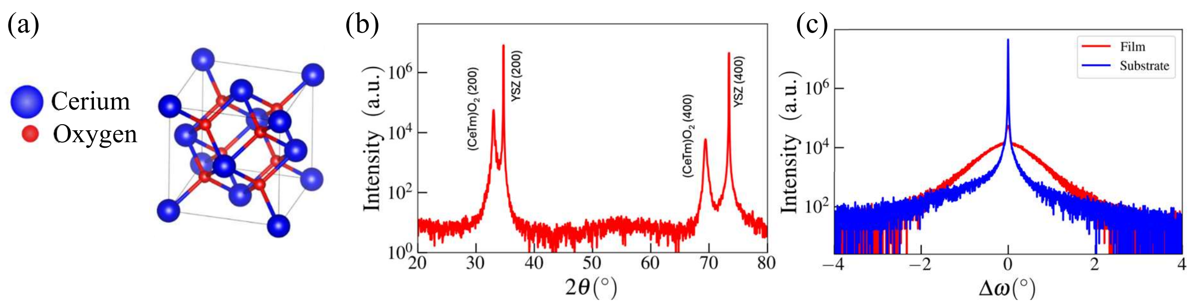

Figure 1 (a) The crystal structure of CeO_2. (b) XRD measurements on a thin film of CeO_2 grown on YSZ substrate indicating single crystal films aligned with the YSZ axis. (c) Rocking curve of the film showing a sharp peak on top of a broader Gaussian background.

to its technological strengths, CeO_2's biocompatibility simplifies lab handling, broadening its applications.

One of the goals of this project is to investigate the potential of rare-earth dopants (specifically, thulium dopants) in CeO_2 for quantum information applications, including sensing and quantum memories. Figure 1a shows the crystal structure of CeO_2. In this project, we are using Pulsed Laser Deposition to grow thin films of CeO_2. The main advantage of PLD is its rapid turnaround time, which allows us to fabricate films with different thicknesses and doping concentrations in a short period of time.

We have managed to grow high-quality CeO_2 thin films doped with trivalent thulium ions (Tm3+) at concentrations of 0.1% on single-crystalline yttria-stabilized zirconia (YSZ) substrates. We have characterized these samples using various techniques, XRD, XRR, and AFM measurements. To achieve the desired doping concentration, we employed two distinct targets, a pure CeO2 target and a 10% Tm-doped CeO2 target and varying the laser pulses directed at each target. Figures 1b and 1c show some of these characterizations.

The XRD pattern of the thin film 1% Tm-doped CeO_2 , as shown in Fig. 1b , exhibits well-defined (200) and (400) diffraction peaks, confirming the epitaxial nature of the film. The presence of interference fringes around the (200) diffraction peak indicates a high degree of crystalline order and smooth interface quality. The measurements of the rocking curve performed on the (200) peak of the YSZ substrate show a sharp peak with a full-width-at-half-maximum (FWHM) of 0.01◦, whereas the corresponding measurement for the CeO_2 film doped with Tm reveals a combination of a sharp peak (FWHM 0.05◦) and a broader peak (FWHM 0.5◦). This suggests that the film remains strained in the initial growth stages near the substrate interface but progressively relaxes as the thickness increases while maintaining oriented growth. As evident from the XRD curves and Rocking curves, we have been able to grow highly crystalline films that are well-aligned with the crystallographic axes of the substrate. Furthermore, our atomic force microscope measurements show very smooth films with roughness below 0.5nm.

We characterized the optical properties of CeO_2:Tm^{3+} using photoluminescence (PL) and photoluminescence excitation (PLE) [3]. The Tm^{3+} ions in the sample were excited by a continuous wave UV laser at 355nm, plotted in in Fig. 2. The spectrum consists of strong emission peaks in red and NIR regimes with main four peaks at 654, 690, 793, and 808 nm which correspond to $^1G_4$-$^3F_4$,$^3F_3$,$^{2,3}H_6$,$^1G_4$-$^3H_5$ and $^3H_4$-$^3H_6$ transitions, respectively.

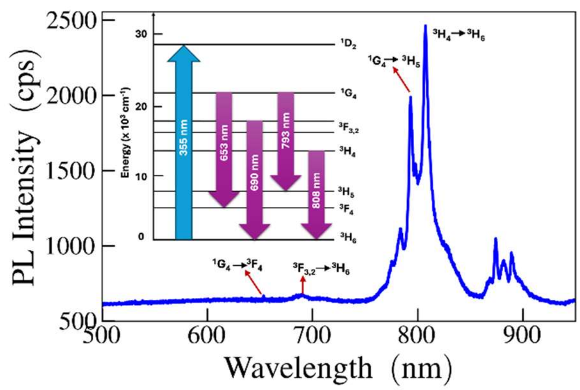

Figure 2 Fig. 2. PL spectrum of 550 nm thick 1% Tm3+ doped film.

These measurements are exciting first steps in our goal of using these dopants for quantum communication and sensing applications. As the next step, we are currently working on characterizing these transitions' quantum efficiency and coherence in pump-probe measurements. Another goal is to embed these films in photonic structures to enhance their collection efficacy. We have developed a novel growth method to directly deposit nanostructures of complex oxides into pre-patterned substrates and are in the final steps of refining this technique [4].

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Project Title: Interactions of QDs' Fast Light in Rb Vapors for Hybrid Quantum Information Science and Technology
Award Number: (unknown)

Abstract Title: Towards Hybrid Quantum Repeaters with Quantum Dots and alkali vapors

Names of PI: Joanna M Zajac (PI)[1]
Collaborators and contributors: Tobias Huber[2], Sven Höfling[2], Monika Dziubelski[1], and Sofia Pereira[1]

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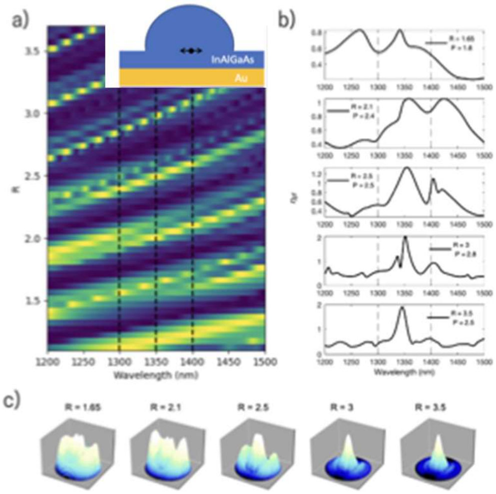
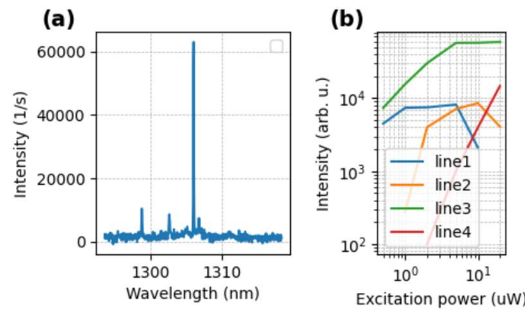
Abstract: III-V epitaxial QDs working at near-infrared (NIR) wavelengths had been a gold standard for quantum emitters and promising candidates for memories. They exhibit high brightness, indistinguishability, and work on demand. Future long distance quantum communication networks will utilize existing optical fiber infrastructure working between 1.3 and 1.5 µm. Shifting QDs operational wavelengths into telecommunication O- and C-bands is thus important for these applications[1]. There are two III-V semiconductor material systems that have been considered, namely, InAs/InP and InAs/GaAs QDs.

In this work, we discuss our novel sample design with QDs based on quaternary alloy InAlGaAs, lattice matched to InP substrates, however, maintaining shorter emission wavelengths using advanced growth techniques. To increase extraction efficiency from these emitters, we embed them into specially designed and optimized microcavity strucutres[2]. We use these QDs to develop hybrid repeater platforms working with alkali memories. We work with diamond configuration in Rb for four-wave mixing process to convert 1.3 µm signal photons to 795 nm idler. This conversion mechanism, unlike ladder configuration such as ORCA, exhibit noise due to spontaneous emission and four-wave mixing process; however, it also has superior storage properties under EIT. I will discuss our latest progress on this topic discussing growth, material characterization and device development under my Carrier Award Project in Quantum Information.





a) transmission maps vs radius (R) and wavelength (nm) for the cavity shown in the inset, b) Transmission cross-sections for fixed radius as a function of wavelength and in c) far-field emission patterns for indicated R.

a) Low temperature μ-photoluminescence (μ-PL) from QDs growth for this project showing high S/N at 60 kHz. b) Excitation power sweep for several QDs within the same wafer.

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Multiparticle Quantum Information Processing in Robust Plasmonic Lattices

DE-SC0021069

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Abstract

One of the most significant achievements of the second quantum revolution is the ability to perform tasks that are infeasible for classical computers. However, the stringent requirements for quantum resources, along with the presence of noise and loss, impose important limitations on technologies for quantum information processing. The aim of this research program is to develop robust protocols for quantum information processing by exploiting the additional interference pathways enabled by optical near fields to control complex multiparticle interactions within plasmonic platforms. Our ability to resolve specific dynamics in multiparticle plasmonic systems will be leveraged to advance quantum information processing in plasmonic lattices that are intrinsically loss-tolerant and operate without error correction. Unlike other quantum architectures, our plasmonic platform supports both bosonic and fermionic interactions, offering a robust foundation for quantum simulation and information processing.

I will describe our work on the nonclassical near-field dynamics of surface plasmons [1]. As shown in Figure 1, we isolated the constituent multiparticle subsystems of classical plasmonic waves and showed that their quantum dynamics are governed by bosonic or fermionic coherence processes [1]. Interestingly, the article *Multiphoton Quantum Statistics from Scattered Classical Light* highlights the implications of extracting quantum multiparticle systems from classical plasmonic platforms in the presence of losses [2]. Our work was selected as the Best of Optics in 2024 by *Optica's Optics & Photonics News* [3]. Remarkably, these findings enabled my team to develop robust multiphoton quantum technologies [4].

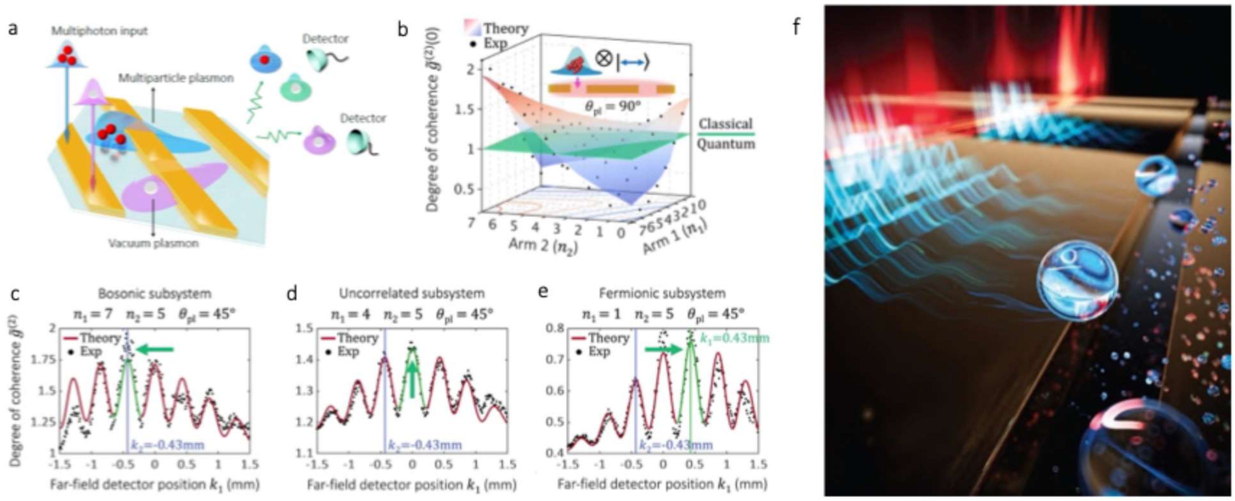

Figure 1. We investigated the nonclassical dynamics of plasmonic multiparticle systems illustrated in **a**. As reported in **b**, the constituent multiparticle subsystems that form classical plasmonic waves exhibit classical and quantum properties of coherence. For example, subsystems with a degree of coherence greater than one, such as the twelve-particle system ($n_1 = 7$, $n_2 = 5$) in **c**, exhibit bosonic correlations. Furthermore, systems with coherence degrees close to one are uncorrelated, as shown in **d**. Interestingly, quantum multiparticle systems with a degree of coherence below one show fermionic anticorrelations. This is shown in **e** for a six-particle system ($n_1 = 1$, $n_2 = 5$) [1]. The illustration in **f** depicts a metallic nanostructure hosting plasmonic waves produced by vacuum fluctuations of the electromagnetic field. It also shows the scattering of surface plasmons, which produces multiparticle systems with either bosonic or fermionic coherence properties [3].

We exploited these capabilities to design the plasmonic metacrystal shown in Figure 2. This structure creates a synthetic plasmonic lattice in the Fock basis, which is used to engineer multiparticle quantum systems with up to forty particles. The plasmonic lattice supports a large number of interactions that induce the formation of forbidden thermalization gaps. This novel quantum effect prevents the propagation of multiparticle states with certain statistical fluctuations.

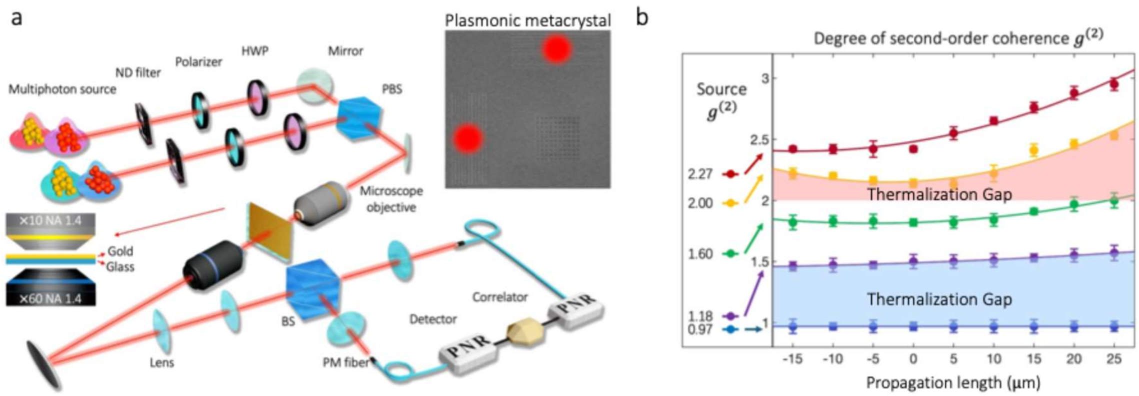

Figure 2. The experimental setup used to investigate thermalization gaps in quantum plasmonic metacrystals is illustrated in panel **a**. Two multiphoton sources are prepared in distinct polarization states using a polarizer and a half-wave plate (HWP). These sources are then combined via a polarizing beam splitter (PBS) and focused onto the two input ports of the plasmonic metacrystal, as shown in the inset of panel **a**. The transmitted multiphoton system is collected by a microscope objective and subsequently imaged using a tunable telescope, which allows measurements at different propagation planes of the plasmonic sample. The selected plane is directed through a beam splitter and analyzed using two photon-number-resolving (PNR) detectors. The metacrystal consists of one hundred plasmonic nanoantennas with varying orientations. Our experimental results are presented in panel **b**. The second-order degree of coherence of our multiphoton sources is shown in the left panel of **b**. Interestingly, the quantum statistical properties of the sources, which are not characterized by the eigenstatistics of the crystal, are modified by the crystal. These results demonstrate the first quantum statistical plasmonic metacrystal.

Future Plans

We are preparing a manuscript on quantum statistical plasmonic metacrystals. This is the first platform that enables the engineering of multiphoton states across all known statistical regimes. Moreover, it represents the first demonstration of plasmonic lattices employed to implement photonic crystals sensitive to the quantum statistical properties of the light field.

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Advanced in-situ analysis and ex-situ studies of 2D Hexagonal Boron Nitride and 2D Ferromegnets via Aberration-Corrected Scanning Transmission Electron Microscopy

DE-SC0023224

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The discovery of room-temperature single photon emission (SPE) in hexagonal boron nitride (hBN) launched it to the forefront of research as a promising platform for room-temperature quantum optics and photonics. Room-temperature quantum emission centers in hBN exhibit single-photon emission and optically addressable spin states, as desired for many quantum technologies. Here we present our published[1–3] results and ongoing work funded by this DOE project. We created and characterized nanopores in hexagonal boron nitride (hBN) via aberration corrected scanning transmission electron microscopy (AC-STEM) drilling and photoluminescent (PL) spectroscopy. Defects on the sub-nanometer to nanometer scale are induced in hBN through electron irradiation and AC-STEM drilling[1]. Confocal PL spectroscopy is subsequently employed to characterize the optical activity of the defects created through electron irradiation and drilling.

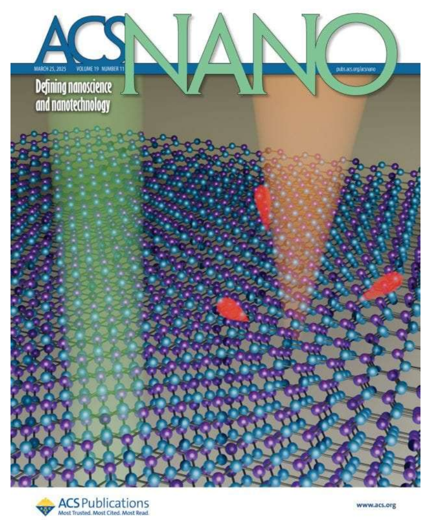

FIGURE 1. Cover art of ACS Nano illustrating optical probing of hBN films exposed to electron beams, as well as thermal annealing and oxygen plasma treatments.[3]

The presented work is focused directly on the two-prong approach (*Thrust A* and *Thrust B*) funded by our BES QIS project (PI: Drndic) for advancing *in situ* and *operando* AC-TEM while investigating new phenomena in 2D hexagonal boron nitride (BN) and 2D ferromagnetic materials. These emerging 2D materials are interesting for the investigation of new fundamental phenomena including single-photo emission (hBN), studies and engineering of single-atom defects, room temperature ferromagnetism and nanostructure applications in quantum nanoelectronics, single-photon emission phenomena, spintronics and quantum information processing. We prepared hBN samples compatible with confocal photoluminescence (PL) microscopy, transmission electron microscopy (TEM), and atomic-force microscopy (AFM), and we used those techniques to quantitatively investigate correlations between fluorescent emission, flake morphology, and surface residue. We find that the microscopy techniques themselves induce changes in hBN's optical activity and residue morphology: PL measurements induce photobleaching, whereas TEM measurements alter surface residue and emission characteristics. We also studied the effects of annealing and oxygen plasma cleaning on the structure and optical activity of hBN.

We investigated two conventional methods for the transfer of 2D materials: viscoelastic stamping with polydimethylsiloxane (PDMS) and a heated transfer with poly bis-A carbonate (PC).[2] We characterized the resulting transfers using atomic force microscopy (AFM), aberration-corrected scanning transmission electron microscopy (AC-STEM) and electron energy loss spectroscopy (EELS). Both transfer methods yield flakes with significant and comparable residue.

We also present results on Iron Germanium Telluride (FGT), a magnetic van der Waals (vdW) material that has attracted significant interest due to its strong out-of-plane uniaxial magnetic anisotropy and high Curie temperature ($T_c \sim 230K$) relative to other vdW magnets. These properties make FGT an exciting candidate for spintronics applications. We probed the effects of geometric confinement on magnetic spin texture in FGT using low-temperature Lorentz transmission electron microscopy (LTEM), a powerful technique which enables direct observation of magnetic domains down to the nanometer scale. Our ongoing work elucidates the micromagnetic properties of geometrically confined FGT – a key step towards scalable spintronics technology.

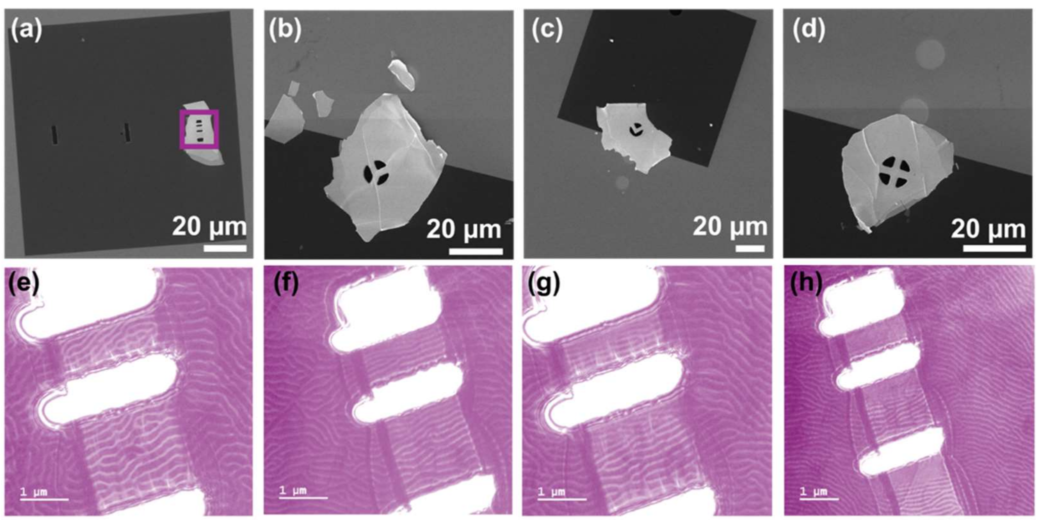

FIGURE 2. Sculpted Fe_3GeTe_2 flakes and corresponding LTEM data. (a-d) Scanning electron microscope images of four devices that have been patterned using focused ion beam milling. (e-h) LTEM temperature series. All images were obtained with a 0.1043T field, 2 mm defocus, and -30° tilt. Panels e, f, g, and h were obtained at 95 K, 133 K, 149 K, and 193 K, respectively.

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Multiscale Quantum and Classical Microscopy for Superconducting Quantum Systems

Award Number: ERKCK51

Benjamin Lawrie, Gábor Halász, Chengyun Hua, Petro Maksymovych, Matt Brahlek, Josh Damron, Sang Yong Song (pd), Yueh-Chun Wu (pd), and Nirjhar Sarkar (pd)

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Abstract: The overarching goal of this project is to discover how charge and spin fluctuations and controlled disorder impact classical and quantum order parameters in magnetic and superconducting materials. In pursuit of this goal, two objectives are defined: (1) understand how local spin fluctuations become correlated across all length scales in magnetic systems under both equilibrium and out-of-equilibrium conditions and (2) unravel the interplay between superconductivity, magnetism, and disorder in unconventional (e.g., Fe-based) superconductors from atomic to millimeter length scales. We will address these objectives with a unique combination of spin-based quantum sensing, variable temperature quantum optical microscopies, nanoscale tunable Josephson junction and Andreev spectroscopies, and operando characterization of superconducting quantum devices. The demonstrated quantum advantage offered by our quantum microscopies will result in an unprecedented understanding of microscopic interactions in functional devices patterned from emerging quantum materials.

Over the past three years, we have made substantial advances in our understanding of the interactions between quantum sensors and magnetic and superconducting materials across a wide range of length scales. As shown in Fig. 1(a), we provided the first experimental demonstration of atomic-scale tunneling Andreev reflection (TAR) spectroscopy, and in doing so, we provided direct evidence of the sign-changing superconducting order parameter in FeSe and of the suppression of superconductivity along the nematic twin boundary[1]. We showed that incommensurate potentials can be used to manipulate vortex interactions and tune vortex lattice parameters in FeSe[2] as part of a first step toward the control of vortex

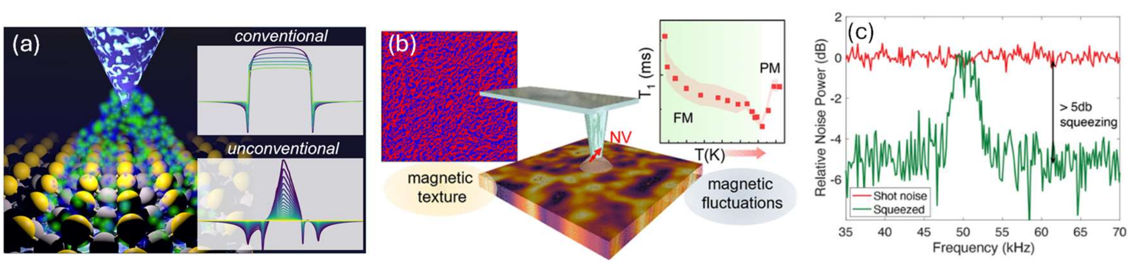

Fig. 1 (a) Schematic illustration of TAR spectroscopy on FeSe illustrating TAR spectra from conventional and unconventional superconducting order parameters[1]. (b) Illustration of nanoscale dc magnetic ordering (left) and temperature dependent NV relaxometry (right) as probes of spin interactions in magnetic thin films[2]. (c) First demonstration of squeezed magnetic circular dichroism measurements with quantum noise reduction 5 dB below the photon shot noise limit[3].

interactions in superconducting quantum devices. As shown in Fig. 1(b), we used spin-based quantum sensors to unveil the universality class of a Sr_2FeReO_6 thin film with a combination of temperature-dependent relaxometric probes of spin fluctuations and nanoscale dc magnetometry measurements[3]. And as shown in Fig. 1(c), we provided the first demonstration of magnetic circular dichroism measurements with a squeezed optical readout field[4], a critical step toward non-perturbative quantum-enhanced squeezed vacuum measurements of Kerr rotation in materials that exhibit spontaneous time reversal symmetry breaking. Finally, we established a theoretical framework for understanding the effects of disorder on spin-triplet superconducting devices[5], and we have recently shown that we can use nanoscale helium-ion patterning to write quantum devices with controlled disorder in iron-based superconducting films.

Each of these quantum sensing modalities addresses a complementary length scale, but until now, each approach has been used in very different temperatures and magnetic fields on very different material systems. We have now established spin-based quantum sensing capabilities that function at temperatures of 50 mK-350K, we have recently shown that we can perform relaxometric probes of spin-defects in hBN in fields of up to 7T with no loss of contrast, and a new MSE-supported 1.4 K STM will allow for TAR spectroscopies to be performed on patterned devices in fields of up to 5 T. By integrating all of these approaches into a comprehensive suite of quantum sensors, we are now able to probe the interplay between superconductivity, magnetism, and disorder in unconventional (e.g., Fe-based) superconductor materials and to understand and control the effects of atomic-to-mesoscale disorder on the behavior of quantum devices patterned from these materials.

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Modeling, probing, and controlling quantum coherence in materials
FWP: SCW1674, DE-SC0020313

Diagnostics to deduce effects of impurities and electron correlations on quantum material systems

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Abstract: In this collaborative QIS project our central aim is to gain a detailed understanding of the fundamental material origins of decoherence in superconducting and semiconducting quantum systems. These material limits originate from defects that are intrinsic to the materials used in fabrication, and also from particle interactions that drive new electronic phases that compete with superconductivity and can affect thermal transport. By combining theoretical efforts with advanced experimental methods, we aim to achieve the following goals: (i) Develop precise theoretical models that link microscopically observed defects to decoherence in quantum devices, (ii) Develop new types of probes that act as powerful diagnostic tools to identify sources of decoherence and eliminate them, (iii) Develop new quantum material and device platforms that exhibit advantages in fabrication and noise reduction, and also create new functional capabilities. Through systematic improvements in quantum materials, this effort will enable new qubit platforms to be created that exhibit much longer coherence times, higher gate fidelities, and improved thermal management.

A main thrust of our efforts has been in developing new scanned probe techniques that can visualize impurities in dielectrics and probe carrier dynamics in materials. We have shown how electrostatic force microscopy (EFM) can be coupled with density functional theory calculations to chemically identify individual near-surface impurities in dielectrics, identifying, for example, those caused by vacancies as well as from surface carbons. We have also advanced scanning tunneling potentiometry (STP) as a tool that can be utilized to directly image carrier flow in materials to quantify impurity scattering and el-el interactions.[1-3] In a third effort, a scanned Josephson junction technique was advanced that has the potential to perform quantum limited microwave absorption measurements at the nanoscale.[4] These measurements utilize high capacitance superconducting SPM tips to probe superconducting surfaces, creating a scannable Josephson junction spectrometer that can probe GHz-THz absorption with extremely high spectral sensitivity an spatial resolution.

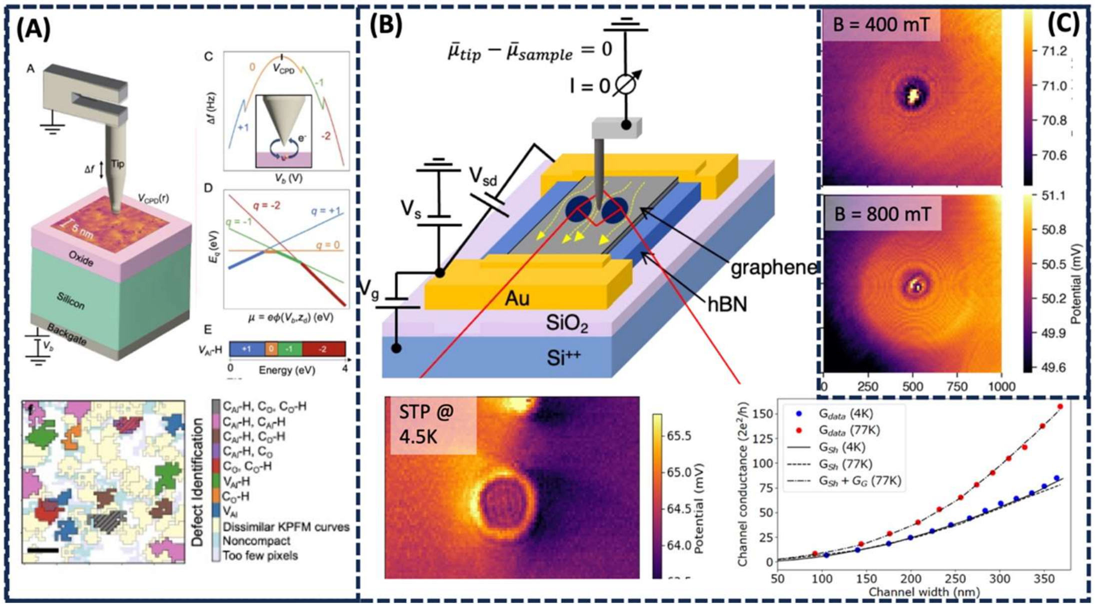

Figure 1: **(A)** Combined electrostatic force microscopy and DFT study localizing and chemically identifying impurities in Al_2O_3. **(B)** Scanning tunneling potentiometry (STP) study of carrier transport around electrostatic barriers in graphene. This work demonstrates how the carrier flow profile evolves as electron-electron interactions increase, driving phase transitions. Figure adapted from Ref. [2]. **(C)** STP study of magnetotransport in graphene near a circular barrier, revealing a transition from Ohmic behavior to quantum Hall transport. Figure adapted from Ref. [3].

A second aspect of this effort is to explore superconducting circuit architectures that allow for direct investigation of defects that drive dechoerence. These experiments include both flux tunable transmons, as well as multi-mode resonators composed of ~20,000 Josephson junctions that act as time domain spectrometers of two-level system dynamics. This latter technique works over a large bandwidth, including the operation frequencies of fluxonium qubits. Combined, these techniques have allowed us to quantify the effect of the junction oxide and deduce the role that liftoff residues play in qubit decoherence. These experiments are being performed in tandem with theoretical efforts that explore the effects of fluctuation-driven noise in superconductors, which can occur in thin superconducting films and narrow constrictions due to heating. [5]

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DNA-Controlled Dye Aggregation – A Path to Create Quantum Entanglement
DE-SC0020089

Molecular Devices to Create Entangled Exciton States and Molecular Dye Polymers for Quantum Entanglement and Quantum Circuits

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Abstract: Our group's high-level goal is to build molecular (dye) aggregate devices that create entangled exciton states at room temperature towards quantum gates and circuits. Our near-term goal is to synthesize oblique dimer dye aggregates and dye aggregate polymers (3-6 dyes in length). The former will be used to establish that electronic quantum beats can be observed in molecular aggregates. The latter will be used as excitonic transmission lines to build excitonic devices that create and control entangled exciton states at room temperature. DNA templates (nanobreadboards) will be used to route the polymer dye aggregate transmission lines to form entanglement devices. *Currently, we are on the verge of two groundbreaking achievements: demonstrating electronic coherence (decoherence time, τ_d, beat frequency, ω_h) in dye aggregates and synthesizing dye polymer aggregates*.

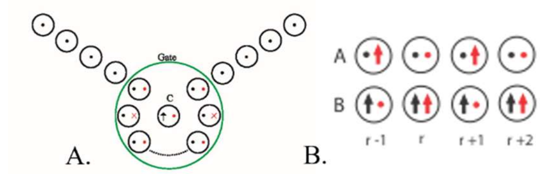

Fig. 1: Excitonic devices comprised of dye aggregate transmission lines that create exciton Bell states. **A**. An exciton transistor. **B**. Controlled phase shifter, a segment of a CNOT gate. Due to the arrangement of μ's and Δd's, excitons cannot hop between transmission lines A and B but can only influence each other through the inter-row two-exciton coupling energy *K*. Note: Black circles represent dyes. Black dots represent μ's orthogonal to the plane of the figure. Black arrows represent μ's lying in the plane of the figure. Red dots represent Δd's pointing out of the plane of page. Red x's are Δd's pointing into the plane of the figure.

We limit our description of studies to those performed by 3 of the 5 highly integrated teams in our research group over the past 2 years. We identified dyes that have large dipole-dipole coupling energy, *J,* based on the transition dipole moment (μ) of each dye, resulting in dye aggregates exhibiting strong exciton delocalization. Theoretical work has been performed on two other understudied parameters fundamentally crucial for creating many-particle entangled states: (1) *K* (exciton-exciton interaction energy), to identify dyes capable of exhibiting large Ks (induced by the difference static dipole, Δd) upon aggregation and (2) tuning the angle (ζ) between μ and Δd within an individual dye to modify J and K in dye aggregates.

Two theoretically explored dye aggregate schemes to produce entangled exciton states were identified capable of producing maximally entangled states (Bell states). The first (**Fig. 1A**) consists of an exciton transistor-like configuration in which the absence or presence (0 or 1) of an exciton on the gate molecule C becomes entangled with whether n excitons (on the source-drain dye aggregate transmission line) remain at the source or are transferred to the drain.[1] The second consists of a CNOT gate consisting of a circuit employing a controlled phase shifter (**Fig. 1B**) and two Haddamard gates[2] which successfully converts a two-exciton product state into a Bell State. In both schemes, J enables excitons to propagate and K gives rise to the entanglement. Both schemes employ two types of dyes: one for which μ and Δd are parallel and one for which they are orthogonal demonstrating the utility of having dyes with differing ζ, μ, and Δd.[3]

To construct the entanglement devices (**Fig. 1**), required are dyes with $J_{m,n} \sim k_bT$, low electronic-vibrational coupling (Huang-Rhys factor [RH]), and tunable ζ, μ, and Δd that can be arranged linearly to form transmission. One such group of dyes are bacteriochlorins (Bchl - found in green and purple bacteria) and chlorins (Chl – found in green plants), of which chlorophyll is a subset. We synthesized synthetic Bchl and Chl monomers with the lowest RH of all dyes we have studied and synthesized) and some tunability of ζ, μ, and Δd.[3, 4] Towards transmission lines, our group is using two approaches – one involves using DNA-templates to form dye aggregates and the other involves using rigid nonconjugating bridges. Here, we focus on the nonconjugating bridge approach. Nonconjugating bridging enables tuning of distance and orientation, thus J, between monomers and imparts greater rigidity to the dye aggregate effectively suppressing detrimental vibrational modes. We have demonstrated a simple click-chemistry approach for nonconjugating bridging of three different synthetic Bchl monomers used to form two different oblique (90° between dye's μ's) heterodimer dye transmission lines.[5] Oblique dimers are quite rare as compared to J- (μ's end-to-end) and H-dimers (μ's cofacial). Dipole-dipole coupling between the two dyes in the oblique dimer results in Davydov splitting of the monomer absorption peak into two new excited states with equal electronic oscillator strengths; thus, they are ideal candidates for the direct measurement of τ_d and ω_h. Our latest effort has resulted in two additional oblique dimers – a third Bchl heterodimer and a Bchl homodimer (**Fig. 2A**), both of which show Davydov splitting with near equivalent and 99% electronic oscillator strengths and $J_{m,n} \sim k_bT$. We are in the process of synthesizing a trimer transmission line (**Fig. 2B**) with near future work focused on synthesizing long polymer aggregates (**Fig. 2C**) and their routing via DNA templated (**Fig. 2D**) nanobreadboards enabling quantum entanglement devices (**Fig. 1**) as well as quantum gate devices and light harvesting systems.

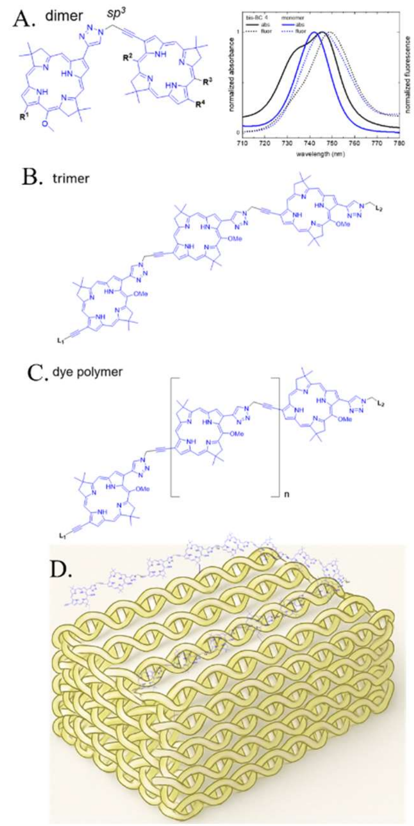

Fig. 2: **A**. Oblique Bchl homodimer and absorbance (& fluorescence) showing Davydov splitting as compared to the monomer. **B**. Trimer Bchl currently being synthesized. **C**. polymer dye aggregate. **D**. routing of a polymer dye aggregate using a DNA brick "nanobreadboard" - 3D routing possible (DNA AI created).

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Direct Observation of Fractional Quantum Hall Quasiparticle Braiding Statistics via Interferometry

DOE QIS DE-SC0020138

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Utilizing recent advances in materials design and anyon interferometry generated under DOE BES QIS funding DE-SC0020138, we report on new set of interferometric and controlled anyon manipulation experiments. Our ultimate goal is to demonstrate the operation of a topologically protected qubit constructed from the non-Abelian fractional quantum Hall state at n=5/2. Our project aims to realize a fundamentally new qubit platform with "built-in" protection from decoherence and provide unambiguous experimental confirmation of a long-sought phase of matter supporting non-Abelian anyons. Here we report on a series of novel experiments in which we first demonstrate the existence and controlled manipulation of Abelian anyons in the fractional quantum Hall regime.

Our most recent results have demonstrated the successful extension of Fabry-Perot interferometry to measure effective charge and braiding statistics at n=2/5, a complex higher order FQHE state with multiple circulating edge modes, effective charge $e^*=1/5e$, and anyonic statistical angle $J_a=4p/5$ [1]. This result provides a necessary steppingstone to investigate interferometry at the non-Abelian n=5/2 state, where the presence of multiple edge modes, small bulk excitation gap, and small effective charge $e^*=1/4e$ will make studies more difficult than at the primary Laughlin state at n=1/3. Fig. 1 shown below displays interference data for the inner mode with charge $e^*=1/5e$ at n=2/5 [1]. We have also recently exploited our device design to demonstrate that the fractionalized edge modes behave as chiral Luttinger liquids as originally predicted by X. G. Wen nearly 40 years ago. Our new data is the first to show a robust tunneling exponent g and proper scaling behavior for the *tunneling of anyons* across a narrow constriction form by a quantum point contact within our interferometer design [2]. Along with our measurements of effective charge and anyonic statistics at n=1/3, measurement of the tunneling exponent g=1/3 completely parameterizes the topological order at n=1/3 and identifies the universality class of the ground state wavefunction (see Fig. 2 below).

Most recently we have designed and implemented a fundamentally new screening quantum heterostructure to interferometric measurements at the fragile non-Abelian n=5/2 state. These new heterostructures place *screening electrons in AlAs quantum wells* to provide the necessary screening while maintaining high mobility and high 2DEG density in the primary quantum well necessary to support the n=5/2. We are optimistic that incisive experiments are now feasible given our recent advances in understanding and control of quantum Hall interferometry, new advances in heterostructure design and device fabrication, and recently added measurement capabilities.

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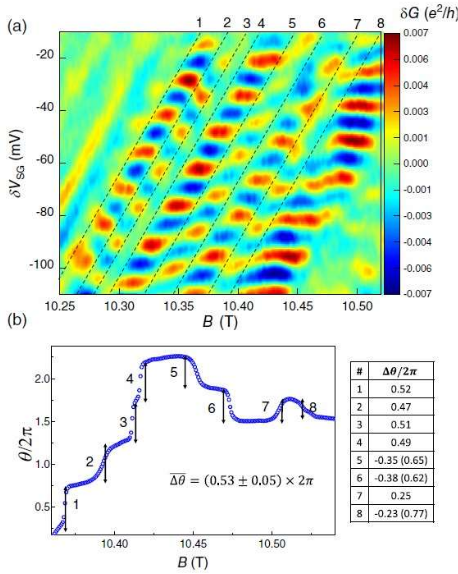
[2] Universal Anyon Tunneling in a Chiral Luttinger Liquid, arXiv: 2502.20551, under review Nat. Phys.)



#	$\Delta\theta/2\pi$
1	0.52
2	0.47
3	0.51
4	0.49
5	-0.35 (0.65)
6	-0.38 (0.62)
7	0.25
8	-0.23 (0.77)

Fig. 1: (a) Conductance versus side gate voltage and magnetic field for the inner mode at ν=2/5 with dashed lines indicating the positions of discrete jumps in phase. (b) Phase extracted via Fourier transform vs. magnetic field. These data indicate an anyonic phase of 4π/5.

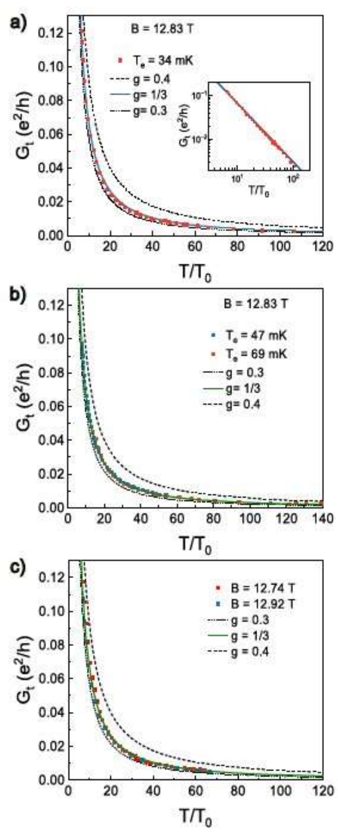


Fig. 2: (a) Scaling of zero bias tunneling conductance at T=34mK and B=12.83 T, at the center of the ν=1/3 plateau. (b) Scaling of zero bias tunneling conductance at T=47mK and T=69mK, at the center of ν=1/3. (c) Scaling of zero bias tunneling conductance at B=12.92T and B=12.74T, +/- 90mT away from the center of the plateau.

Mapping the genome of coherent quantum defects for Quantum Information Science DE-SC0022289

Stark shift effects from first principles in quantum defects

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Abstract: The operation of spin qubits in quantum networks requires long spin but also optical coherence. Optical coherence is limited by the interaction of color centers with electrical fields. Electrical fields around the quantum defects from surfaces or other impurities can shift the emission wavelength (the zero-phonon-line, ZPL) through a Stark shift leading to spectral diffusion. Characterizing the Stark shift of quantum defects is of importance for designing spectrally-stable defects or to understand how to mitigate the spectral diffusion through active control.

While first principles computations have been used to understand and compute many defect properties (e.g., emission wavelength, Debye-Waller, …), the Stark shift of color centers has not been commonly computed and diverging values are available in the literature. We report on a study of the Stark shift effect of the NV center in diamond through a slab technique as well as Modern Theory of Polarization.[1] We show that the two techniques agree, outline a methodology to obtain these values and provide computed values of Stark shift that we estimate as a change in dipole moment between excited and ground state of 2.68 to 2.23 Debye (depending on the level of theory).

Building on our benchmark on the NV center in diamond, we also present some recent results on the T center in silicon. The T center is a doublet color center made of carbon and hydrogen and has emerged as the main spin qubit defect in silicon.[2–4] The defect-bound exciton nature of the excited state of the T center brings interesting question on the physical nature of the Stark shift as well as technical challenges. We have computed this Stark shift for the T center and estimate a change in dipole of around 0.79 Debye and oriented along the C-H direction. We highlight the importance of second order (polarizability) effects in bound excitons and compare our results to recent experimental data.[5]

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Next-Generation Parametrically-Induced Quantum Engineering (PIQUE 2.0)

Award Number: DE-SC0019461

Names of PI, coPI(s), and unpaid collaborators: <u>Archana Kamal</u>[1,2], Leonardo Ranzani[3], Guilhem Ribeill[3], Luke Govia[3], Raymond Simmonds[4], Jose Aumentado[4], Zhihao Xiao[1], Taewan Noh[1,4]**,** Emery Doucet[1]**,** Tristan Brown[1,3], and Tarush Tiwari[1,3]

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Abstract:
This program aims to develop and demonstrate novel functionalities for **creation, control and characterization** of quantum states under a unified framework, which we refer to as PIQUE (Parametrically-Induced QUantum Engineering). Building and extending the results and unique insights generated in first-generation PIQUE (Parametrically-Induced QUantum Engineering) program**,** in the next-generation PIQUE program, we employed strong and tunable parametric interactions for fast, scalable and robust state preparation and control of open quantum systems. We committed to a co-design approach by developing theoretical framework and protocol design closely informed by near-term experimental modalities: specifically, use of linear dissipation and tunable couplings via parametrically-activated sideband interactions between transmons and a lossy oscillator, demonstrated in the first generation of PIQUE program. Beyond PIQUE, the new quantum control functionalities and techniques enabled by this program have already spurred several theoretical and experimental follow-up works by the superconducting circuits community.

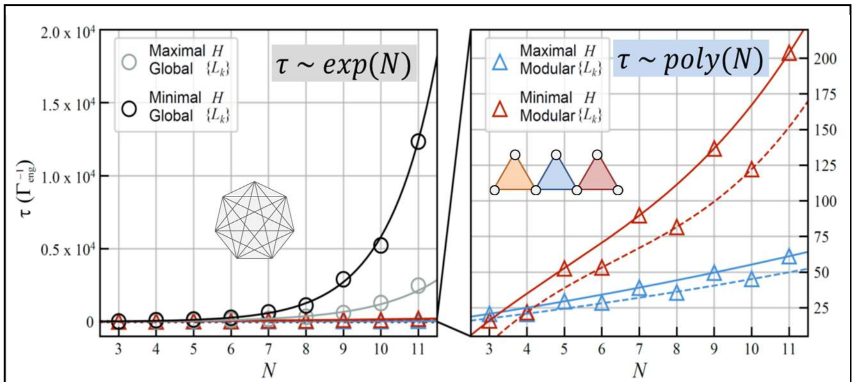

Figure1: Stabilization time vs system size (N) for global (left) vs modular (right) dissipation-engineered protocols for N-qubit W-state entanglement generation. The insets shows the all-to-all vs modular dissipative coupling graph for N=7 protocol

In this program, we developed the first reservoir-engineered entanglement stabilization protocols that exhibit a concurrent scaling of steady-state fidelity and speed allowing a simultaneous reduction of preparation error and time [1]. Crucially, we found that using several overlapping modular dissipators (instead of a single global dissipator) can allow an exponential improvement in stabilization performance, with the stabilization time growing according to some low-degree polynomial in the system size system size (N) for fixed amplitude drives [4] (Figure 1). Remarkably, such modular architectures allow use of bounded-depth interactions and only O(N) resources [i.e. number of dissipative and unitary drives] for stabilizing an N-qubit entangled state making such protocols both "modular" and "scalable". Further, straightforward extensions to high-dimensional entanglement is possible by using parametric drives on successive qudit levels for the stabilization of AME (2, m) state (with 1< m <N).

We have also introduced the paradigm of "parametric circuit-QED" where similar architectures, but now pumped with off-resonant parametric drives can realize large dispersive shifts, which are *in-situ* tunable in sign and magnitude, support exact decoupling or "blind spots" [2], and frequency-selective multi-qubit readout [3]. We developed an ab-initio theoretical framework based on sequential time-dependent Schrieffer-Wolff transformations that enables analytical calculation of dispersive shifts for strong parametric interactions (Figure 2).

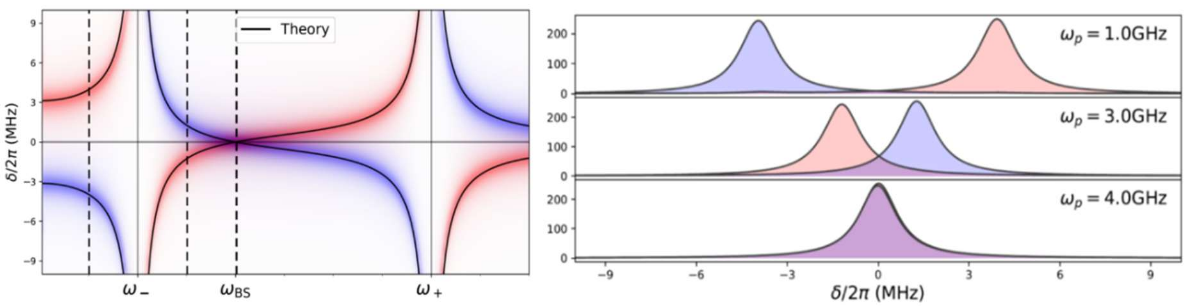

Figure 2: Comparison of analytical results for dispersive shift on resonator obtained using time-dependent Schrieffer-Wolff technique (black curves), with the resonator spectrum obtained from exact numerical simulations of the time-dependent Rabi model (color). Blue (red) correspond to profiles obtained for qubit in ground (excited) state. The line cuts show, negative and zero dispersive shifts tunable via choice of interaction frequency set by the pump ω_p

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Planar Systems for Quantum Information

DE-SC0019481

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Abstract: The scope of this research program is to develop 2D moiré materials as a quantum simulator to implement model Hamiltonians and their phase diagrams and dynamics. Specifically, we aim to (i) develop homogeneous moiré materials with controllable superlattice potential for quantum simulation; (ii) develop methods for continuous tuning of moiré parameters, including length and energy scales, and charge density; (iii) experimentally map the equilibrium quantum phase diagram of model many-body Hamiltonians such as the Hubbard models and compare with theory; and (iv) develop methods to dynamically control the many-body Hamiltonians and investigate non-equilibrium quantum dynamics.

The emergence of 2D moiré materials has opened a new platform to explore quantum phases of matter that arise from electronic topology and correlations. In particular, twisted transition metal dichalcogenides (TMDs) in the AA-stacking structure form a honeycomb moiré lattice with two sublattice sites residing in separate TMD layers. The topmost moiré valence bands, originated from the K- and K'-valley states of the monolayer TMDs, are spin-split and spin-valley locked by a large Ising (out-of-plane) spin-orbit field, which defines the spin quantization axis. It was first predicted by our team member from the continuum model band structure calculations that flat Chern bands with spin/valley-contrasting Chern numbers emerge in small-angle tTMDs as a result of skyrmion-like interlayer hopping[1]. Recent experiments[2], including one from our team, have demonstrated both an integer Chern insulator at moiré band filling factor $\nu = 1$ and fractional Chern insulators at $\nu = 3/5$ and 2/3 in tMoTe2 under zero magnetic field.

In this work, we report transport evidence of a fractional quantum spin Hall (QSH) insulating state in 2.1-degree-twisted bilayer $MoTe_2$, which supports spin-S_z conservation and a series of Landau-level-like flat Chern bands[3]. QSH insulators are 2D electronic materials that have a bulk band gap like an ordinary insulator but have topologically protected pairs of edge modes of opposite chiralities. Previous experiments have found only integer QSH insulators with counter-propagating up-spins and down-spins at each edge leading to a quantized conductance $G_0 = e^2/h$ (with e and h denoting the electron charge and Planck's constant, respectively). We perform both local and local transport measurements (Fig. 1). At filling factor $\nu = 3$ of the moiré valence bands, each edge contributes a conductance 3/2 G_0 with zero anomalous Hall conductivity. Further, at $\nu = 2$, 4, and 6, we observe a single, double and triple QSH insulator with each edge contributing a conductance G_0, $2G_0$ and $3G_0$, respectively. Our results open the possibility of realizing time reversal symmetric non-abelian anyons and other unexpected topological phases in highly tunable moiré materials.

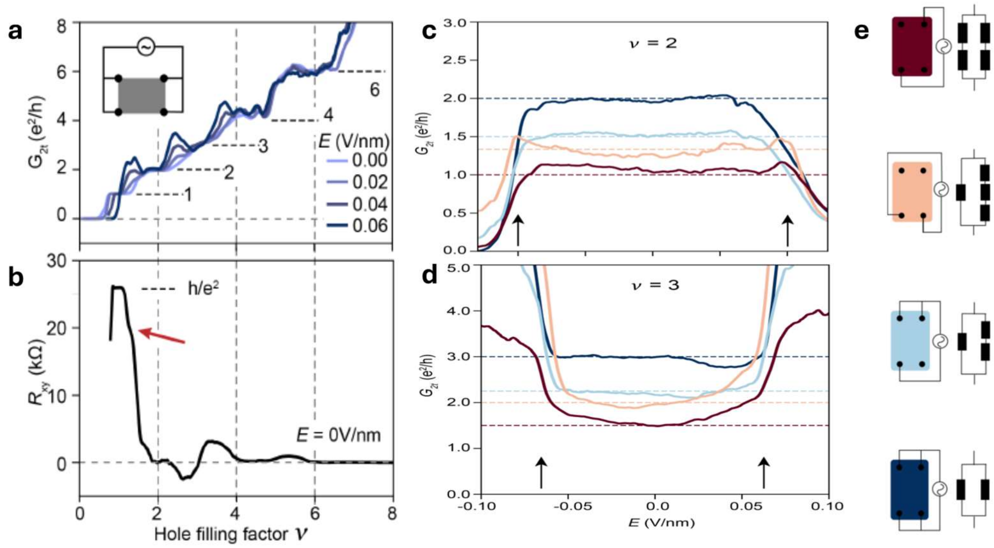

Figure 1. Integer and fractional quantum spin Hall effect in 2.1-degree twisted bilayer $MoTe_2$. **a,b,** Hole filling factor dependence of two-terminal conductance (**a**) and Hall resistance (**b**) at 20 mK for representative electric fields in the layer-hybridized regime. The two-point conductance is quantized at νG_0 while the Hall resistance is nearly zero for ν = 2, 3, 4 and 6. **c,d,** Out-of-plane electric-field dependence of two-terminal conductance for different measurement configurations around ν = 2 (c) and ν = 3 (d). Dashed lines denote the predicted values from the Landauer-Büttiker analysis. **e,** Two-terminal nonlocal measurement configurations (left) and equivalent circuits (right) for helical edge mode transport. The shaded rectangles represent twisted bilayer $MoTe_2$ with four contacts (black dots). Unconnected dots are floated contacts. The shade color matches the line color in **c,d**.

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Multiscale Quantum and Classical Microscopy for Superconducting Quantum Systems
Award Number: ERKCK51

Disorder-enabled control of superconducting devices

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Abstract: The primary goal of this thrust is to understand and control the interplay between disorder and superconductivity on the device scale. The key phenomenon that we aim to control through disorder is vortex tunneling across a narrow superconducting strip in a mesoscopic loop (i.e., Little-Parks) geometry. Theoretically, we study the relevant vortex-tunneling processes via Ginzburg-Landau equations in the one-dimensional limit or the London limit. Experimentally, we fabricate mesoscale superconducting rings of Little-Parks geometry using conventional electron-beam lithography patterning techniques to show integer / fractional magnetoresistance oscillations in Nb and $Pd_3Bi_2Se_2$. An unconventional direct ion-writing fabrication technique has also been developed to obtain cleaner devices.

Over the past three years, we theoretically established universal signatures of unconventional spin-triplet superconductors that are observable on the device scale. Spin-triplet superconductors are interesting from the fundamental perspective and because they host half-quantum vortices that are predicted to bind Majorana zero modes. The signatures we proposed directly reflect the tunneling of such half-quantum vortices, thereby revealing not only the unconventional superconducting state but also the Majorana zero modes that may allow topological quantum computing.

We primarily focused on the Little-Parks effect – a key hallmark of superconductivity that results from the macroscopic quantum coherence of Cooper pairs. Due to fluxoid quantization, the resistance of a mesoscale superconducting ring oscillates as a function of the magnetic flux going through the ring. For a conventional

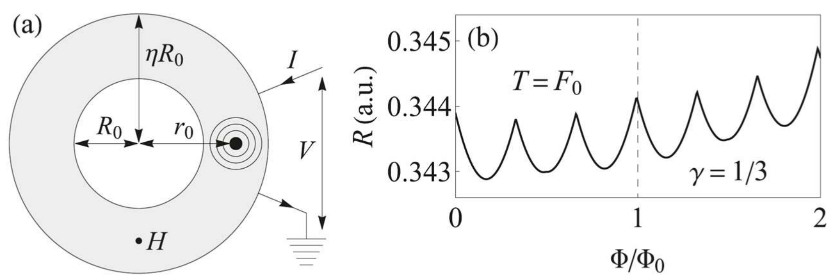

Figure 1: (a) Schematic illustration of the mesoscopic Little-Parks setup with a vortex (bullseye) tunneling across the superconducting ring[1]; the voltage V resulting from a bias current I is measured. (b) Disorder-induced Little-Parks oscillations with fractional periodicity $\Delta\Phi = \Phi_0 / 3$ in the resistance $R = V / I$ against the flux Φ through the loop.

(spin-singlet, *s*-wave) superconductor, the Little-Parks oscillation periodicity is given by the flux quantum $\Phi_0 = h / 2e$, and the minima of the resistance correspond to integer multiples of Φ_0. Unconventional superconductors may exhibit different kinds of Little-Parks oscillations. For example, in gapless superconductors with *d*-wave pairing, the Little-Parks oscillations acquire an enlarged periodicity $2\Phi_0$, while polycrystalline spin-triplet superconductors feature shifted Little-Parks oscillations with the resistance minima corresponding to half-integer multiples of Φ_0.

In our theoretical work[1], we showed that, for a range of temperatures in the presence of disorder, spin-triplet superconductors exhibit unconventional Little-Parks oscillations with an emergent fractional periodicity Φ_0 / n, where the integer $n \geq 3$ is entirely determined by the ratio of the spin and charge superfluid densities.

For a different temperature range studied in another theoretical work[2], it was found that the Little-Parks oscillation periodicity remains Φ_0, but each maximum is split into two distinct peaks, with the peak width and peak separation following distinctive scaling laws as a function of temperature. These novel features in the Little-Parks effect all reflect the existence of half-quantum vortices that are exclusive to spin-triplet superconductors. Current efforts[3] aim to understand how the same half-quantum vortices may be revealed in color-center relaxometry, which ties this thrust to other parts of the project.

Experimental challenges in Little-Parks measurements lie in fabricating superconducting nano-rings without introducing significant sidewall damage that suppresses superconductivity, broadens transitions, and introduces uncontrolled vortex leakage paths, resulting in noisy oscillations with poor signal-to-noise ratios. This effect was observed during our experimental work on $Pd_3Bi_2Se_2$ and Nb. Therefore, we developed focused ion beam (FIB) patterning techniques using helium/xenon ions with sub-2 nm resolution. This technique selectively converts superconducting regions to normal states through controlled ion implantation rather than material removal, preserving pristine current paths while creating well-defined boundaries. FIB patterning has successfully fabricated high-quality Josephson junctions in various superconductors and could potentially enable reproducible Little-Parks devices with enhanced signal quality. This approach also enables engineering of 0-π junctions for topological qubits and SQUIDs as well as superconducting nanowire single-photon detectors, an opportunity that we are just beginning to explore.

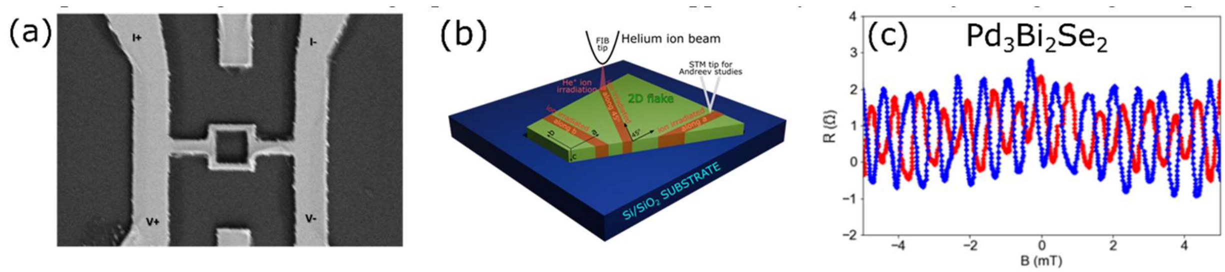

Figure 2: **(a)** Superconducting Nb nano-ring for Little-Parks measurements from standard electron-beam lithography and reactive ion etching. **(b)** Schematic sketch of FIB patterning with helium/xenon ions. **(c)** Little-Parks oscillations in $Pd_3Bi_2Se_2$.

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Project Title: Optical Information Processing Through Jointly-Optimized Diffractive Surfaces and Electronic Neural Networks

Award Number: DE-SC0023088

Name of PI and unpaid collaborators: **Md Sadman Sakib Rahman**, Tianyi Gan, Emir Arda Deger, **Çağatay Işıl**, Mona Jarrahi, and **Aydogan Ozcan**

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Abstract: In this project, we are exploring the science at the intersection of AI, materials science and optics/photonics to harness the diffraction of light through structured materials composed of trainable surfaces (forming a 3D structured volume) that collectively execute a desired computational task, not achievable with standard optical design principles. These diffractive systems form optical processors that are passive (no external power), and operate at the speed of light, calculating the desired solution as the input light propagates through a thin physical diffractive system trained for a specific task. Benefiting from the scalability and parallelism of optics, diffractive processors and materials that are trained using, e.g., deep learning, create task-specific computational imagers, sensors and machine vision systems with various new capabilities. In this project, we have been investigating the information processing capacity of trainable diffractive surfaces, to inquire a deeper understanding of the limits of their computational capabilities [1-3].

Optical information processors created by trainable diffractive surfaces that are used as the front-end or back-end of electronic neural networks form hybrid information processing units. One of our accomplishments [1] on the topic of jointly optimized collaborations between electronic neural networks and diffractive surfaces was on learning diffractive optical communication of information around arbitrary opaque occlusions. We demonstrated an optical architecture for directly communicating optical information of interest around zero-transmittance occlusions using electronic encoding at the transmitter and all-optical diffractive decoding at the receiver [1]. In our scheme, an electronic neural network, trained in unison with an all-optical diffractive decoder, encodes the message of interest to effectively bypass the opaque occlusion and be decoded at the receiver by an all-optical decoder, using passive diffraction through thin structured layers. This all-optical decoding is performed on the encoded wavefront that carries the optical information or the message of interest, after its obstruction by an arbitrarily shaped opaque occlusion. The diffractive decoder processes the secondary waves scattered through the edges of the opaque occlusion using a passive, smart material comprised of successive spatially engineered surfaces, and performs the reconstruction of the hidden information at the speed of light propagation through a thin diffractive volume that axially spans < 100 wavelengths. We showed that this combination of electronic encoding and all-optical decoding is capable of direct optical communication of any arbitrary message between the transmitter and the receiver even when the opaque occlusion body entirely blocks the transmitter's field-of-view. We also reported an experimental demonstration of this scheme using a 3D-printed diffractive decoder that operates at the THz spectrum [1]. We demonstrated that this scheme could be configured to be misalignment-resilient as well as highly power efficient, reaching output diffraction efficiencies of >50%. We also showed that our encoder/decoder framework can be jointly trained to be resilient against unknown, random dynamic changes in the occlusion size/shape, without the need to retrain the encoder or the decoder. This makes the presented concept dynamic and easy to adapt to external and uncontrolled/unknown changes that might happen between the transmitter & receiver apertures.

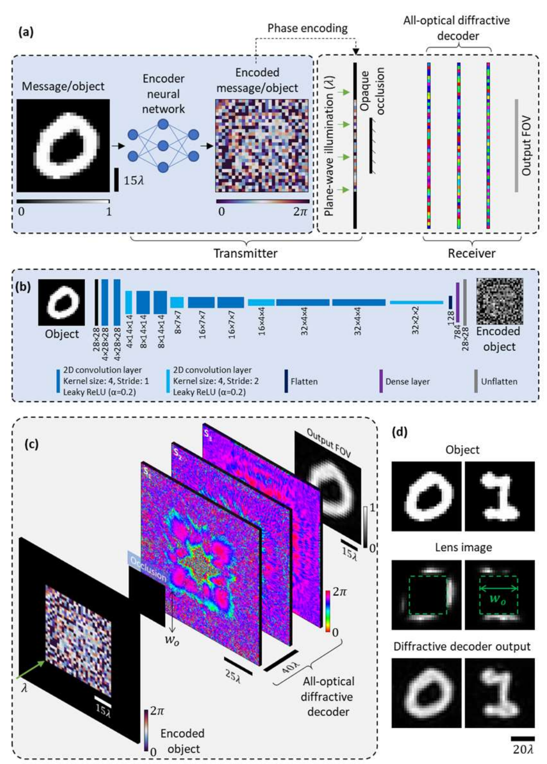

Fig. 1: Schematic of the optical communication framework around fully opaque occlusions using electronic encoding and diffractive all-optical decoding [1]. (a) An electronic neural network encoder and an all-optical diffractive decoder are trained jointly for communicating around an opaque occlusion. For a message/object to be transmitted, the electronic encoder outputs a coded 2D phase pattern, which is imparted onto a plane wave at the transmitter aperture. The phase-encoded wave, after being obstructed and scattered by the fully opaque occlusion, travels to the receiver, where the diffractive decoder all-optically processes the encoded information to reproduce the message on its output field. (b) The architecture used for the convolutional neural network (CNN) encoder throughout this work. (c) Visualization of different processes, such as the obstruction of the transmitted phase-encoded wave by the occlusion of width w_o and the subsequent all-optical decoding performed by the diffractive decoder. The diffractive decoder comprises L surfaces $(S_1, \cdots, S_L)$ with phase-only diffractive features. In this figure, $L = 3$ is illustrated as an example. (d) Comparison of the encoding-decoding scheme against conventional lens-based imaging.

As another exciting accomplishment [3] on the topic of collaborations between electronic neural networks and diffractive surfaces, we demonstrated a jointly optimized electronic encoder neural network and an all-optical diffractive decoder model for optical information transfer through random unknown diffusers. This electronic-optical design is composed of a CNN that encodes the input image information of interest (to be transmitted) into a 2D phase pattern, like an encrypted code, which is all-optically decoded by a jointly trained/optimized diffractive processor that reconstructs the image of the input information at its output plane, despite the presence of random unknown phase diffusers that are constantly changing/evolving. We experimentally validated the success of this approach using terahertz radiation and a 3D-printed diffractive network [3], demonstrating the feasibility of the diffractive decoder with electronic encoding for optical information transmission through random unknown diffusers. The optical decoder of this hybrid model can be scaled physically (through expansion or shrinkage) to operate at different parts of the spectrum without retraining its diffractive features.

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Multimodal Quantum Material Control Monitored with Ultrafast Coherent X-Rays Award number FWP LANLE1NT (connected to DE-SC0019126)

Enhanced and twisted nonlinear optical responses in quantum nanomaterials

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Abstract: In recent years, explorations of the nonlinear optical (NLO) responses of van der Waals (vdW) crystals have proven to be integral to the study of their fundamental material symmetries, while also providing a platform to revolutionize nonlinear optoelectronic and photonic devices. Here, we will discuss our recent efforts to probe the NLO properties of an emerging class of polar vdW semiconductors and extend vdW nonlinear optics to include the often-overlooked spatial degree of freedom of light. First, we will highlight our studies of bismuth telluro-halide crystals [1], where we observe giant second-order optical nonlinearity enhancements that are highly correlated with the orientation of their permanent dipole moments and domain character (Fig. 1). We will then turn our focus to nonlinear frequency-mixing processes in vdW crystals driven by twisted light [2]. Here, we demonstrate the harmonic scaling of orbital angular momentum and the free tuning of the wavelength, topological charge, and radial index of vortex light-fields, both supported by atomically thin semiconductors (Fig. 2). Our work points to new materials and structured-illumination-based methods for future vdW-based classical and quantum communication nanotechnologies.

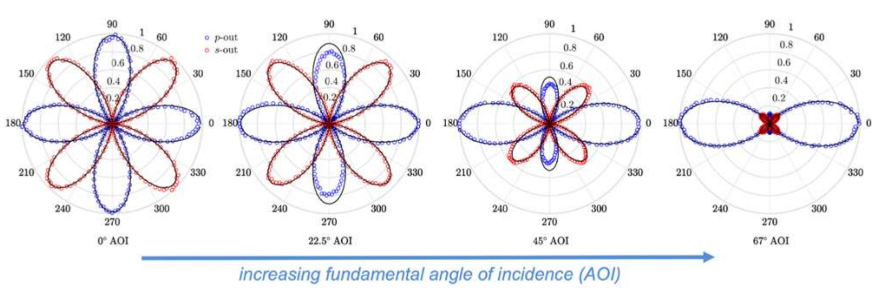

Fig. 1. p- (blue dots) and s-polarized (red dots) second harmonic generation output from BiTeBr, as a function of the linear input polarization angle with respect to the crystal's armchair direction, for increasing angles of incidence with respect to the *ab*-surface normal direction. The data shows significant enhancement of the nonlinear optical output and alternation of the nonlinear output's fundamental polarization dependence as the driving field's polarization becomes increasingly aligned with the *c*-axis oriented permanent dipole moment through the activation of the crystal's d_{33} nonlinear susceptibility tensor element.

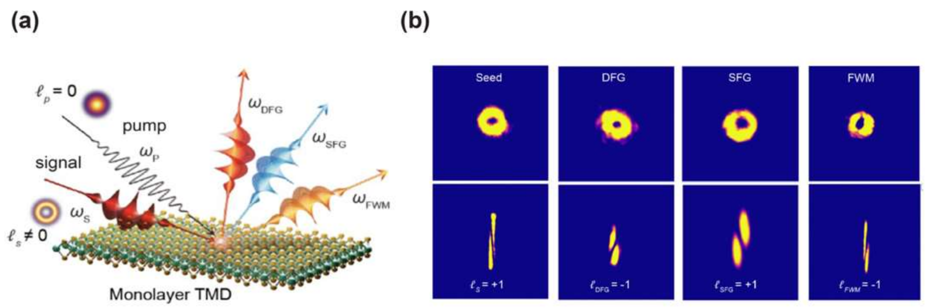

Fig. 2. (a) Schematic of structured difference frequency generation (DFG), sum frequency generation (SFG), and four-wave mixing (FWM) nonlinear optical responses from a monolayer transition metal dichalcogenide (TMD) driven with a Gaussian pump beam and Laguerre-Gaussian vortex seed, the latter having non-zero quantized orbital angular momentum indexed by the topological charge ℓ. (b) Real-space images (top panels) of the donut shaped intensity profile of the vortex seed ($\ell_{\mathrm{S}} = +1$, $\hbar\omega_{\mathrm{S}} = 1.18$ eV) and resulting DFG ($\ell_{\mathrm{DFG}} = -1$, $\hbar\omega_{\mathrm{DFG}} = 1.92$ eV), SFG ($\ell_{\mathrm{SFG}} = +1$, $\hbar\omega_{\mathrm{SFG}} = 2.81$ eV) and FWM ($\ell_{\mathrm{FWM}} = -1$, $\hbar\omega_{\mathrm{FWM}} = 1.92$ eV) outputs. The lower panels show the corresponding momentum space maps imaged at the focal plane of a cylindrical lens. For all data, the pump beam is always Gaussian ($\ell_{\mathrm{P}} = 0$, $\hbar\omega_{\mathrm{P}}^{\mathrm{DFG}} = 3.1$ eV, $\hbar\omega_{\mathrm{P}}^{\mathrm{SFG}} = 1.63$ eV, $\hbar\omega_{\mathrm{P}}^{\mathrm{FWM}} = 1.55$ eV).

[1] K. W. C. Kwock and T. Norden *et al.*, manuscript in preparation – to be credited to LANLE1NT

[2] T. Norden *et al.*, under review at *ACS Nano*

Poster Session I

Quantum Computing Approaches for Correlated Multi-Orbital Materials Simulations

FWP Number: A19-510-100

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1. Ames National Laboratory 2. Iowa State University 3. Saarland University, Germany

Abstract: This project leverages and contributes to an expanding toolbox of quantum algorithms to carry out accurate simulations of correlated multi-orbital quantum materials. Noisy intermediate-scale quantum computers represent the first generation of quantum computing hardware that can be programmed remotely through the cloud and applied to a wide range of computational tasks. Although such devices are currently too error-prone to outperform classical computers in simulations of static and early-time dynamical properties of correlated materials, rapid progress in hardware improvements, algorithm development, and problem reformulation indicates that practical quantum advantage may soon become attainable.

The central goal of this project is to achieve practical quantum advantage by applying quantum algorithms to calculate static, dynamic, and finite-temperature properties of correlated multi-orbital quantum materials, focusing on layered transition metal oxide and rare-earth-bearing half-Heusler compounds. To realize this, the work adopts a hybrid quantum-classical Gutzwiller variational embedding (GQCE) approach that employs quantum computers as impurity solvers. It also directly applies quantum algorithms to study complex quantum spin and fermionic models, which both provide steppingstones for these algorithms' applications within the Gutzwiller approach and provide important reference points for the Gutzwiller calculations of itinerant electron systems in the strong coupling limit. Calculations will be carried out on IBM and Quantinuum quantum processing units (QPUs).

To achieve the goal of ground state simulations of multi-orbital correlated materials, we first developed and benchmarked best practices for ground-state preparation of the d-sub-shell embedded impurity models using adaptive variational quantum algorithms, a crucial step in GQCE simulations [1]. These adaptive methods enable significant circuit compression compared to standard variational quantum eigensolvers. Ground-state energies were measured on both IBM and Quantinuum quantum hardware, achieving a relative error of 0.7% using a combination of zero-noise extrapolation and symmetry-based post-selection. To further enhance circuit expressivity, trainability, and noise resilience while optimizing quantum–classical resource balance, we integrated variational quantum circuits with a quantum subspace expansion [2]. We demonstrated that coordinating the number of layers in the Hamiltonian variational ansatz with the Krylov subspace expansion order significantly improves performance. Benchmark simulations of ground-state energies for one- and two-dimensional mixed-field Ising spin models were performed on noisy simulators and IBM QPUs, incorporating probabilistic error reduction techniques. Additionally, we improved the automatic ansatz generation procedure used in adaptive algorithms to reduce circuit depth and proposed the integration of tensor-network methods to reduce quantum resource requirements [3]. These combined achievements will allow us to extend simulations to full-shell d- and f-embedded impurity models, thereby enabling GQCE ground-state calculations of transition-metal and rare-earth compounds.

To advance simulations of dynamical properties of quantum systems, we integrated the adaptive variational quantum imaginary- and real-time evolution algorithms (AVQITE, AVQDS) for ground state response-function calculations, improving circuit compression by leveraging degrees of freedom in the entire ansatz [4]. Specifically, adaptive variational quantum algorithms were developed to measure real-time single-particle Green's functions and nonlinear susceptibilities. Accurate benchmark calculations are showcased for fermionic Hubbard models, molecules, and spin systems. Compared to existing methods, our adaptive algorithms yield significantly shallower quantum circuits. The next step is to combine this approach with a real-time generalization of GQCE, enabling response function simulations of complex materials. This will support direct comparison with experimental probes, including ARPES, neutron scattering and 2D coherent spectroscopy.

To address the goal of including finite-temperature effects in correlated quantum materials simulations, we combined AVQITE with classical minimally entangled typical thermal states (METTS) sampling, resulting in the AVQMETTS algorithm [5]. The hybrid approach enables calculations of equilibrium properties at finite temperature. Using local spin models, we demonstrated approximately linear system-size scaling of quantum circuit complexity at fixed temperature. We also simulated the magnetic field dependence of the phase transition temperature in a two-dimensional transverse-field Ising model and analyzed the impact of hardware noise on AVQMETTS results. Moving forward, we will integrate this approach with the imaginary-time generalization of GQCE to enable finite-temperature simulations of correlated materials.

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Scalable quantum photonic materials based on precision doped diamond nanostructures

DE-SC0025295
Focused Ion Beam Implantation for Diamond Color Center Fabrication

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Color centers in diamond are gaining recognition as powerful tools for quantum information science (QIS), particularly in quantum communication and sensing applications. These atomic-scale defects exhibit unique optical and spin properties, making them ideal candidates for integration into quantum devices [1]. However, a major challenge lies in the deterministic fabrication of color centers, as precise control over their spatial placement and density is essential for device functionality. Focused ion beam (FIB) technology, especially when combined with liquid metal alloy ion sources (LMAIS), offers a promising solution to overcome these challenges and enable scalable fabrication of color centers, which we have addressed as part of previous projects [2, 3]. This part of our project focuses on the fabrication of tin-vacancy (SnV) centers in diamond using FIB implantation of tin (Sn) ions generated by a AuSn alloy, as well as the development of a sodium (Na) source for creating Na-based color centers.

SnV centers in diamond have recently attracted attention due to their ability to operate at elevated temperatures compared to lighter group-IV-based color centers. Additionally, SnV centers exhibit high quantum yield and promising optical properties, including narrow linewidths and high photostability, making them suitable for QIS applications. Like other group-IV-based color centers, SnV centers benefit from their high symmetry, which reduces spectral diffusion and enhances coherence.

Sodium-based color centers are theoretically predicted to outperform group-IV-based color centers in certain aspects. For example, they are expected to exhibit a higher Debye-Waller factor, and depending on their charge state, they may possess larger spin than SnV centers. These characteristics make Na-based color centers a promising candidate for next-generation quantum devices.

The deterministic creation of color centers at specific locations remains a bottleneck for their integration into quantum devices. Typically color centers are fabricated either through broad-beam ion implantation and annealing or by incorporating impurity atoms during crystal growth, in both cases resulting in poor spatial control. In contrast, FIB technology enables the direct implantation of desired impurity atoms with nanoscale precision, typically on the order of tens of nanometers. This level of accuracy is both critical and sufficient for integrating color centers into photonic structures, such as waveguides and cavities, which are essential for fabricating quantum devices. Liquid metal alloy ion sources (LMAIS) are particularly suited for QIS applications with FIB due to their ability to emit a variety of ion species with isotopic resolution. This versatility makes LMAIS an ideal choice for creating color centers with high spatial precision.

As part of this project we have FIB implanted high-purity diamond with 117-Sn (nuclear spin-1/2 isotope) and 120-Sn (nuclear spin-0 isotope) and performed activation anneals, with the PI currently working on characterization of the implanted sample.

As part of our planned effort to study Na-based color centers in diamond, we are developing a Na FIB source. Specifically, we aim to create both sodium substitutional (Na_C) and sodium-vacancy (NaV) centers through different annealing recipes. Once a FIB source is available, the ability to place Na_C and NaV centers with nanoscale precision using FIB will enable experiments to explore color-center cavity interactions, a critical step for integrating Na-based color centers into quantum devices.

Developing a stable Na source for FIB implantation presents unique challenges. Reports in the literature describe the use of pure-metal Na and NaLi sources, but both are prone to oxidation, making it difficult to achieve the stability required for FIB implantation. Our initial approach involved using a AuSi eutectic alloy with Na added as an impurity. While the source emitted ions from Au and Si, no Na emission was observed. Sodium is expected to be a prolific ion due to its low first ionization energy (495 kJ/mol), but mass spectra revealed no Na emission, shown in Figure 1. Sodium's high vapor pressure likely causes it to preferentially boil off during source preparation.

To address the evaporation issue, we identified $Na_{62}Pb_{38}$ as a suitable alternative source material due to its lower melting point of 301°C and eutectic-like behavior. However, our initial attempt at creating a NaPb source resulted in rapid boil-off of the material. Further investigation revealed that the alloy was overheated during source preparation. While the AuSi eutectic melts at 380°C, our standard fabrication procedure heats the material to 800°C to ensure full melting of the AuSi boat. This excess heat likely caused the NaPb material to boil, as the vapor pressures of Na and Pb are lower than those of Au and Si.

To prevent overheating, we developed a thermometry system to monitor and control the temperature of the NaPb crucible during source preparation. Using this system, we were able to fabricate a filament, however upon generating emission from the filament, the alloy again evaporated, ascribed to electron self-heating of the filament during emission.

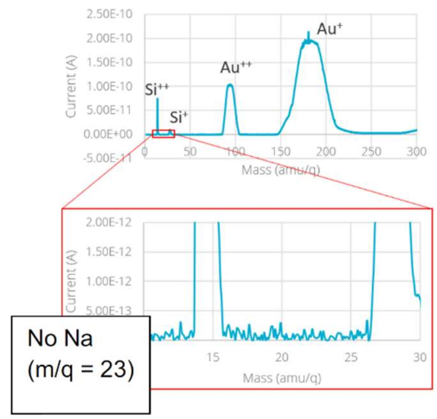

Figure 4 Mass spectrum of AuSiNa source.

FIB technology, combined with LMAIS offers a powerful solution for the deterministic fabrication of color centers. Our efforts to develop a Na source for FIB implantation represent a critical step toward studying and integrating Na-based color centers into quantum devices. While challenges remain, we have made progress with alternative materials like NaPb.

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Project Title: Holographic quantum simulation of strongly correlated electron systems

Award Number: DE-SC0022102
Names of PI, coPI(s), and unpaid collaborators**:**

PI: Shyam Shankar, University of Texas at Austin; Co-PI: Andrew Potter, University of British Columbia; Postdoctoral associates: Joseph Sullivan, University of British Columbia; Josiah Cochran, University of Texas at Austin; Graduate students: Ameya Riswadkar, Jian Jun Liu, Aikaterini Kargioti, Theodore Shaw, University of Texas at Austin; Yuxuan Zhang, University of Texas at Austin and University of Toronto; Travis Hosack, University of British Columbia; Daoheng Niu, University of Texas at Austin; Undergraduate students: Shahin Jahabani, University of Texas at Austin; Alice Xiong, University of British Columbia; and Noah Mugan, University of Texas at Austin

Abstract: Quantum simulations of many-body systems in materials science and chemistry are promising application areas for quantum computers. However, the limited scale and coherence of near-term quantum processors pose a significant obstacle to realizing this potential. In this project, we have been theoretically and experimentally exploring a recently developed method for implementing quantum operations to sequentially build up a matrix product state (MPS) representation of a many-body quantum state. This approach can be used to demonstrate quantum advantage over classical simulations by efficiently using the resources of moderate-size, noisy quantum processors. We will present a theoretical and experimental study of how to employ this sequential quantum simulation algorithm to simulate the ground state energy of a highly entangled many-body spin chain.

First, we will describe how a single-mode circuit quantum electrodynamics (cQED) device, consisting of

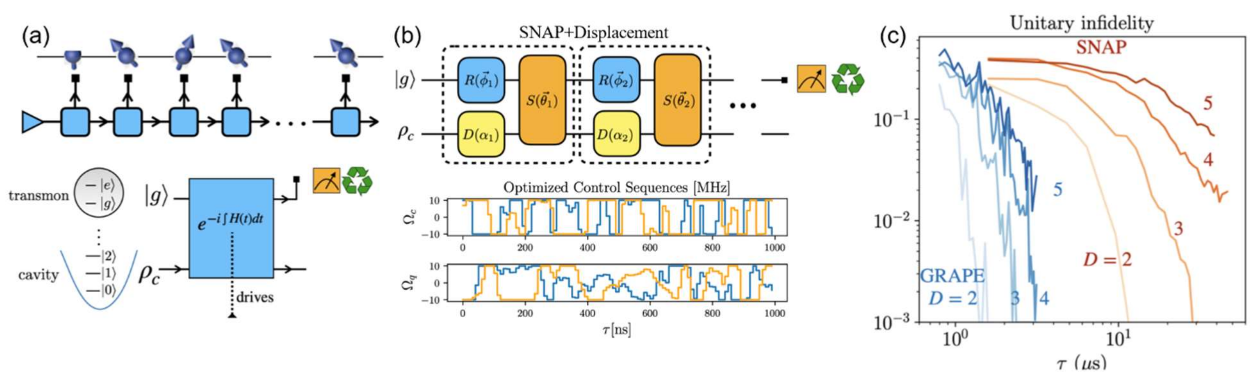

Figure 5. (a) Graphical representation of a matrix product state for a spin chain (top) and its implementation as a sequential circuit with qubit reuse (b) Synthesis of time-dependent Hamiltonian with SNAP gates or GRAPE pulse-level control. (c) Infidelity for synthesizing a unitary representing various bond-dimension D approximations of a critical Ising chain ground state.

a transmon qubit coupled to a long-lived cavity mode, can be used to implement sequential quantum simulation of a spin chain [1]. The approach, described in Figure 1, employs the transmon qubit to read out the state of each spin in the chain and exploits the large state space of the cavity as a quantum memory encoding inter-site correlations and entanglement. We numerically compared analog pulse-level control generated by Gradient Ascent Pulse Engineering (GRAPE) to digital control based on Selective

Number Arbitrary Phase (SNAP) gates. We find that analog control can accurately prepare the MPS representation of a quantum-critical spin chain in less time, thus reducing infidelity due to decoherence.

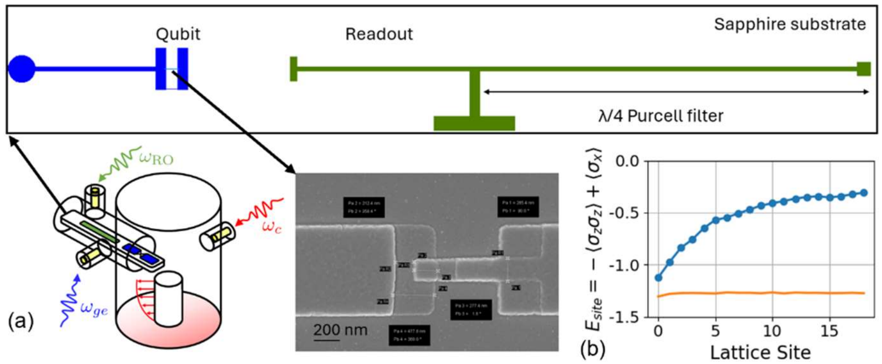

Figure 6. (a) cQED hardware consisting of a long-lived superconducting cavity (red), transmon qubit (blue) and readout (green). Qubit nonlinearity is realized with a Josephson junction. (b) Measured ground state energy (blue) compared to ideal calculation (orange).

Next, we will describe our experimental implementation of the sequential simulation protocol with superconducting quantum hardware, shown in Figure 2. We found experimentally that GRAPE based analog control is sensitive to miscalibration resulting in significant error in the measured ground state energy. To address this hurdle, we will present on progress towards realizing sequential simulation using SNAP gates.

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Integrated Materials Platforms for Topological Quantum Computing Devices

DE-SC0019274

Abstract Title: Modeling and testing of materials and devices for topological and spin qubits

Vlad Pribiag[1], **Sergey Frolov**[2], **Chris Palmstrøm**[3], **Noa Marom**[4], **Kunal Mukherjee**[5], Shixiong Zhang[6], Tomasz Story[7], Moïra Hocevar[8], and Vladimir Strocov[9]

1. University of Minnesota, 2. University of Pittsburgh, 3. University of California Santa Barbara, 4. Carnegie Mellon University, 5. Stanford University, 6. Indiana University Bloomington, 7. Institute of Physics, Polish Academy of Sciences, 8. CNRS Grenoble, 9. Swiss Light Source, Paul Scherrer Institute

Abstract: A significant challenge towards developing robust quantum computing systems with large numbers of logical qubits is rapid decoherence. A promising approach to addressing this challenge is based on the concept of fault-tolerant quantum computing architectures that leverage topological quantum states such as Majorana zero-modes (MZM). Substantial progress toward realizing and detecting topological states has been enabled by quantum devices which combine semiconductors with superconductors due to the combination of processing-friendly materials that offer high spin-orbit coupling, large g-factors, ballistic transport, and the ability to create transparent interfaces with various superconductors. However, further breakthroughs leading to topological quantum computation require next-generation quantum nanostructures that offer substantial improvement in the electronic purity together with optimal coupling across the superconductor-semiconductor interface. *Here we discuss our recent results under this project, focusing on first-principles modeling of interfaces, simulation of device properties and experimental testing of IV-VI nanowire materials for spin qubits.* This work is a subset of our larger effort supported by this grant, which combines quantum device fabrication and characterization with materials synthesis and data interpretation informed by ab initio simulations and machine learning to develop robust, tunable and scalable platforms for the manipulation of quantum states.

(i) Modeling MZM materials: We have employed an array of numerical methods, including DFT-U and micromagnetic simulations, as well as tight-binding modeling, to explore the question of materials most suitable for the realization of MZMs in hybrid superconductor-semiconductor nanowire systems. Using DFT-U, we have explored the potential of two-shell nanowire architectures, where a core of a III-V semiconductor is first passivated by a shell of a larger bandgap II-VI semiconductor. This shell serves as a tunnel barrier to the outer shell of a metallic superconductor. We have explored the combinations of InSb-CdTe-Al, as well as InAs-ZnTe-Al and InAs-CdSe-Al which happen to be lattice matched [1,2]. We identified that their interlayer of group II-VI semiconductor cannot exceed the thickness of 5-10 monolayers and established the band alignment. The results for individual bulk layers were compared to ARPES.

(ii) Modeling MZM-relevant devices: Using micromagnetic and tight-binding modeling, we have demonstrated that stray fields from proximate micromagnets can induce sufficient spin splitting in a nanowire to in principle generate MZMs. We proposed designs that are best suited for the basic 2-MZM nanowires, as well as for the more advanced fusion and braiding devices [3]. We have also established that stray fields can strongly suppress superconductivity in thin shells of Al or Sn, which can lead to false MZM positives in experiments [4].

(iii) Towards spin qubits in IV-VI quantum dots: Quantum dot spin qubits are a leading quantum computing platform, with most of the recent progress concentrated on the group IV materials family (Si and Ge or combinations thereof). This aspect of our project is exploring group IV-VI compounds which feature strong spin-orbit interaction, large Landé g-factors, high mobilities, low resistance electrical contacts, and the possibility of isotopic purification. Some of the materials in the family are expected to be topological (SnTe) and some feature a strong dielectric response which can be leveraged e.g. for long-distance coupling of qubits. To this end, we have fabricated single and double quantum dots and explored their dot energies, spin-orbit interaction strength and anisotoropy, and the structure of their spin states. These results are in preparation for publication. We find unusual 45-degree anisotropy in both g-factor and SOI strength (Fig. 1), which hint at the crystalline origin of the phenomena. We also identify that the dots typically have quantum Coulomb energies, which allows study of an unusual regime for spin qubits. We also fabricated quantum dots on PbTe nanowires to study the fractional charge jumps that can be associated with anyon quasiparticles. We observed charge jumps that are nearly 1/3 of the quantum dot's conductance period. This was observed near zero magnetic field in a weakly interacting material, thus not in the regime where anyons are expected. We identify an explanation based on additional uncontrolled quantum dots coupled capacitively to the primary dot [5].

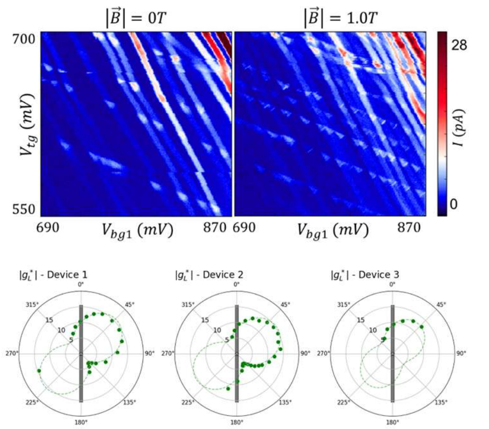

Fig. 1: Stability diagram (top) and g-factor anisotropy (bottom) of PbTe nanowire DQDs. These devices are useful for mapping out the properties spin-orbit coupling and Landé g-factors in this material, for topological and spin qubit applications.

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Multiscale Quantum and Classical Microscopy for Superconducting Quantum Systems
Award Number: ERKCK51

Light-matter interaction with quantum light

Chengyun Hua, Benjamin Lawrie, Yueh-Chun Wu (pd)

Oak Ridge National Laboratory

Abstract: Magneto-optical probes of condensed matter have drawn increasing attention over the past decade because of the potential for unveiling steady-state and out-of-equilibrium spin interactions and for probing spontaneous time-reversal-symmetry breaking in unconventional superconductors. However, a major obstacle for such spectroscopies is the photoinduced heating that can perturb the material under study especially in cryogenic environments. Quantum light sources open the possibility to couple to matter without the need for strong lasers. For example, quantum sensing using squeezed light for applications ranging from gravitational wave detection to scanning probe microscopies has enabled measurements with sensitivity beyond the shot noise limit (SNL) without introducing excessive heating. Our team has substantial experience in both optical manipulation of materials in low-temperature environments and the development and application of squeezed light sources.[1,2] Recently, we have demonstrated two-mode squeezed light sources can be used to improve the minimum detectable signal in magnetic circular dichroism (MCD) measurements by three decibels compared with state-of-the-art classical measurements, even with relatively lossy samples like terbium gallium garnet.[3] The two-mode squeezed light source is generated via four-wave mixing in ^{85}Rb vapor before the MCD signal is transduced onto the probe field and a balanced photodetection scheme is used to suppress noise below the SNL. By employing a two-mode squeezed light source to suppress photon shot noise, the sensitivity of the MCD measurement is enhanced, allowing detection of weaker signals with lower laser power—thereby reducing photothermal disturbances during cryogenic optical probing of quantum materials. These results establish a foundation for quantum-enhanced magneto-optical microscopies, which are especially vital for probing environmentally sensitive materials and for low-temperature measurements where increased optical power can cause unacceptable thermal disturbances.

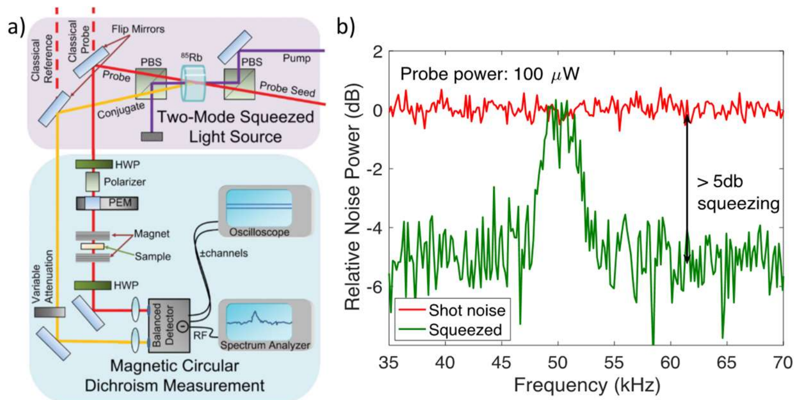

Figure 1: (a) Schematic of quantum enhanced magnetic circular dichroism experiment. (b) Demonstration of a quantum-enhanced polarization-sensitive signal (green) without a sample together with a shot noise limited trace (red) acquired with a probe power of 100 μW.

To further minimize perturbations induced by the macroscopic photon population in bright squeezed states of light, squeezed vacuum states of light readout through dual homodyne measurement schemes (as illustrated in Fig. 2) offer the potential for truly non-perturbative measurements of material properties in cryogenic environments. Previously, we demonstrated the first practical measurement of an AFM microcantilever displacement using a squeezed dual homodyne readout scheme, resulting in classically inaccesible measurement sensitivity of 1.7 fm/√Hz.[2] This scheme can be adapted for low-temperature magneto-optical Kerr effect measurements with the addition of a set of polarization optics. This quantum-enhanced approach is predicted to achieve a sensitivity of 10 nrad/√Hz with negligible photothermal interactions compared with the cooling power of our dilution refrigerators.[4] Such a combination of high sensitivity and low operating temperature cannot be achieved with any classical measurement technique. This approach will enable the detection of subtle spontaneous Kerr rotation arising from time-reversal symmetry breaking in a topological superconductor, and it could also unlock new quantum enhanced probes of phonon angular momentum that have recently been explored with classical readout schemes in the core MSE program at ORNL[5].

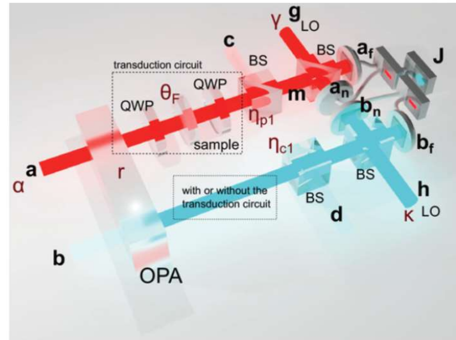

Figure 2: Optical setup for generating a two-mode squeezed light source comprising a probe and conjugate beam that exhibit intensity difference and phase-sum squeezing.

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Multiscale Quantum and Classical Microscopy for Superconducting Quantum Systems
Award Number: ERKCK51

Elucidation of Quantum Spin Defect Interactions with Quantum Materials

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Oak Ridge National Laboratory

Abstract: This thrust focuses on developing and integrating advanced quantum sensing platforms using spin defects such as the boron vacancy center in hexagonal boron nitride (hBN) and nitrogen-vacancy (NV) centers in diamond to probe magnetic fluctuations and superconducting phenomena from nano to mesoscale dimensions. The use of different sensing modalities matched to the application of interest addresses critical limitations in quantum magnetometry. By using scanning NV center magnetometry and relaxometry to investigate the critical behavior of a high-Tc ferromagnetic oxide near its Curie temperature[1], we addressed the technical challenges of probing the relevant length and time scales during thermally driven ferromagnetic-to-paramagnetic phase transitions, as highlighted in Figure 1. We have recently adapted this approach to capture the static and dynamic aspects of critical behavior in van der Waals ferromagnets like Fe_3GaTe_2, providing new insights into the universal properties governing magnetic phase transitions and demonstrating the power of NV-based techniques for studying critical phenomena across the relevant spatial and temporal scales.

Spin defects in hBN provide unique advantages for sample-sensor proximity due to their 2D and (in principle) arbitrary operational thickness as well as out of plane spin orientation allowing for close distances between the defect plane and probe system while simplifying bias field alignment. With this approach, we have recently monitored spin dynamics near a phase transition between collinear and incommensurate spiral antiferromagnetic phases using T_1 relaxometry of boron vacancy defects in an hBN flake transferred onto $LuMn_6Sn_6$, as shown in Figure 2. We are currently mapping the spin dynamics near different phase transitions in $LuMn_6Sn_6$ with all optical relaxometry measurements in a low temperature high-field environment in order to better understand the complex phase diagram of this Kagome magnet. In addition to these efforts, we have probed the energetics and dynamics of silicon vacancy (SiV) centers in diamond under arbitrary magnetic field orientations in a dilution refrigerator.[2,3] These experimental approaches will be adopted for NV centers and boron vacancy spin defects to advance mK spin-based quantum sensing protocols over the next few years. Finally, in collaboration with the ORNL BES-MSE core program[4] we are developing new e-beam-induced etching techniques to optimize the

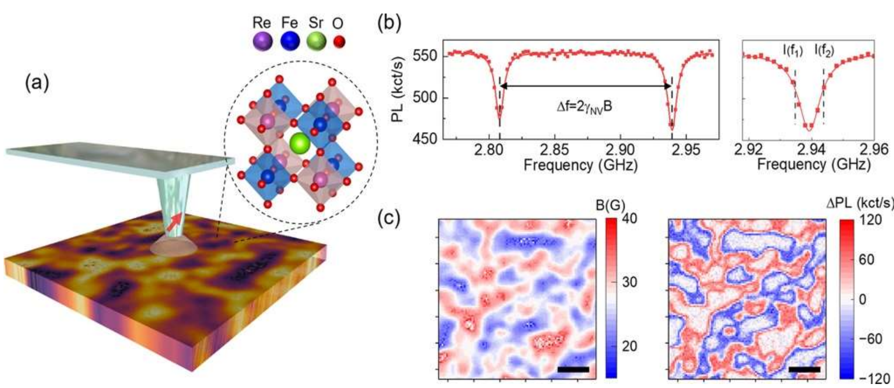

Figure 7 (a) Schematic illustration of nanoscale spin-based probe of perovskite ferromagnetic oxide Sr_2FeReO_6. (b) Measured ODMR spectra for imaging of local magnetic textures (scale bar 400 nm) depicted in (c).

brightness and directionality of emission from spin defects while controlling surface chemistry to optimize spin-based sensing functionality.

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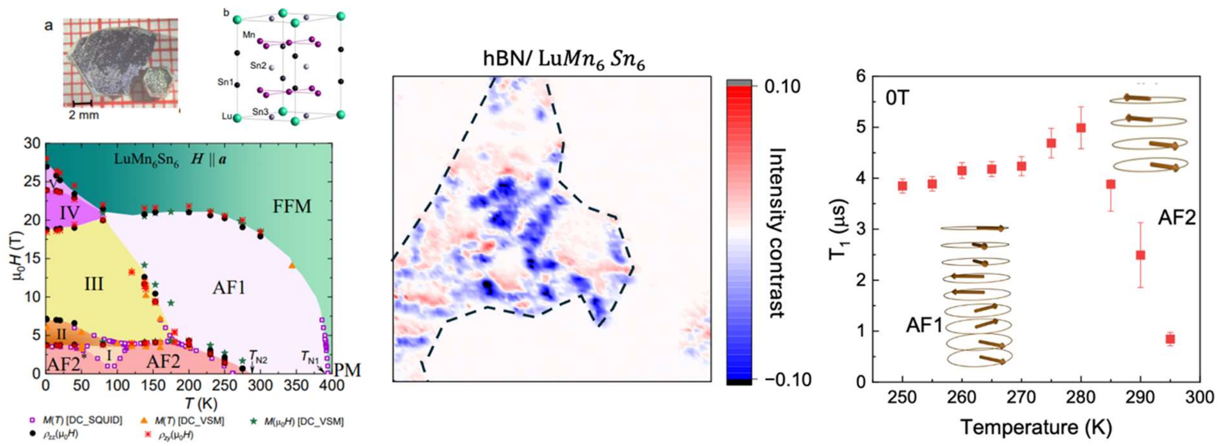

Figure 8. Antiferromagnetic kagome compound $LuMn_6Sn_6$ with a rich phase diagram prepared with hBN flake on top of polished surface (left). Map of magnetic domains in $LuMn_6Sn_6$ measured with boron vacancy spin defects (middle) and associated zero-field temperature dependent T_1 relaxation times spanning the AF2-AF1 transition (right).

centers in off-axis magnetic fields. *Phys. Rev. B* **111**, 155426 (2025). doi: 10.1103/PhysRevB.111.155426

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Project Title: Understanding the Many-body Properties of Rare Earth-Based Materials Relevant to Next Generation QIS Applications
Award Number: PM-062

Abstract Title: Neutron Scattering Studies of Quantum Coherence in Yb Spin Chains

Names of PI, (co-PI): Igor A. Zaliznyak[1]**(co-PI)**, **Andreas Weichselbaum**[1] **(co-PI), Robert Konik**[1] **(PI)**, **Lazar Kish**[1] **(Postdoc),** Daniel M. Pajerowski[2], Andrei T. Savici[2], Andrey Podlesnyak[2], Leonid Vasylechko[3], Alexei Tsvelik[1] (unfunded collaborators)

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Abstract: The goal of this research program is to use neutron scattering to investigate the coherence properties of Yb and other rare earth (RE) based solid state qubit candidate systems and how these properties can be modified and manipulated using time-structured external stimuli such as laser excitation tuned to the RE specific optical transitions.

In the past and current funding periods, this program has developed an optical fiber-based setup for in-situ laser pumping of optically active samples during neutron experiments, implementing laser pump neutron probe capability for scattering experiments on QIS-relevant materials. The setup has been used for a set of initial experiments on HYSPEC and SEQUOIA spectrometers at the SNS and at WAND2 diffractometer at HFIR. The laser setup allows time-resolved stroboscopic measurements of relaxation processes in quantum systems following interaction with coherent laser photons with time resolution of about 20 to 40 microseconds. The initial experiments yielded promising results and allowed developing experimental protocols and data processing workflows for time-resolved laser pump neutron probe measurements.

One key question for potential applications of RE-based solid-state qubits in QIS is that of thermal decoherence. Thermal effects can be important for optical coupling to quantum states in crystals where interaction with energetic photons can lead to substantial heating effects resulting in a loss of quantum coherence. Indeed, conventional wisdom dictates that quantum effects become unimportant at high temperatures. In magnets, it has been expected that when thermal energy exceeds interactions between atomic magnetic moments, the moments become uncorrelated, and classical paramagnetic behavior is restored. This thermal decoherence of quantum spin behaviors is a major hindrance to quantum information applications of spin systems. Remarkably, our neutron scattering experiments on Yb chains in $YbAlO_3$, an insulating perovskite crystal, defy these conventional

expectations [1]. We observe a sharply defined spectrum of spinons, fractional quantum excitations of spin-1/2 chains, to persist to temperatures much higher than the scale of the interactions between Yb magnetic moments. The observed sharpness of the spinon continuum's dispersive upper boundary indicates a spinon mean free path exceeding approximately 35 inter-atomic spacings at temperatures more than an order of magnitude above the interaction energy scale (Figure 1A). While the coherence of quantum excitations persists, the lower bound on quantum entanglement as quantified by quantum Fisher information, demonstrates universal power-law decline with temperature (Figure 1B).

[1] (funded by this project) "High-temperature quantum coherence of spinons in Yb spin chains", Lazar Kish, Andreas Weichselbaum, Daniel M. Pajerowski, Andrei T. Savici, Andrey Podlesnyak, Leonid Vasylechko, Alexei Tsvelik, Robert Konik, Igor A. Zaliznyak. Nature Communications (acceptance offered, 2025), https://doi.org/10.48550/arXiv.2406.16753.

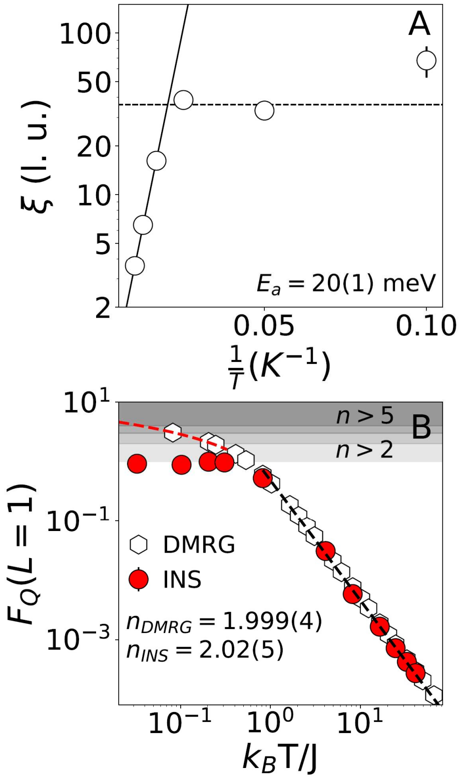

Figure 1. (A) Quantum spinon coherence length in $YbAlO_3$ obtained using the measured spinon dispersion and lifetime extracted from the spectral broadening. (B) Temperature dependence of maximal quantum Fisher information, $F_Q(L = 1)$. Dashed black line is a power-law fit to the data in $T \geq 2$ K range capturing asymptotic high-temperature behavior, FQ ~ $(J/T)^n$, with n = 2. Dashed red curve, shown in the region below T_N = 0.8 K (shaded), is a fit of DMRG data below 4 K to a logarithmic dependence, illustrating the low-T asymptotic behavior.

Project Title: Understanding the Many-body Properties of Rare Earth-Based Materials Relevant to Next Generation QIS Applications
Award Number: PM-062

Abstract Title: Witnessing Quantum Entanglement Using Resonant Inelastic X-ray Scattering

Names of PI, coPI: Mark P. M. Dean[1](co-PI), **Jonathan Pelliciari[1](co-PI)**, **Robert M. Konik[1] (PI)**, **Igor Zaliznyak[1](co-PI)**, **Wei He[1]** (**Postdoc**), **Lazar Kish[1]** (**Postdoc**), Yao Shen[1], Sophia TenHuisen[4], Mary H. Upton[2], Diego Casa[2], Petra Becker[3], Matteo Mitrano[4], Jennifer Sears[1] (unfunded collaborators)

Affiliations: [1]Brookhaven National Laboratory, [2]Argonne National Laboratory, [3]University of Cologne, [4]Harvard University

Abstract: Quantum entanglement is a fundamental property underpinning quantum many-body physics and emerging quantum technologies, such as quantum metrology, communication, and sensing. Although there are many approaches for quantifying entanglement in synthetic quantum systems, most of these are impractical for solid-state quantum materials, due to either the large number of atoms in such materials or the lack of fine-scale control. The creation and control of coherence in solid-state quantum systems requires improved methodology to detect entanglement.

Recent years has seen impressive progress in the use of neutron spectroscopy to detect entanglement in solids [1]. This has been proven to be a sensitive means to quantify spin entanglement. Resonant inelastic x-ray scattering (RIXS) is an advanced experimental method capable of detecting the spin, lattice, orbital, and charge degrees of freedom in quantum materials, with capabilities for measuring under device like conditions such as under electrical current or after illumination with a laser [2]. The RIXS process, show in the left panel of Figure 1, involves the resonant absorption of an x-ray photon, creating an intermediate state with a core hole and a valence excitation, before the hole is filled via the emission of another x-ray photon. By measuring the energy and momentum change of the x rays, one can infer the properties of the excitations created in the material.

At Brookhaven we have developed methods to use RIXS to quantify orbital entanglement of spin states in solids [3] as illustrated in Fig. 1. Our approach builds on the so-called quantum Fisher information (QFI) metric, which is connected to the probability distribution of measurements and the corresponding parameter estimation in a multiparticle quantum system. If the precision of a parameter estimation exceeds the classical limit, then it can be deduced that the system must have multipartite entanglement. Since the quantum operator associated with neutron scattering operator is Hermitian, it can be directly applied to obtain the QFI. For RIXS, the operator is non-Hermitian, and we needed to develop a new approach.

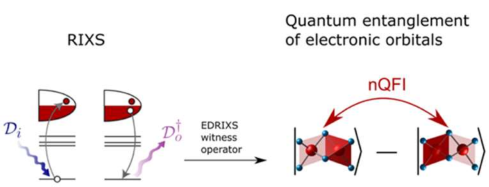

Figure 9: Witnessing orbital entanglement using RIXS. In this work, we develop a joint experimental-theoretical method to obtain an orbital entanglement witness from the RIXS intensity. We have developed a method to use RIXS to obtain the normalized quantum Fisher information (nQFI) for detection of the quantum entanglement.

The first step in the process is to measure the RIXS spectrum of the material of interest. We are currently working on transition metal and rare earth systems such as $Ba_3CeIr_2O_9$, Yb:YAG, Yb:YAP. We first obtain the RIXS operator as a function of incident energy and incoming and outgoing x-ray polarization for the orbital system in question using the numerical codebase EDRIXS [4] by fitting the observed empirical spectrum to a simulation as shown in Fig. 2. We then compute the nQFI from the RIXS operator and determine the optimum conditions to detect entanglement. For example, the right panel of Figure 2 shows show the RIXS-determined nQFI for $Ba_3CeIr_2O_9$ varies with x-ray momentum. Choosing specific angles corresponding to constructive interference between dimer sites allows the detection of multipartite entanglement based on the values exceeding the dotted red line.

We are currently completing our studies of Yb spin systems Yb:YAG and Yb:YAP under laser illumination corresponding to the J=7/2 to 5/2 transition that is widely used in laser and communication technologies. We are also extending our work to Yb implanted van der Waals materials.

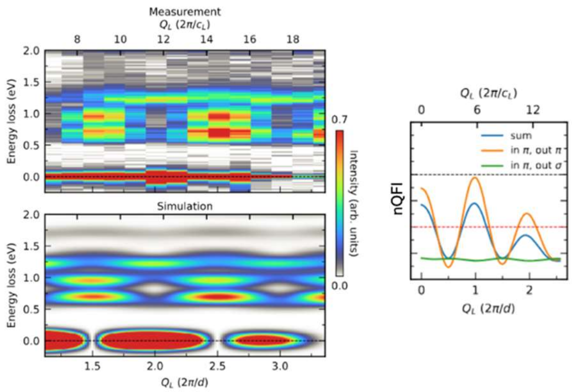

Figure 10: Top left: Ir L_3-edge RIXS spectrum of $Ba_3CeIr_2O_9$ as a function of energy loss and momentum. Bottom left: EDRIXS simulation of the spectrum above made using the EDRIXS software based on a model for the active Ir $5d^5$ orbitals. Right: nQFI as function of momentum for different outgoing x-ray polarizations.

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DNA-Controlled Dye Aggregation – A Path to Create Quantum Entanglement

DE-SC0020089

Toward Measuring an Exciton-Exciton Coherence in a DNA-Assembled Molecular Aggregate

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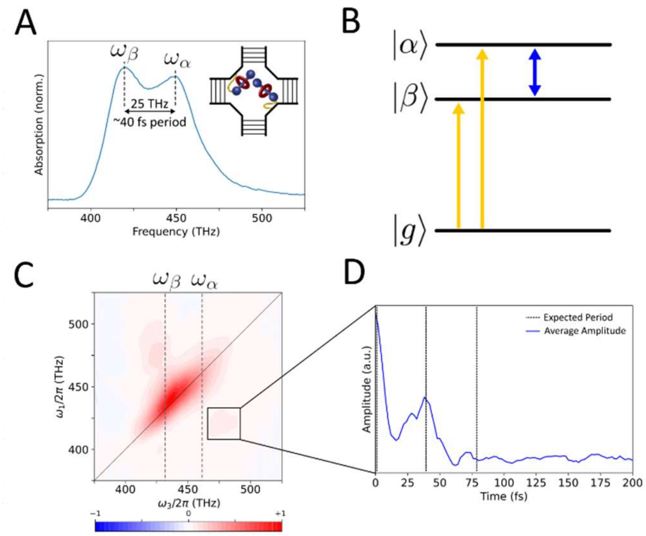
Abstract: Our emphasis has been on engineering and assembling materials and building tools and analysis techniques to directly measure an exciton-exciton coherence, a key ingredient in quantum entanglement, in DNA-assembled molecular aggregates. A key significant prior development, which refined our scope, includes the discovery of squaraine:rotaxane dimers that have an oblique orientation and as such optical properties making them ideally suited to produce exciton-exciton coherence (ω_h and τ_d) and entanglement. In addition, we are working to characterize the difference dipole moment (Δd) of chromophores, which governs the amplitude of exciton-exciton interactions (K), also important in creating entanglement in molecular aggregates.

In pursuit of our goal to measure an exciton-exciton coherence, we refined a noncollinear optical parameter amplifier (NOPA) for producing very stable, spectrally flat, excellent spatial mode quality, and transform-limited ultrabroadband laser pulses,[1] designed and constructed a phase-stable interferometer enabling 2D spectroscopy in the pump-probe geometry,[2] which has higher stability and is more compatible with cryostats, and applied analysis techniques (previously only theorized) to determine if measured quantum beats are purely electronic (*i.e.*, exciton-exciton coherence), mixed electronic-vibronic, or purely vibrational.

We recently incorporated a 77 K cryostat into our optical setup, which should extend the quantum beats and improve our ability to resolve them. The figure at right shows the preliminary measurements that seemingly indicate exciton-exciton coherence, however we are conducting numerous control measurements to confirm this conclusion. Our future plans include incorporating a 4 K cryostat into our optical setup, which will dramatically improve our ability to resolve and analyze the exciton-exciton coherence. After directly measuring and confirming the presence of exciton-exciton coherence, we will perform polarization measurements to determine whether the coherence is indicative of entanglement, a longstanding goal in studies of molecular aggregates for the past two decades.



A) Schematic and steady-state absorption of the oblique SeTau670 dimer tethered to a DNA Holliday junction. B) Energy level diagram, with exciton-exciton coherence between states |α⟩ and |β⟩ shown in blue. C) Normalized 2D spectrum with states |α⟩ and |β⟩ (dashed lines). D) The average amplitude of the cross peak between |α⟩ and |β⟩ as a function of time, with expected period of the coherence (dashed lines). One period of the exciton-exciton coherence appears to have been detected.

In parallel with the efforts above, we identified mechanisms in molecular materials that act to drastically reduce the singlet excited-state lifetime (τ_p), which can, under certain conditions, be detrimental to τ_d. While in-house synthesized bacteriochlorins (BCs) featuring both dimethylamino and nitro groups appear to increase Δd,[3] the latter substituents have a detrimental influence on τ_p in polar solvents.[4] We also identified a material system that is able to overcome aggregation-induced quenching (AIQ), a phenomenon we ascribe to the introduction of new internal conversion pathways that drastically reduce τ_p upon aggregation (manuscript in revision). Our results suggest that it may be possible to overcome AIQ by tailoring the environment of the aggregate, that is, tailoring the local sterics and electrostatics.

Additionally, in pursuit of our goal to characterize Δd of chromophores, we constructed an electro-absorption spectrometer.[5] We used the spectrometer to characterize Δd of field standards, common dyes, and organic semiconductors with biocompatible tags (manuscript in preparation).

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Robust Quantum Computing for Condensed Matter Physics

DE-SC0023231

Alexander Kemper (PI), Department of Physics, North Carolina State University; and **James Freericks (co-PI)**, Department of Physics, Georgetown University

Abstract:

In this talk, I will discuss advances we have made in bringing the power of quantum computers to bear on solving problems in condensed matter systems, in particular focusing on predicting or simulating spectroscopic experiments of systems in and out of equilibrium, which is the primary goal of this QIS project.

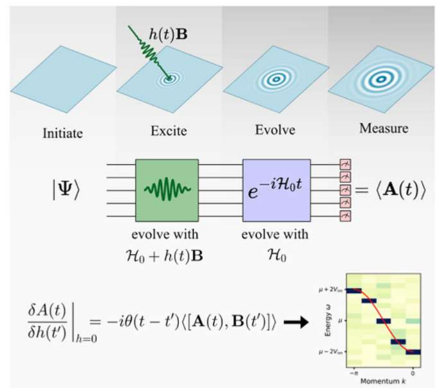
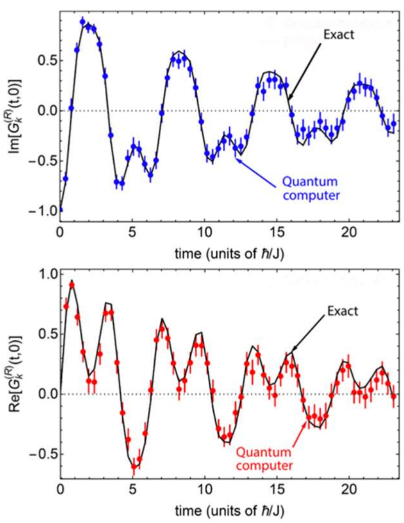


Fig. 1 | Linear response method. We establish an equivalence between the experimental measurement of a response function and an ancilla-free quantum simulation under a time dependent Hamiltonian that includes the perturbative excitation $h(t)\mathbf{B}$. Following excitation, the system is evolved under $\mathcal{H}_0$, and $\mathbf{A}$ is measured. The functional derivative of $A(t) = \langle \mathbf{A}(t) \rangle$ with respect to $h(t')$ yields the retarded response function shown in the figure. The data shown is taken from Fig. 3.

First, I will a **linear response framework** for quantum simulation, which establishes a direct bridge between what we can measure experimentally and the fundamental **quantum many-body state** of a material. The core of this method is its ability to compute crucial **bosonic and fermionic correlation functions** directly on a quantum computer. Unlike previous techniques, it allows for a wide range of operators within a single quantum circuit, which means you can acquire multiple types of correlation functions—both frequency- and momentum-selective—simultaneously. This is a significant advantage because it avoids compounding errors and eliminates the need for extra "ancilla" qubits, making the process more efficient and reliable. We demonstrate this framework by applying it to a **charge-density-wave material** using IBM's quantum hardware (specifically, the ibm_auckland superconducting quantum computer). Even in a noisy quantum computing environment, the method successfully extracted meaningful data, highlighting its robustness. This research significantly advances the possibility of using **quantum computers** to accelerate the discovery and design of new **quantum materials**. By providing a streamlined way to calculate these complex response functions, it not only enhances the capabilities of current and near-term quantum devices but also suggests potential improvements for classical simulations by offering a path to bypass computationally intensive vertex corrections. NatComm 2024, 10.1038/s41467-024-47729-z.

As an interlude, I will briefly introduce a new property of Green's functions and other observables, which allows for a form of error correction of experimental spectra (including quantum simulated spectra). PRL 2024, 10.1103/PhysRevLett.132.160403

Finally, I will pivot to the problem of open quantum system dynamics. Our work explores the simulation of **open quantum systems** on near-term quantum hardware, addressing the critical challenge of how quantum systems interact with their environment, and highlighting the potential of current quantum computers to tackle problems traditionally difficult for classical methods, particularly in the context of driven and dissipative phenomena.



Our work, completed under this project, shows that algorithms for open quantum systems outperform similarly complex non-dissipative algorithms on noisy hardware. The first paper, "Simulating open quantum systems on quantum hardware with robustness to hardware errors," focuses on the remarkable **robustness of open quantum system simulations to hardware errors**, even with deep circuits involving up to two thousand entangling gates. This capability raises the need to be able to probe these systems spectroscopically, so int a second paper, "Measuring n-point correlation functions in open quantum systems on a quantum computer," we propose a **unified hierarchical method** for measuring n-point correlation functions in open or non-equilibrium quantum systems. This method involves repeatedly interrupting the system's time evolution with an ancilla qubit interaction and immediate measurement. We emphasize the robustness of this technique compared to other ancilla-based interferometric methods like the Hadamard test, making it well-suited for noisy, near-term quantum computers. The paper demonstrates its efficacy by measuring single-particle Green's functions of a driven-dissipative fermionic system on a quantum computer, proving that dynamical correlation functions for such systems can be reliably obtained. NPJQI 2025, 10.1038/s41534-025-00964-8, and PRL 2024 0.1103/PhysRevLett.132.100601.

Both papers contribute to the growing field of quantum simulation of open systems. Together, they offer compelling evidence for the feasibility and promise of using near-term quantum computers to explore complex, environmentally coupled quantum phenomena.

Trade off-free entanglement stabilization in superconducting circuits

Award Number: DE-SC0019461

Names of PI, coPI(s), and unpaid collaborators: **Leonardo Ranzani[1]** (RTX-BBN Technologies), **Archana Kamal[2,3], Guilhem Ribeill[1], Luke Govia[1], Raymond Simmonds[4], Jose Aumentado[4], Daniel Campbell[5],** Tristan Brown[1,2], Zhihao Xiao[2], Emery Doucet[2], Tarush Tiwari[1,2]

RTX BBN Technologies[1], University of Massachusetts Lowell[2], Northwestern University[3], NIST Boulder[4], Air Force Research Laboratory, Rome[5]

Abstract:
In the PIQUE project we aim to demonstrate novel techniques and devices to control and measure quantum systems using parametric interactions and engineered dissipation [1]. In this abstract we focus on the demonstration of autonomous state stabilization of a maximally entangled state of two transmons [2]. Unlike traditional protocols, PIQUE minimizes coherent leakage out of the target state, thus avoiding any inherent trade off between target state preparation fidelity and convergence time. We accomplish this by creating a purely dissipative channel for population transfer into the target state (Figure 1(a)), mediated by strong parametric interactions coupling the second-excited state of a superconducting transmon and the engineered bath resonator. Our scheme achieves a state preparation fidelity of 84% with a stabilization time

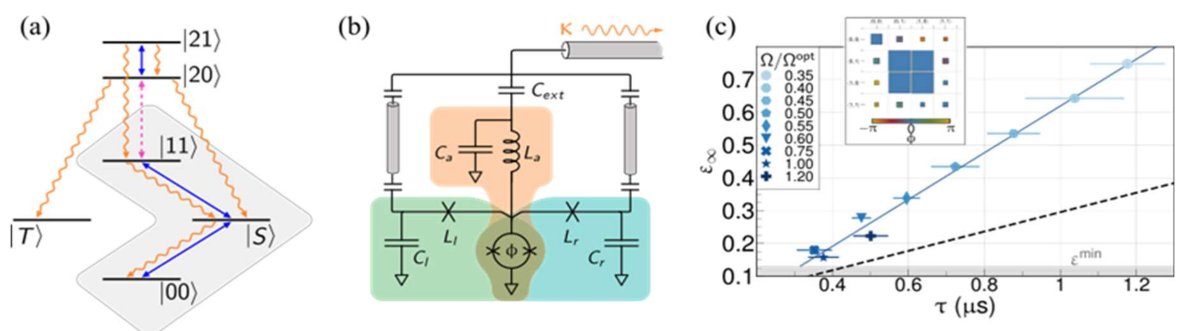


Figure 1 – (a) Energy level diagram showing dissipative preparation of Bell state. (b) Circuit schematic of the device, consisting of two transmons and a resonator connected by a joint SQUID. The resonator provides a controlled bath with finite linewidth. (c) As we increase the drive strength, preparation error and time decrease monotonically, achieving state fidelity limited only by qubit intrinsic decoherence (quantum state tomography in the inset).

constant of 339 ns, leading to a state-of-the-art 54 ns error-time product in a solid-state quantum information platform (Figure 1(c)).

In order to scale our technique to multi qubit systems, we developed a new modular tunable coupler that minimizes spurious crosstalk and maximizes linearity. The coupler (Figure 2(a)) consists of two ancilla qubits connected by a small Josephson junction. The qubits hybridize and mediate a strong interaction with an intrinsic cancellation point (Figure 2(b)), which is first order insensitive to the data qubit frequencies and results in low ZZ interaction. Moreover, at the cancellation point the effective coupling rate is approximately linear in flux, enabling fast parametric coupling and suppressed nonlinear frequency shifts.

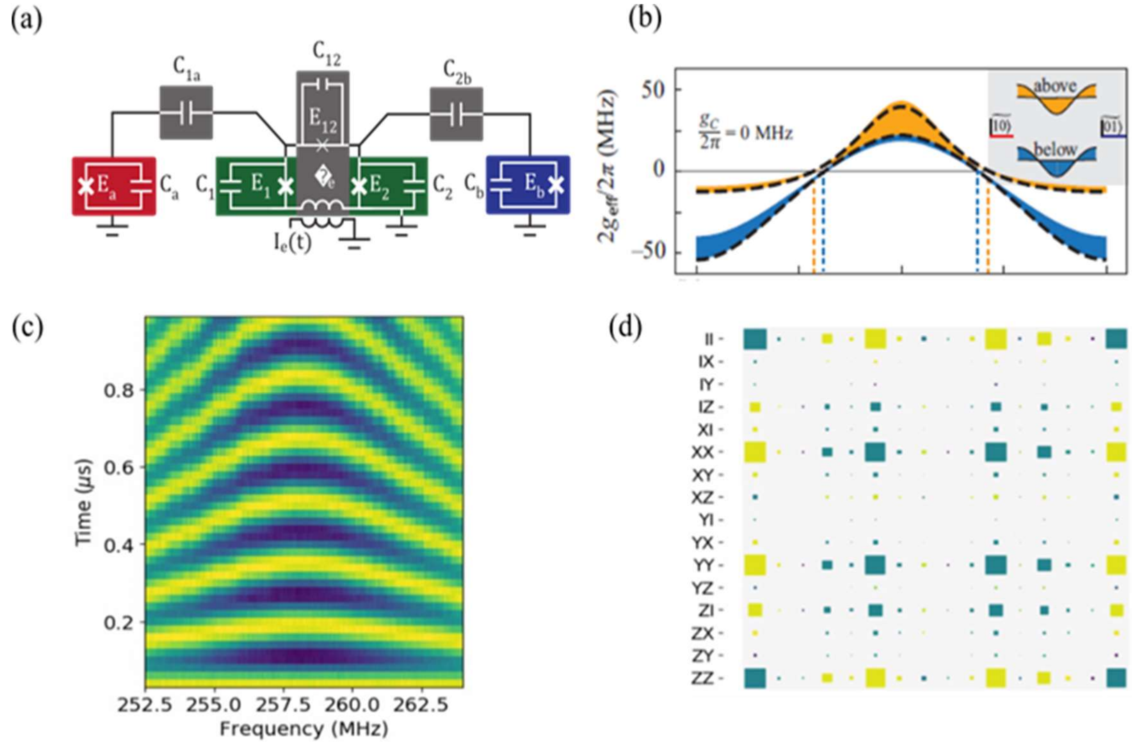


Figure 2 – (a) Improved dual transmon coupler to reduce crosstalk and improve linearity. (b) The coupler has an intrinsic zero coupling flux point near 0.25. (c) Measured parametric iSWAP between the data qubits and (d) quantum process tomography with 97% gate fidelity.

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Hybrid Magnon Systems
DE-SC0022060

Quantum Sensing of Magnons Using a Qubit

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Abstract

We are investigating coherent dynamics of propagating magnons in hybrid magnetic devices down to the single quantum regime and implement these effects in quantum coherent operations with circuit quantum electrodynamics (cQED) systems. Towards this end, our ultimate goal is to develop magnetic field-tolerant superconducting quantum devices and use them to engineer interactions between qubits and magnons. As a first step, we realized an integrated, hybrid cQED-magnon system integrating a superconducting transmon qubit with a YIG sample biased locally by permanent magnets [1]. Using this hybrid system, we performed high-sensitivity/high-dynamic range sensing of magnons and their dynamics (see Fig. 1): Using qubit spectroscopy we sensitively measured the occupation number of the Kittel mode of YIG with a sensitivity of a few $\text{magnon}/\sqrt{\text{Hz}}$ over a dynamic range of about 2000 magnons. Using quantum control of the qubit dynamics we also measured the decay dynamics of the Kittel mode with a sensitivity of a few $\text{ns}/\sqrt{\text{Hz}}$, and we demonstrated a technique for resolving fast decay dynamics using a parametric pumping technique.

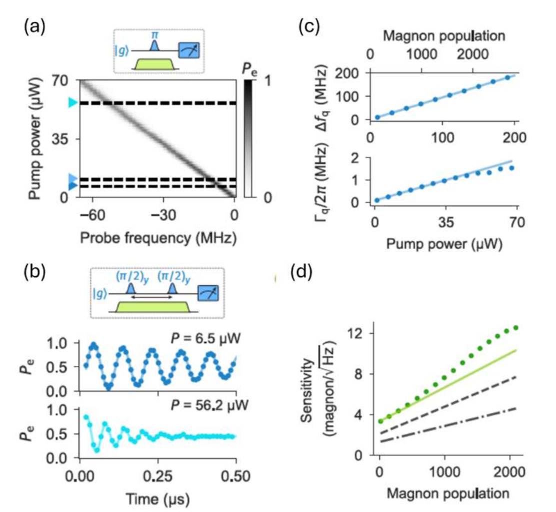

Fig .1: (a) Qubit frequency shift as a function of pump power applied to magnon mode. (b) Ramsey decay of qubit for different pump powers. (c) Stark-shifts and dephasing rates as a function of pump power and magnon population. (d) Magnon sensitivity as a function of population.

Since standard transmons are highly susceptible to magnetic fields we have begun to investigate field-compatible superconducting thin films as platforms for field-resilient quantum circuits. We fabricated and characterized thin-film co-planar waveguide and lumped-element resonators made from disordered superconductors (TiN and NbTiN) that have high critical fields in intrinsic nonlinear kinetic inductance (NLKI). We found high quality factors in such devices and characterized their nonlinearity. We observed strong pump-induced Stark shifts of the resonator frequency and obtained initial evidence that this nonlinearity is sufficient to dynamically control the hybridization between statically coupled resonators.

For future quantum circuit development, we will work towards the development of a field compatible quantum circuit platform with tunable interactions. Towards this end, we will work with magnetic-field-compatible eNe qubits and single-crystal YIG. The goal is to achieve strong coupling between the magnon state of a YIG sphere and the qubit state of an eNe qubit, mediated by microwave photons in a coplanar superconducting resonator. We will explore the magnetic field tolerance of the eNe qubit and study the evolution of coherence under strong magnetic fields in order to explore the frequency limitation of this architecture.

We also plan to study the integration of eNe qubit with YIG thin-film samples and explore quantum sensing of a single propagating magnon. For this planned work, we will continue working on magnon-photon coupling with a coplanar superconducting resonator fabricated on YIG thin films and explore time-domain coherent control of propagating magnons, similar to our recent Nature Communications publication about time-domain coherent magnon control in YIG spheres. Furthermore, we plan to explore the operation of an eNe qubit which is fabricated on YIG thin film instead on a Si substrate and study the compatibility of eNe qubit on magnetic thin film.

With respect to using superconducting qubits, we will pursue two complementary strategies. First, we will investigate the interplay of non-linear kinetic inductance (NLKI) and intrinsic quality factor, Q. If high NLKI and high Q can coexist, we will be able to develop nonlinear high-Q resonators that are field-compatible, and that can be coupled on-demand to magnon-polariton modes through a microwave pump. Second, we will use thin-film NbTiN as base layer to realize field-compatible superconducting "SNAIL-mon" qubits that combine high coherence with 3-wave mixing capability. If these qubits can be made with high-coherence in an external magnetic field, they will allow for strong tunable interactions between quantum circuit and magnon-polariton modes.

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Modeling, probing, and controlling quantum coherence in materials
FWP: SCW1674, DE-SC0020313

Modeling, probing, and controlling defects in materials for superconducting qubits and spin qubits

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[1]Lawrence Livermore National Laboratory, [2]University of Wisconsin — Madison, [3]University at Buffalo, [4]University of California, Berkeley

Different superconducting qubits on the same chip, or the same qubit at different times, can exhibit different relaxation times. This may be caused by different distributions of TLSs in frequency, electric dipole, and location in the qubit device. To explore the effect of TLS distribution in frequency and space, we developed a model of the interactions between the qubit and over 300 different baths consisting of 200 TLSs each. We found that TLSs in regions of high electric field in the resonator can significantly reduce the qubit coherence time, with the strongest electric fields contained in the Josephson junction (JJ). However, TLSs away from the JJ can also reduce qubit lifetime. We find that in some cases a single TLS reduces the qubit lifetime and in other cases a collection of more weakly coupled TLSs also reduce qubit lifetime [1].

To understand the nature of the TLS landscape under different conditions, we have performed TLS spectroscopy on a superconducting resonator by applying an electric bias that shifts the TLS asymmetry energy, thus bringing TLSs into and out of resonance. TLSs can then be seen as avoided crossings in the resonator transmission spectra that are hyperbolic. We also applied a large alternating electric bias while at low temperature and found that TLS hyperbolae are no longer visible, indicating a profound change in the TLS landscape. Engineering the TLS landscape could allow for qubits with dramatically longer coherence times. In another study, we analyzed TLS density and loss in tantalum superconducting resonators, finding that the grain structure and growth conditions affect loss [2]. We also used scanning electron microscopy, x-ray diffraction, low-T transport, secondary ion mass spectrometry, and transmission electron microscopy to reveal the dependence of residual impurities and screw dislocation density on Ta growth conditions [3]

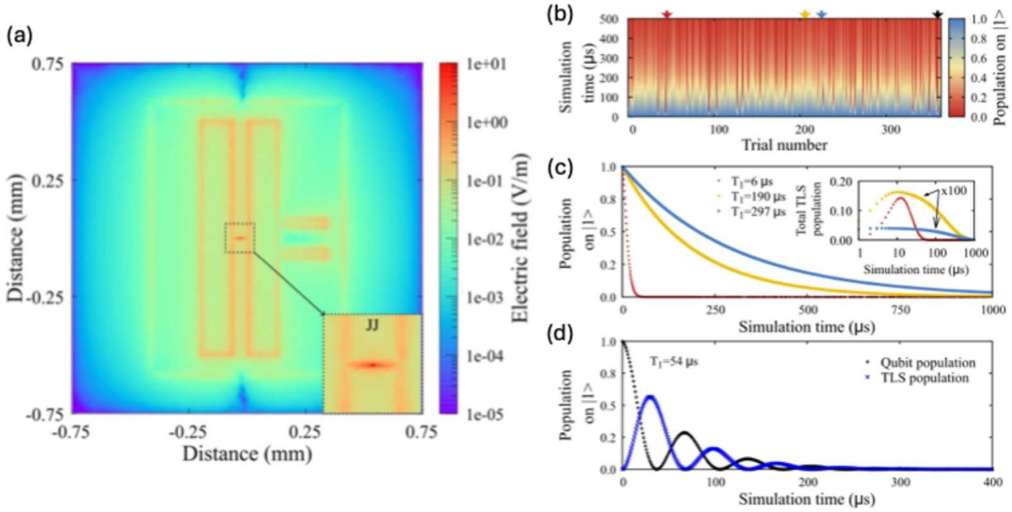

Figure 1: (a) Electric field distribution of the superconducting qubit, Josephson junction indicated. (b) Time-dependent qubit population for 364 simulated TLS baths, blue (red) indicates the excited (ground) state. (c) Linecuts for the three cases indicated by the colored arrows in panel (b), showing qubit relaxation curves when the qubit T_1 is shortest (6.33 μs, red), average (189.96 μs, yellow), and longest (296.81 μs, blue). Inset: corresponding total population of TLS excited states as a function of time. (d) When Δ_0 of the strongly coupled TLS is small and $\mathbf{p}\cdot\mathbf{E}$ is large, corresponding to the trial indicated by the black arrow in panel (b), we observe an exchange of population between the qubit excited state and the collective TLS excited state. Figure adapted from Ref. [1]

We have investigated charge noise in a tantalum transmon qubit and found the charge-offset dynamics are dominated by rare, discrete jumps between a finite number of quasistationary charge configurations, a novel behavior [4]. In another study related to charge noise, calculations on defects in a $Si_{1-x}Ge_x$ spin qubit revealed that Ge dangling bonds are more problematic charge-trapping centers than Si dangling bonds [5]. Similar computational studies on defect levels in Al_2O_3 have informed upcoming work on identifying defects in that material using KPFM.

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Carbon-Based Quantum Information Science with Symmetry Protected Topological States DE-SC0023105

Abstract Title: Engineering low energy modes in 1D and 2D quantum materials

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Abstract: The unmatched quality and atomically precise control over quantum electronic states in low-dimensional materials provided by a synthetic bottom-up approach represents a fertile ground for the discovery and engineering of new phases of matter with potential to advance quantum information transport, storage, and processing (*1–12*). In these materials emergent phenomena arising from unique interactions of energy and disparate states of matter at atomic and molecular length-scales can be rationally designed and shaped by theory, molecular design, and quantum physics. Research efforts throughout this grant period have been organized into two themes.

Progress in 1D systems: Bottom-up synthesized graphene nanoribbons (GNRs) have emerged as testbeds for realizing simple quantum systems due to extensive opportunities to engineer their electronic structure and the ease of creating localized symmetry protected topological (SPT) states within a bulk energy gap. We successfully implemented topological zero modes (ZMs) as the basis states for the engineering of effective low-energy designer Hamiltonians in the strongly hybridized regime. In the symmetric case, realized in the topological junction interface of a 9/7/9-double quantum dot, the strong hybridization between adjacent ZMs gives rise to an exceptionally small quasiparticle gap of 0.16 eV, corresponding to a carbon-based small molecule long-wavelength infrared (LWIR) absorber (*6*). The case of staggered basis state energies is realized in the dilute nitrogen core-doped armchair GNRs. Here the interaction of topological boundary states at the interface between doped and undoped segments can be captured by an effective Hamiltonian where the hybridization interaction strength is tuned by the distance between topological ZMs (*8*). To explore the weakly hybridized regime we successfully stabilized SPT end states lining the zigzag edges of armchair GNRs by lateral fusion with 5-membered rings (*3*). As a function of GNR length we observe a gradual decrease of the magnetic exchange coupling 136 meV $> J >$ 9 meV between off-zero mode end states reflected in an evolution from a closed-shell to an open-shell singlet ground state. Critical advancements to molecular on-surface synthesis protocols have enabled the realization of inversion symmetry breaking in the structure of [3]triangulene-GNRs (*7*). WE developed the head-to-tail fusion of [3]triangulene cores featuring a $S = 1$ ground state to ensure and effective mixing of the majority and minority sublattices between molecular building blocks. The resulting electronic structure is characterized by a strongly hybridized gapped bulk phase featuring magnetic SPT end states.

Progress in 2D systems: Building on our initial success in assembling metal-organic Kagome lattices from N-heterocyclic carbenes (*5*) we expanded the concept of ZM engineering to access hopping frustrated flat bands in nanographene frameworks. We reported the design and synthesis of a strongly hybridized π-conjugated COF featuring high-spin ($S = 3/2$) D_{3h} symmetric aza-[3]triangulene (A[3]T) cores linked by ethynylene groups (–C≡C–) fused normal to their zigzag edges (Fig. 1). The electronic structure of this edge-sharing 2D A[3]T crystal can be modeled as a diatomic Kagome lattice. Each A[3]T unit cell hosts a set of symmetry-related sixfold degenerate Wannier basis states giving rise to a pair of valence band (VB) and conduction band (CB) complexes, each featuring a non-trivial Kagome flat band, bracketing the Fermi energy (E_F) (*11*). In a related effort we demonstrated the covalent assembly of a honeycomb lattice of corner linked high-spin ($S = 3/2$) D_{3h} symmetric [4]triangulene cores. While electronic theory had predicted a nonmagnetic ground state reminiscent of a ying-yang diatomic Kagome lattice we were able to show that weak hybridization between corner sharing [4]triangulene cores favor a magnetically ordered ground state best described by a Néel antiferromagnet.

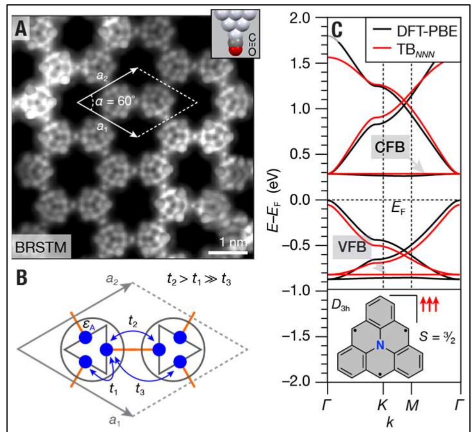

Fig. 1| Molecular engineering of hopping frustrated flat bands in A[3]TCOFs. (A) Constant-height BRSTM image of a representative segment of the *p6mm* lattice of A[3]TCOF. (B) Intra- and inter-unit cell electron hopping parameters (t_1, t_2, t_3) and onsite energy (ε_A) for a diatomic Kagome lattice. (C) DFT-PBE band structure for a freestanding A[3]TCOF Kagome lattice (*11*).

Future directions: Engineering geometric frustration in 1D and 2D spin lattices with bottom-up synthesized nanographenes (e.g. triangular/Kagome lattice, diamond Heisenberg chain).

Publications: (9 published, 1 accepted, 2 under review)

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Van der Waals Reprogrammable Quantum Simulator

DE-SC0022277

Benjamin Hunt [1], Dengyu Yang[1], Qingrui Cao[1], Junwon Choi[1], Erin Akyuz[1], Jeremy Levy[2], Patrick Irvin[2], Ranjani Ramachandran[2], Muqing Yu[2], Jon-Paul Maria[3], John Hayden[3], **Josh Nordlander[3]**, Ian Mercer[3], **Di Xiao[4], Nishchay Suri[1], and Chong Wang[4]**

1-Carnegie Mellon University, 2-University of Pittsburgh, 3-Pennsylvania State University, 4-University of Washington

Abstract: The primary motivation of this DOE-QIS project is to realize a condensed-matter (c-m) quantum simulator with an unprecedented degree of configurability. Quantum simulators have been realized in ultracold atoms, trapped ions, and superconducting qubits. However, it remains an open challenge to engineer Hamiltonians with access to emergent long-range-ordered phases at low temperatures. Consequently, efforts have focused on clean c-m quantum simulators; one prominent effort has been in moiré or artificial superlattices (SLs) in 2D van der Waals (vdW) heterostructures. However, moiré SLs are limited as a (practical) quantum simulator because the symmetry and functional form of the SL potential are entirely determined by the interlayer interactions and strain present in the twisted structure. For artificial SLs in vdW materials, such as etched holes in a gate dielectric, limits are generally set by nanofabrication techniques. The central aim of this DOE-QIS project is to combine two elements – two-dimensional (2D) vdW heterostructures and electron-beam-programmable substrates – to create a versatile new materials platform ("VR-QuSim") with primary application to quantum simulation, but also in quantum sensing, quantum information science, and materials research in strongly correlated and topological systems. The basic idea is shown in Fig. 1a. A vdW heterostructure is placed on top of a substrate that can be "programmed": an electron beam can be used to write nanostructures in the substrate *through* the vdW heterostructure, so that the vdW + substrate can be assembled first and programmed as a last step. Several candidate programmable substrates are possible; this abstract will specifically discuss our experiments on one type -- insulating ferroelectric thin films of boron-substituted aluminum nitride (AlBN) -- in which the electron beam creates nanoscale domains of electric polarization. The nanostructures in the substrate couple electrostatically to the active vdW layer and modify its electronic properties. As a key example of an advantage of our technique, we can in principle directly program superlattices with a *basis*, such as the Lieb or dice lattice, by writing the multiple sublattices with different electron beam doses (Fig. 2a,b; ref. [1]). A major motivation for this is our recent theoretical work that shows certain superlattice designs for massive Dirac fermions can induce topological transitions from a trivial insulator to C=1 and even C=2 Chern insulators (Fig. 1b, c; ref. [2]) for C_3 and C_4-symmetric SL potentials.

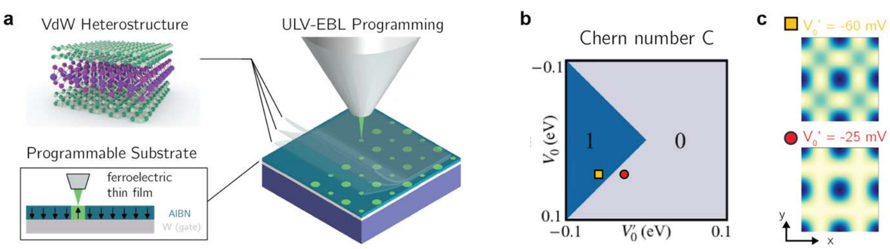

Figure 11. (a) Principle of VR-QuSIM. (b) Topological transition in bipartite superlattice by varying relative strength of sublattice potentials V_0 and V_0'

Here we present our initial experiments on reversing the polarization of a ferroelectric thin film (boron-substituted aluminum nitride, $Al_{1-x}B_xN$, x=0.1) using an electron beam, characterizing the resolution of our technique using piezoforce microscopy (PFM). In a graphene/hBN Hall bar on AlBN substrate, we write half the device with a uniform pattern. This shifts the Dirac point of the patterned half relative to the unpatterned half, and we observe a p-n junction across the interface.

In graphene/hBN devices on unprogrammed AlBN films, we discovered that graphene exhibits magnetoconductance at charge neutrality that is reminiscent of the "quantum Hall topological insulator (QHTI)" phase [3]. We show evidence for quantized helical edge modes, using non-local transport measurements (Fig. 2b). We show that the usual insulating behavior of graphene at charge neutrality in the quantum Hall regime can be restored by heating the sample to 130ºC. This does not alter the mobility of the device nor the visibility of broken-symmetry Landau levels. We interpret this as suggesting the important role of the hBN-AlBN interface via interfacial water molecules and ions that screen the Coulomb interaction.

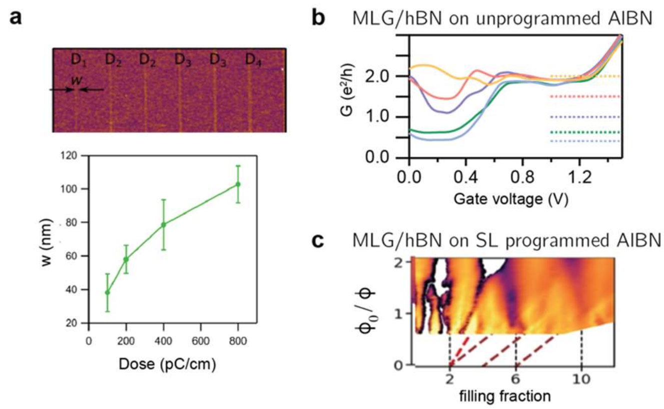

Figure 12. (a) Resolution of e-beam programming of AlBN. (b) Two-terminal (2T) transport of Graphene/hBN on unprogrammed AlBN at 4 Tesla. Dashed lines show expected conductance for different 2T configurations. (c) graphene/hBN on FE programmed with 20 nm superlattice. Color scale proportional to conductance.

We studied four graphene devices with triangular patterns of lattice constants between 10-25 nm programmed into the FE thin film. These devices show preliminary evidence of superlattice effects: Landau levels emerging from replica Dirac points (Fig. 1c).

The most pressing current and future challenges of this work include expanding experiments to other vdW materials such as monolayer transition-metal dichalcogenides (TMDs), in which realization of certain Hamiltonians may be more easily achieved; verification of the ability to program superlattices with a basis or with controlled disorder; and experiments beyond magnetotransport, such as optical or scanned probe measurements.

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Poster Session II

Mapping the genome of coherent quantum defects for Quantum Information Science
DE-SC0022289

Atomic Engineering and Characterization of Targeted Quantum Emitters in 2D Transition Metal Dichalcogenides through Scanning Tunneling Microscopy and First Principles computations

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Abstract: Quantum emitters are a foundational component of quantum information science (QIS) technologies, serving critical roles in quantum networks, quantum sensing, and photon-based quantum computing. Key technical requirements for quantum emitters include the ability to emit identical photons, sufficient brightness, and deterministic emission (ideally emitting one photon per excitation). A central challenge across all quantum emitter platforms is the precise, atomistic control of defect creation, ensuring emitters meet these stringent requirements and enabling the production of identical photons capable of entanglement.

To address the challenge of creating identical color centers, we explore two-dimensional (2D) semiconducting host materials—specifically, monolayer WS_2 and MoS_2—which allow atomically precise defect engineering. These materials also provide direct access to defect structures and their electronic and optical properties with atomic resolution, achieved using state-of-the-art photo scanning tunneling microscopy (STM) and spectroscopy techniques. We have developed three distinct methods to create sulfur vacancies: thermal annealing [1], helium ion bombardment [2], and argon ion bombardment [3]. In the course of exploring these vacancy-creation pathways, we identified sulfur divacancy complex formation and discovered a method to engineer atomically defined one-dimensional defect lines, which host a correlated electron system known as a Tomonaga–Luttinger liquid [3].

To determine optimal substitutional defects to incorporate into these vacancies, we employed theoretical high-throughput modeling to narrow down potential candidate elements [4]. Guided by these theoretical predictions, we have experimentally studied cobalt (Co)[4] substitutions, confirming with STM the theoretical prediction that the cobalt on sulfur substitution is a paramagnetic defect with two levels in the band gap. Such a candidate spin qubit had never been identified experimentally in WS_2. We are currently investigating other defects in WS_2 ytterbium (Yb), silicon (Si), and titanium (Ti). Additionally, we are working on creating ensembles of identical Co defects in WS_2 to measure critical QIS-relevant properties, such as optical emission and coherence times.

Finally, we will present a novel experimental technique—gate-assisted scanning tunneling luminescence—that enables, for the first time, electrically driven exciton creation with atomic-scale precision. This breakthrough allows us to excite individual defects and measure their optical responses directly and precisely at the atomic scale. This capability not only provides an unprecedented direct correlation among structural, electronic, and optical properties of defects in 2D materials, but also significantly accelerates our progress in atomically precise color-center engineering for next-generation quantum emitters.

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Mapping the genome of coherent quantum defects for Quantum Information Science
DE-SC0022289

Long-lived entanglement of a spin-qubit register in silicon photonics

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Abstract: Spin-photon interfaces based on color centers in solids are well-suited as optically interconnected multi-qubit registers for quantum information processing. A series of well-established quantum defects in diamond have shown promise in demonstrating entanglement at long distance. However, scaling diamond-based qubit platforms for quantum applications remains a significant challenge due to fabrication difficulties. The goal of our project is to advance our understanding and quantum control of color centers in silicon, a mature host material for scalable fabrication of qubits.

Among the silicon color centers studied so far, the T center is the only system combining a telecom O-band emission (desirable for quantum communication) with a coherent electron spin qubit in the ground state (desirable as quantum memory). In our project, we focus on advancing our understanding of optical and spin coherence of this defect in nanoscale structures with guidance from first-principles hyperfine interaction calculations. After a brief introduction to earlier work funded by this program on indistinguishable photon generation from a single color center in silicon [1] and nanoscale sensing with T-centers [2], we will present recent progress on demonstrating a multi-qubit register based on T-centers in silicon [3]. We demonstrate the initialization, coherent control, and state readout of a three-qubit register based on the electron spin of a T center coupled to a hydrogen and a silicon nuclear spin. The spin register exhibits long spin echo coherence times of 0.41(2) ms for the electron spin, 112(12) ms for the hydrogen nuclear spin, and 67(7) ms for the silicon nuclear spin. We use nuclear-nuclear two-qubit gates to generate entanglement between the two nuclear spins with a fidelity of F=0.77(3) and a coherence time of T^{2*}=2.60(8) ms. Our results show that T centers can realize a long-lived multi-qubit register with an optical interface in silicon photonics. We conclude by discussing ongoing work on improving qubit readout fidelities for T-centers in nanoscale devices, understanding electronic excited state dynamics under electric fields and modeling through first principles spin bath interactions and coherence times in silicon.

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Mapping the genome of coherent quantum defects for Quantum Information Science
DE-SC0022289

Sharp emission and fine structure splitting from localized excitons in Vanadium-doped monolayer WSe_2

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[1]Lawrence Berkeley National Laboratory, [2] Rice University, [3] National University of Singapore, [4] National Institute for Materials Science

Abstract: Two-dimensional transition metal dichalcogenide (TMDC) semiconductors have attracted attention for light-matter interaction due to their strongly bound excitons arising from reduced dielectric screening and quantum confinement. Point-like defects in 2D materials hold promises as individually accessible sources of single photon emission and as a spin qubits in the same material. Our previous study has demonstrated that neutral Cobalt substitution on the sulphur site (Co_S^0) in WS_2 is a promising spin-photon interface for quantum communication[1].

The more established and easier to synthesis Vanadium substitutional defect in WSe_2 (V:WSe_2) has shown a broad below-gap emission associated with this defect in addition to evidence of long-range magnetism. In this work, we encapsulate high quality V:WSe_2 in hBN which allows us to see multiple PL emission peaks (linewidths < 1 meV) that have so far been hidden under a single broad emission. We attribute these peaks to recombination of dark excitons bound to V_W^{-1} site. By examining the magnetic field dependent photoluminescence, we observe a Zeeman splitting yielding g-factors between 8.4 to 9.7, as well as a zero-field, fine structure splitting of about 0.9 meV in one of the peaks. We calculate the electronic structure associated with the observed defect states using Density Functional Theory and time-dependent density functional theory. These calculations help us identify the nature of the observed transition, describe the electronic structure of the defect and rationalize the magnetic field interaction. The computations and transmission electron microscopy suggests that complex defects that could be attractive emitters in the telecom might be as well present in the sample. We also discuss the potential for Fermi level tuning to prepare the defect in a neutral charge, triplet state, that would offer a spin degree of freedom and current experimental efforts in that direction.

1. Thomas, J. C. *et al.* A substitutional quantum defect in WS2 discovered by high-throughput computational screening and fabricated by site-selective STM manipulation. *Nat Commun* **15**, 3556 (2024).

Project Title: Seeking quasiparticles in perturbed matter from low-energy spin dynamics

Award Number: DE-SC0022986

Names of PI, coPI(s), unpaid collaborators: **Arnab Banerjee**, Catherine Pappas, and Roger Pynn

Affiliation(s): Department of Physics and Astronomy, Purdue University, West Lafayette IN. Department of Physics, Technische Universitaet (TU) Delft, Netherlands. Department of Physics, IU Bloomington, Indiana.

Abstract: In this project we set up a platform and workflow that allows us to directly probe exotic states, including those predicted by the near-term analog quantum simulators and quantum computers, their statistics and effects of spin quantization in the low energy spectrum using state-of-the-art neutron sample environments and novel bulk approaches. The project builds upon a successful set of developments in the past three years spanning several frustrated spin systems as well as development of neutron in-situ probes. We have continues exploring the enigmatic states of Kitaev candidate a-$RuCl_3$ while introducing new thallium-based delafossites $TlYb(S,Se)_2$ and 1-6-6 Kagome metal RCr_6Ge_6 (R=Tb, Yb). In $TlYbSe_2$ we discover a quantum disordered ground state to very low temperatures, where a possible spin liquid state is punctuated by a spin glass state from free Yb spins[1]. A likely breakdown of the Wiederman-Franz law in $YbCr_6Ge_6$ due to interactions between fluctuating spins and itinerant electrons that can pave the path to understand the effects of metallic gating of frustrated spin systems. In Shastry-Sutherland magnet HoB_4, we discover new phases and transitions, highlighting the importance of defects in an Ising model[2]. Finally, in $RuCl_3$, a careful neutron measurement of the 8-9 T spin gap reveals a 9 T discontinuity and an overall gap evolution amenable support a distinct topological phase.

In the ensuing research, we develop ferromagnetic neutron spin echo spectrometry, and high field quenched magnets, as well as predictions based on quantum computers by our team, to perform a set of experiments that can help with identification of exotic magnetic states of matter with no classical analog. We desire to confirm the predictions of emergent magnetic quasiparticles, which can potentially replace electrons as the building blocks for decoherence-resistant quantum electronics. Neutron scattering, magnetic susceptibility and thermal transport, mated to in-situ and in-operando techniques for electric currents and magnetic fields, in both bulk and mesoscale systems, offers unique advantages for the discovery and study of magnetic quasiparticles. Here we use a three-pronged approach: (1) We use of low energy spectroscopy techniques, including ferromagnetic neutron spin echo[3], backscattering and other Larmor techniques to measure low energy excitations to directly probe exotic spin statistics in quantum magnets and spin liquid candidates such as $TlYbSe_2$ and $RuCl_3$ to validate quasiparticle levels; (2) We leverage the perturbation of the ground and excited states fast quenched magnetic fields to understand emergence of new quantum ordering phenomena in transverse field Ising magnets (such as TmB_4), leveraging the progress and validating our predictions obtained from quantum

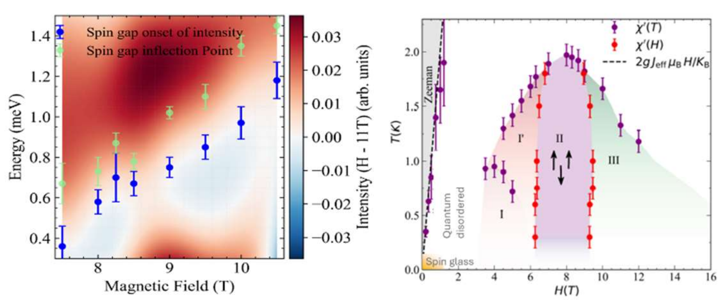

Fig. (Left) The evolution of the low energy spectral intensity in $RuCl_3$ shows a discontinuity at 9 T in both the scattering and the spin gap. (Right) The in-field phase diagram of $TlYbSe_2$ with a spin-glass below 35 mK and a quantum disordered state [1].

computers such as D-Wave[4], QuEra and Google for prediction of new quasiparticle states; (3) We clarify the interaction between itinerant electrons and frustrated magnetic states in RCr_6Ge_6 (R=Rare Earth) which could be critical to understand gating of magnetic devices, in select materials where both co-exist. We expect to probe the neutron spectrum of these magnets as a function of changing in-operando and in-situ conditions – with changing magnetic flux and drive voltages. Our framework to cross-understand and cross-validate magnetic excitations both using quantum computation and real experiments allows us to prepare towards a future where exotic magnetic materials could be tuned towards a desired functionality based on inputs from quantum computer simulations.

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