

Center for Molecular Magnetic Quantum Materials (M²QM)
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Mission Statement: *To provide the materials physics and chemistry understanding of molecular magnetic quantum materials essential for quantum and conventional computing beyond Moore's Law.*

Molecular magnetic materials (MMMs) are quantum materials, often with multiple exotic physical properties arising from the quantum spin states of the molecules and their coupling to the external environment and fields. The future of molecular magnetic systems (MMSs) as quantum material technology candidates depends critically on three fundamental scientific objectives: (i) mastering the chemistry of quantum magnets at the molecular scale in order to design and control all aspects of the energy states and their dissipation and dynamics; (ii) achieving the ability to design and make such materials as stable aggregates (crystals, adsorbates, nanostructures, etc.) while preserving or even enhancing their key physical properties; (iii) developing methods to interrogate and reversibly switch their quantum states (e.g. spin states). Making major advances towards these three critical capabilities is the primary goal for the Center for Molecular Magnetic Quantum Materials – M²QM.

With an overarching goal of *turning molecular magnets into quantum materials useful for both quantum computing and quantum current conventional devices*, M²QM aims to provide the materials physics and chemistry understanding needed to get beyond Moore's Law, *but with a failsafe strategic twist*. The Center's approach is equally targeted to both quantum devices (qubits) and quantum-current (e.g., spin) systems. For qubits, the MMS approach gives rigorous mono-dispersity (identical molecular magnets), enables genuine bottom-up assembly (including self-assembly) into complex aggregated systems, and enables electronic control and detection of quantum magnetic states by diverse means. For quantum currents, MMSs have inherently stronger coupling to the lattice than inorganic competitors (multiferroic oxides, transition-metal- dichalcogenides, etc). This allows the magnetic state to be sensed and manipulated more easily with low-energy electric or strain fields.

In the specific setting of this EFRC, molecular magnetic systems (MMSs) are molecular and nano-structured systems, including single molecule magnets, molecular-metal complexes, metal-organic quantum magnets, and counterpart multiferroics. MMSs exhibit immense spin configuration richness, spin crossover, or spin-state transition, all of which offer new ways to achieve multiferroic functionality. The distinction from materials phenomena rooted in the familiar quantum mechanical underpinnings of structure, bonding, and quasi-particle response illuminates the opportunity and challenge posed by quantum materials. The materials physics and chemistry must be right in two very distinct but intertwined ways: making a stable material from promising molecular constituents *and simultaneously enabling the emergence of the target phenomenon or phenomena (rather than causing its suppression)*. Tasks in M²QM are organized into three scientific thrusts as 1) Magneto-electric couplings in Quantum Materials, 2) Quantum Superposition and Entanglement of Molecular Spin States, 3) Spin-Charge Coupling on surfaces and interfaces, and a cross-cutting fundamental theory effort.

The "bottom-up" molecular/nanoscale approach (molecules → MMSs) to construction of spin logic devices long has been viewed as advantageous. Molecular structure can be designed to control low-energy electron/nuclear spin states. Indeed synthetic chemists now can design molecules with desired quantum states. As they are molecules, each is a precise replica of all others. Such replication obviously is critical for eventual scaling up to quantum devices with predictable performance. Templated substrates

or scaffolds (such as polymers or nucleic acids) can attach and organize functionalized molecules, thereby providing routes towards development of quantum circuits. Other functionality can be built in, e.g., optically active transitions to provide fast sensing of the single-molecule electron spin state. Although information encoding and read-out can be done with the nuclear spin states of an individual molecule, that is inherently slow because of the low nuclear Larmor frequency in terrestrial magnetic fields. Thus electron spin qubits are vastly preferable. Other advantages of molecular electron-spin qubits versus some other qubit architectures are their relatively small size, monodispersity (uniform size), and the possibility of self-assembly into ordered crystalline ensembles. These advantages have allowed discovery of nebulous and fragile quantum properties in nanomagnetism that were difficult, if not impossible, to identify with traditional “top-down” magnetic nanoparticles. Those properties include quantum tunneling of the magnetization vector (QTM), followed by discoveries (involving members of this EFRC team) of exchange-biased QTM (EBQTM), quantum superposition states and entanglement (QSE), spin-spin cross relaxation (SSCR), and others. EBQTM, QSE, and SSCR were discovered in a hydrogen-bonded dimer of Mn_4 single-molecule magnets (SMMs) with spin $S = 9/2$ ground states.

The monodispersity provided by bottom-up synthesis also enables very precise spectroscopic characterization of both the static and dynamic quantum properties of assemblies of magnetic molecules, something central to M^2QM . Beyond control over the physics of individual qubits, supramolecular design principles allow for connectivity between magnetic molecules and the incorporation of additional functionalities, such as optically active components that provide a means to switch and/or sense spin states at the individual molecule level. Other advantages include solution syntheses at or near room temperature, solubility in common solvents; formation of molecular crystals and structural characterization (to atomic precision) by single-crystal X-ray diffraction; and monolayer shells of organic ligands that can be varied conveniently to allow isotopic labeling (2H , ^{19}F , etc). Additional merits include modulation of redox properties, crystal symmetry, and other properties.

M^2QM research will focus on the following topics: How intermolecular coupling changes structure and spin states of molecules via substitutions of ligands and linkers, and how such coupling is affected by pressure and fields; effect of coupling to substrate on the structure and magnetism of molecules and effect of molecules on the substrate such as the formation and variation of the Schottky barrier; spin state detection and manipulation; multiferroicity in molecular crystals manifested as coupling between spin state transition, strain, and charge polarization, in particular the magneto-electric effect; non-equilibrium dynamics in spin-phonon coupling; and effects of strong interaction and extreme fields. All studies will be carried out using combinations of experiment, theory, and computation.

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