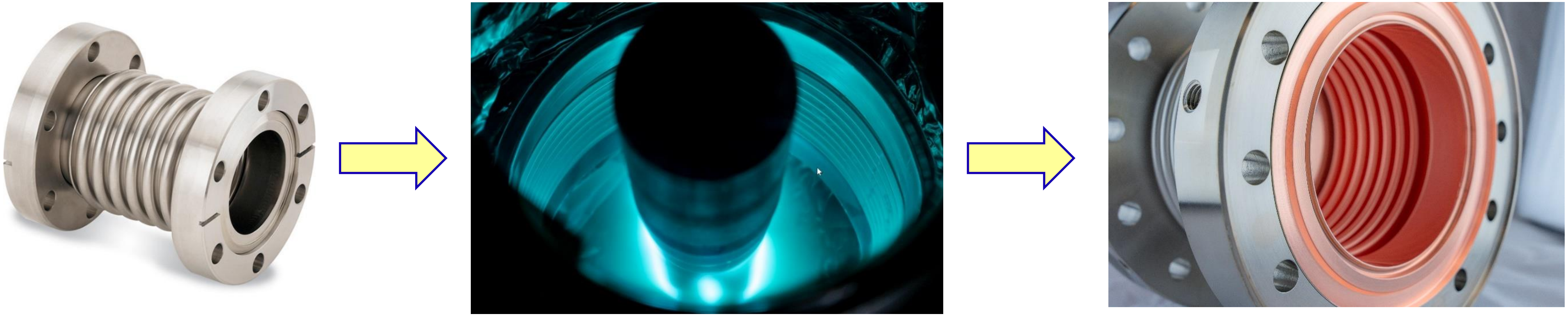


High-Quality, Conformal Bellows Coatings Using Ultra-Fast HiPIMS with Precision Ion Energy Control

starfire industries

2023 DOE-ONP SBIR-STTR EXCHANGE MEETING

About Starfire Industries LLC

Champaign, IL USA (near the University of Illinois)

- Vertical integration from R&D, manufacturing, applications testing and customer support
- "Deep Tech" nuclear, plasma and radiological team

Particle Accelerator Solutions:

- **nGen® portable neutron generators**
- **Centurion® ultra-compact MeV particle accelerators**

Plasma Processing Solutions:

- **IMPULSE® pulsed power modules for sputter/etch**
- **RADION™ microwave plasma sources for PECVD/etch**

Two Business Groups Within One Organization

Products on 6 Continents + Space!

Patent Portfolio Across Products

Facility Expansion

Starfire has recently relocated all operations to a 194,000 sq.ft. facility in Champaign, IL

Design/Engineering

R&D Prototyping

Manufacturing/Production

Warehouse/Shipping

Coatings & Materials Processing

Particle Accelerator Testing/Integration

Starfire’s Product Families

PARTICLE ACCELERATOR SOLUTIONS

nGen®

- Build/Sell Sealed Neutron Generators
- Design/Build Hardware For Custom OEM Solutions
- nGen® Systems Solutions For End-Users

Centurion®

- Build/Sell LINACS
- Custom OEM Sales To Development Partners
- Protons/Neutrons “As A Service”

PLASMA SOURCE SOLUTIONS

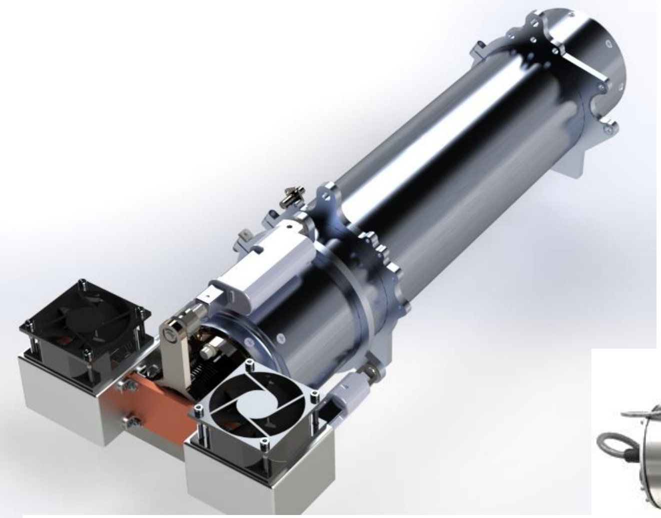
IMPULSE®

- Build/Sell Pulse Modules
- Design/Build Systems Using IMPULSE®
- Coatings “As A Service”

RADION™

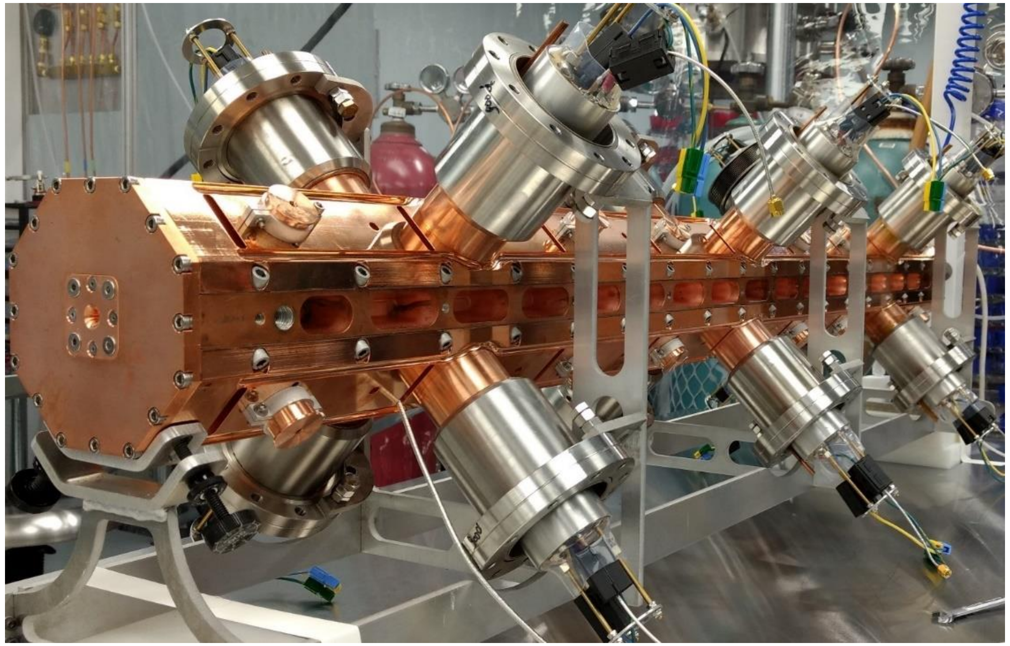
- Build Microwave Plasma Sources
- Atmospheric Coating System Solutions
- Vacuum Etching/PECVD Systems Solutions

Particle Accelerator Solutions



Patented nGen®
Portable Sealed
Neutron Generators
w/Emission On End

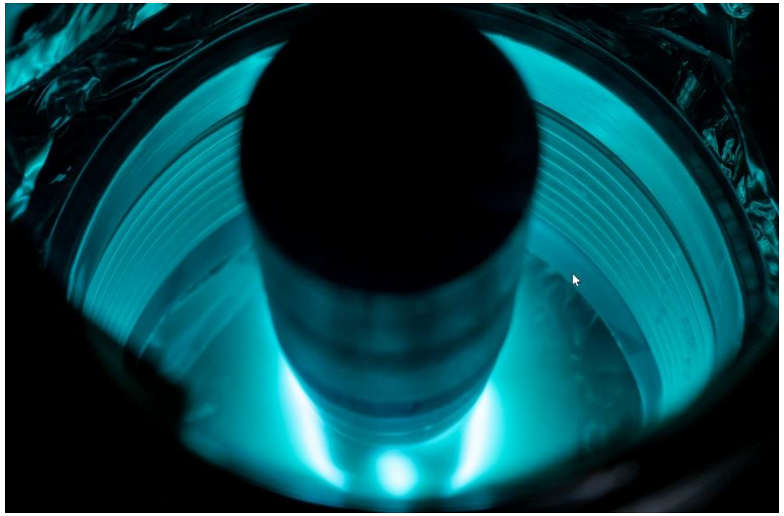
Patented Centurion®
Ultra-Compact RFQ
Linear Accelerators
For Protons &
Deuterons



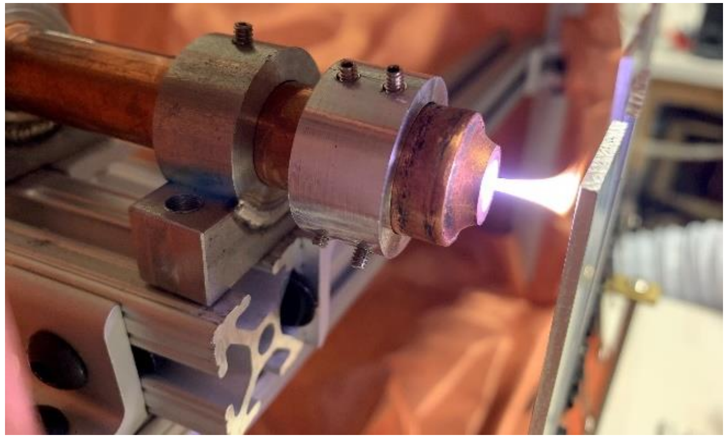
Plasma Source Solutions

Patented IMPULSE® + Positive Kick™
Next-Generation Thin-Film HiPIMS PVD/Etch

Patented RADION™ Microwave Plasma Sources



Pat. Pend. Radial & Inverted Cylindrical Magnetron Systems



Pat. Pend. Atmospheric Cold Plasma Jet Coatings

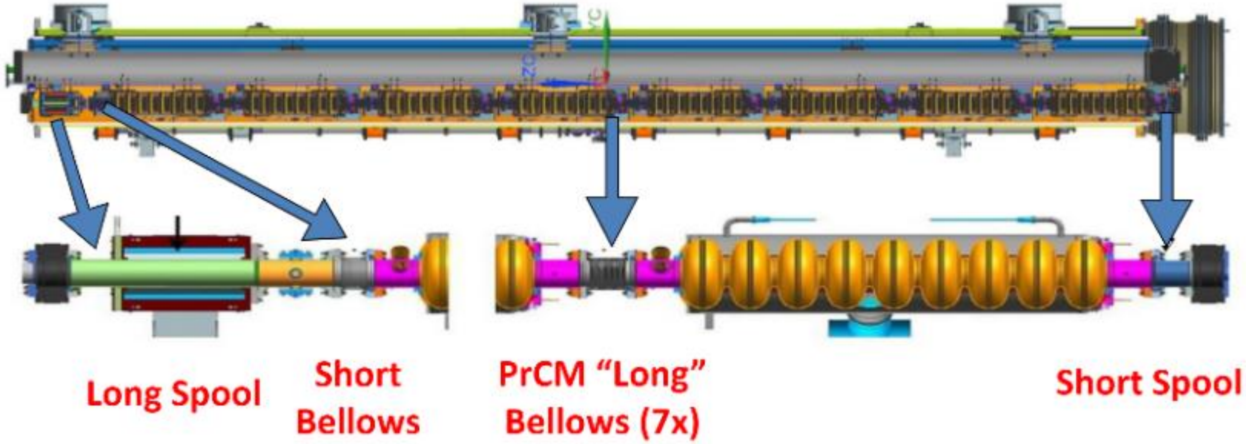
The Problem

Modern particle accelerators employ numerous specialized components requiring metal films

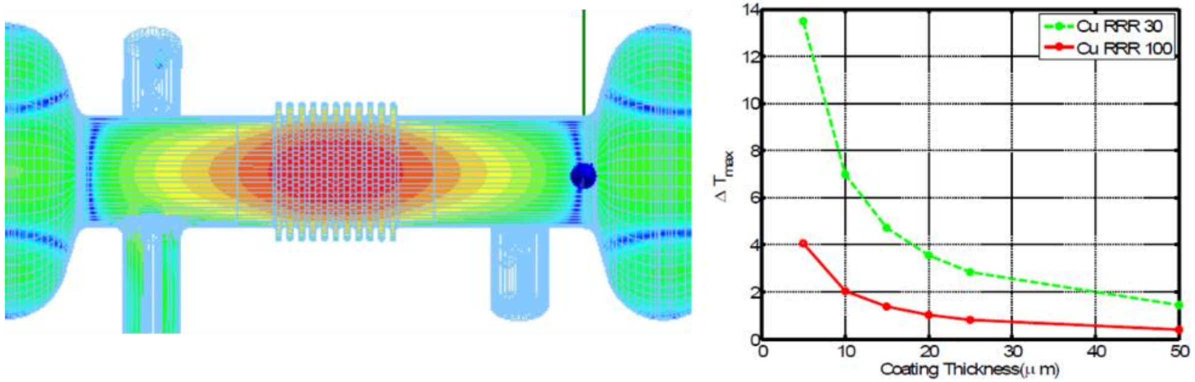
Bellows sections are a great example (and the focus of this work)

- Want high electrical conductivity on the inner surface for low beam-losses
- … & low thermal conductivity in the bulk for low thermal losses

So, a stainless-steel bellows w/ a copper film on the inner surface solves this problem



Some of the LCLS-II cryomodule bellows and spool sections that see RF energy and beam. Additionally, there are many other feedthroughs and bellows on the accelerator platform that are plated and material controlled. Figure from Ref [1].



(left) A simulated trapped RF mode that exists within the bellows that couples two cryomodules (Figure from Ref. [2]). (right) Calculated maximum rise in temperature as a function of layer thicknesses for Cu layers having RRR values of 30 and 100 (Figure from Ref. [3]).

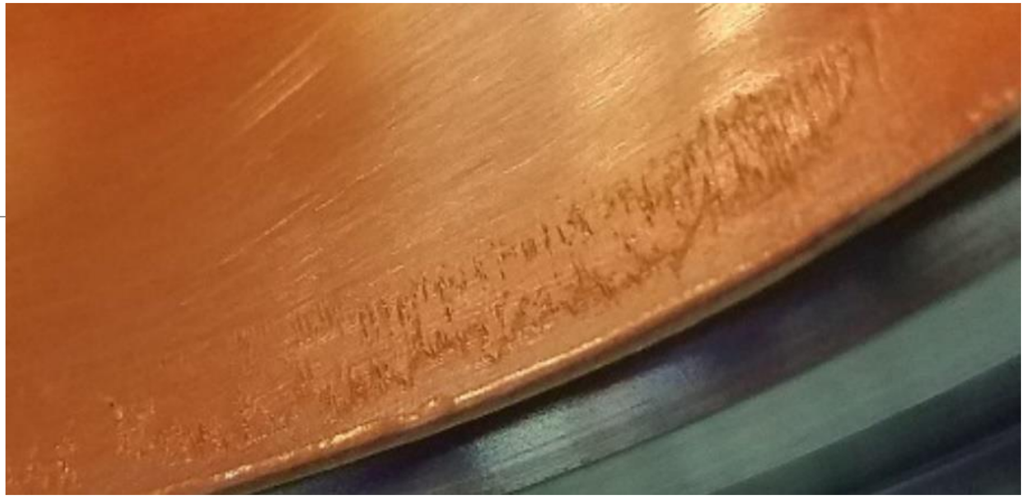
[1]: K. Wilson et al., "Production of Copper-Plated Beamline Bellows and Spools for LCLS-II," in 8th International Particle Accelerator Conference (IPAC 2017), Copenhagen, Denmark, 2017.
[2]: A. Saini et al., "RF Losses in 1.3 GHz Cryomodule of The LCLS-II Superconducting CW Linac," in 28th International Linear Accelerator Conference (LINAC16) , East Lansing, Michigan, 2017.
[3]: A. Saini et al., "LCLS-II TECHNICAL NOTES: Temperature Rise in LCLS-II Cavity Bellows," 29 June 2015. [Online; Accessed 01 October 2019].

What is done now?

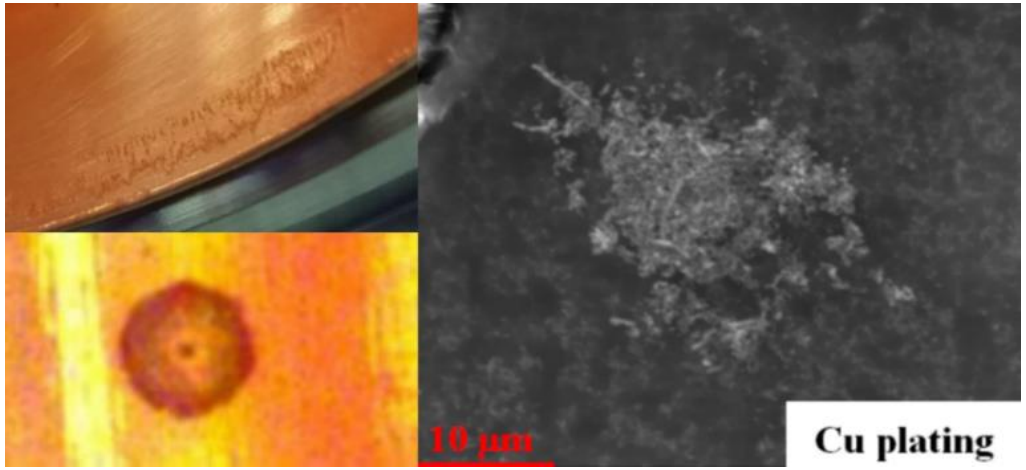
Copper films on stainless-steel bellows are presently deposited via a ‘wet’ electrochemical plating process

This has some inherent limitations/drawbacks:

- Defects/inclusions, flaking, etc. can lead to sparking or increased power deposition
- Hazardous waste streams are generated by these processes
  - These processes are being legislatively phased out in the EU where possible (e.g. where alternatives exist)
  - The US may be soon to follow
- Difficulty sourcing parts
  - Years of attenuation have left only a small handful companies in the US that perform such coatings
  - Infrequent orders result in plating shops having to re-learn some of the techniques and consideration that are lost with personnel turnover



A photograph showing striations in plating attributed to a leak in a seal during the plating process (Figure from Ref. [1]).



Images showing striations in plating (upper left, from Ref. [1]), a possible inclusion (lower left , from Ref. [1]), and a Cu plating particle (right, from Ref. [3]), plus distribution of particulate types by size Ref. [3].

[1]: K. Wilson et al., "Production of Copper-Plated Beamline Bellows and Spools for LCLS-II," in 8th International Particle Accelerator Conference (IPAC 2017), Copenhagen, Denmark, 2017.
[4]: L. Zhao et al., "Study on Cleaning of Copper Plated Bellows for LCLS-II," in 29th International Linear Accelerator Conference (LINAC18) , Beijing, China, 2018.

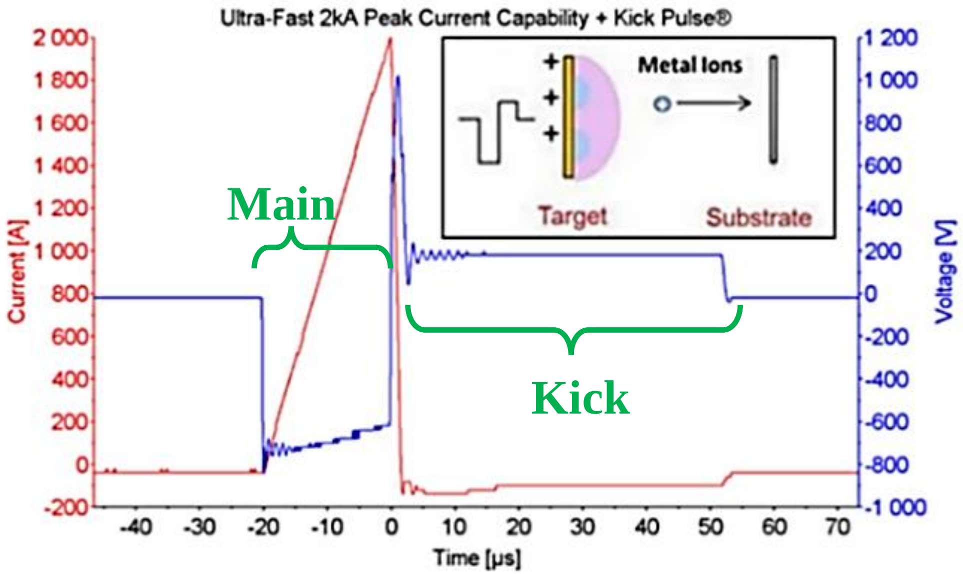
High-Power Impulse Magnetron Sputtering (HiPIMS)

HiPIMS involves driving a sputtering magnetron with high-intensity pulses (typ. $\gtrsim 1\ \mathrm{A/cm^2}$)

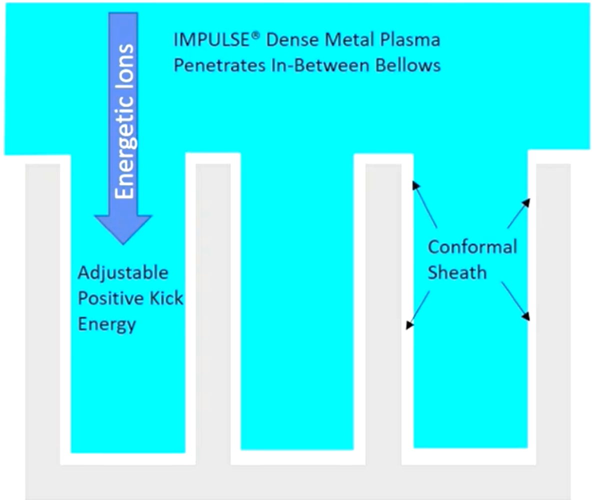
- Pulse structure allows for plasma density and ion energy to be effectively decoupled
- HiPIMS pulse parameters can be adjusted to achieve the desired plasma and film characteristics

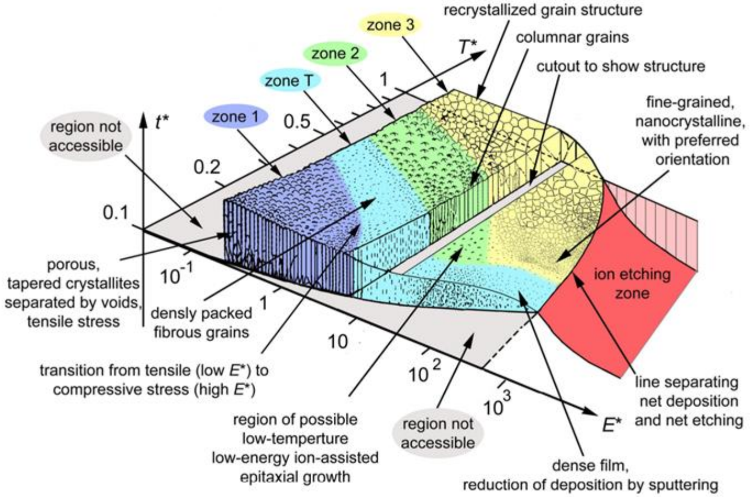
We can use our IMPULSE® w/ Positive Kick™ to achieve adherent, dense, conformal Cu films

- However, a custom magnetron is required for this geometry



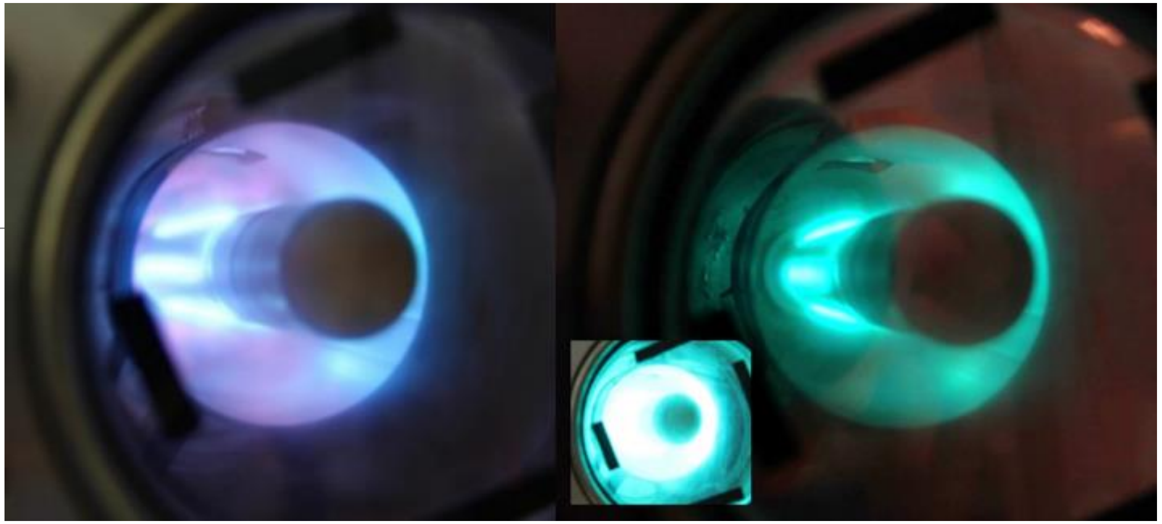
A set of prototypical voltage and current waveforms produced by the IMPULSE®, with the main- and kick phases labelled.





Patented IMPULSE® + Positive Kick™

Radial Magnetron


The radial magnetron shown during an in-situ plasma clean step (left) and during a Cu deposition (right) with a reduced exposure time setting on the camera. The inset image is of the same Cu deposition step without any reduction in exposure time.

A plasma generated on the OD of a Cu10100 cylinder sputters material radially outward

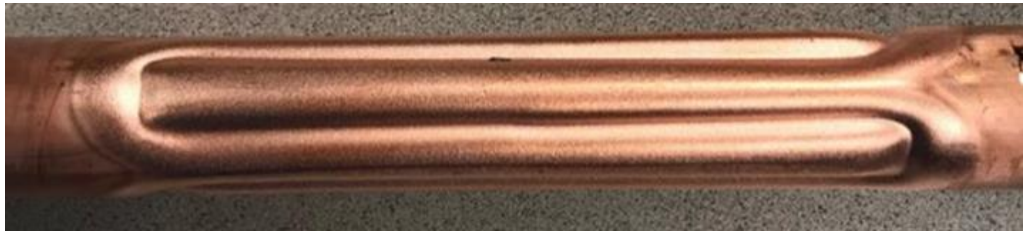
- HiPIMS w/ Positive Kick™ allows for both conformal Cu deposition AND *in-situ* plasma cleaning/etching

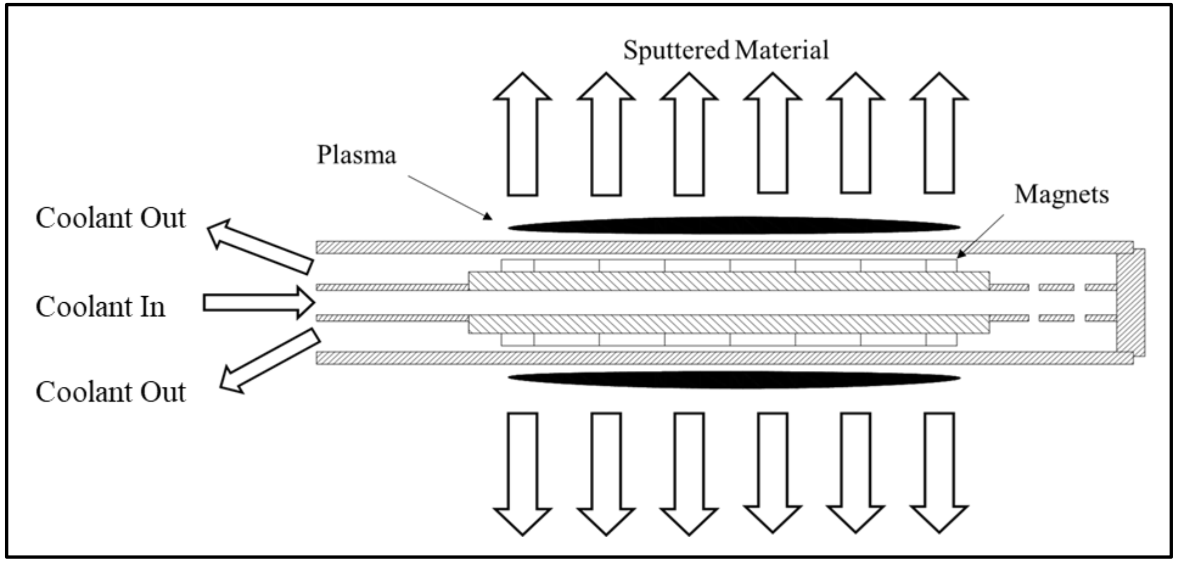
Designed to have a single, serpentine racetrack

- A single racetrack ensures plasma uniformity
- The 1” OD design incorporates 6 passes down the length of the magnetron

HV and cooling are at one end of the magnetron

- Facilitates easy loading/unloading of cylindrical parts at the other end


Erosion pattern produced by the serpentine magnet pack



A cross-sectioned drawing of the radial magnetron design used for this work.

Target Erosion and Utilization

An internal rotating magnet pack offers several advantages

Hard electrical contact with the cathode for HiPIMS pulses

- Stationary cathode → No brushed electrical contacts
  - > 1 kA easily achievable
- A stationary design also minimizes particle generation

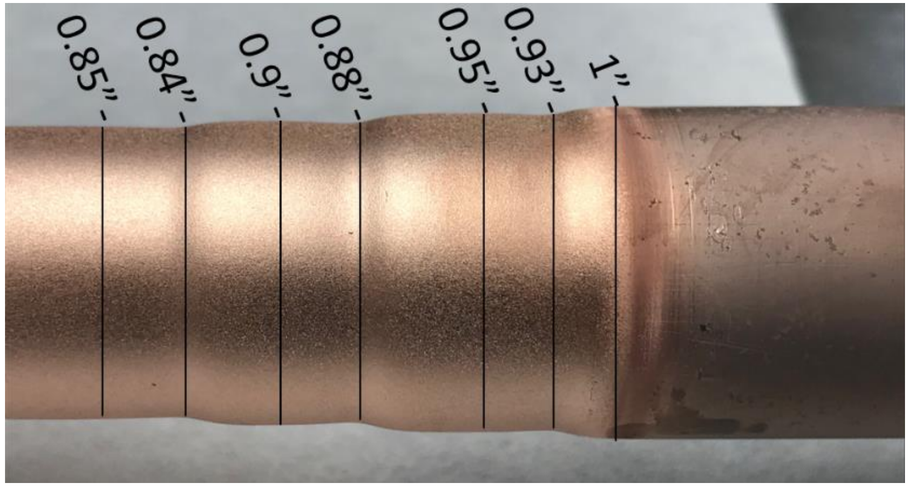
Target utilization is estimated to be $\gtrsim$ 90% over the active region

- Erosion from the stationary pack was observed to be greatest at the turn-around regions
- The turn-around regions for the rotating design were staggered
  - The resulting erosion pattern is extremely uniform

Aging effects are minimal

- There is virtually no change in the surface geometry during operation
- As the surface erodes and the target thins, the surface B-field does increase slightly; the effect on operation is small

Erosion patterns for the stationary (top) and rotating (bottom) serpentine magnet packs.



Close-up view of the erosion at the end of the rotating design. Measurements of the target diameter are indicated.

Substrates and Deposition Conditions

Three types of substrates were used for these experiments:

- 316 stainless steel coupons, LCLS-II bellows sections, and full bellows assemblies

Depositions were typically split up into 4 phases:

1. *In-situ* plasma clean/etch (~10—20 minutes)
    - A short (1 μs) negative pulse strikes a plasma
    - A high-voltage (~400 V) kick pulse is applied to etch the surface of the substrate
2. Cu implantation/intermixing (~5 minutes)
    - The negative pulse is lengthened to rarefy the plasma
    - A high- to moderate-voltage kick pulse is applied to drive Cu ions into the surface of the substrate
3. Transition layer (~5 minutes)
    - Pulse settings are transitioned from the previous step to the next step
4. Cu thick film (dep. at ~ 100 nm/min)
    - Low- to moderate-voltage kick pulse for adatom mobility
    - Pulse rep. frequency and peak current are adjusted for film property control

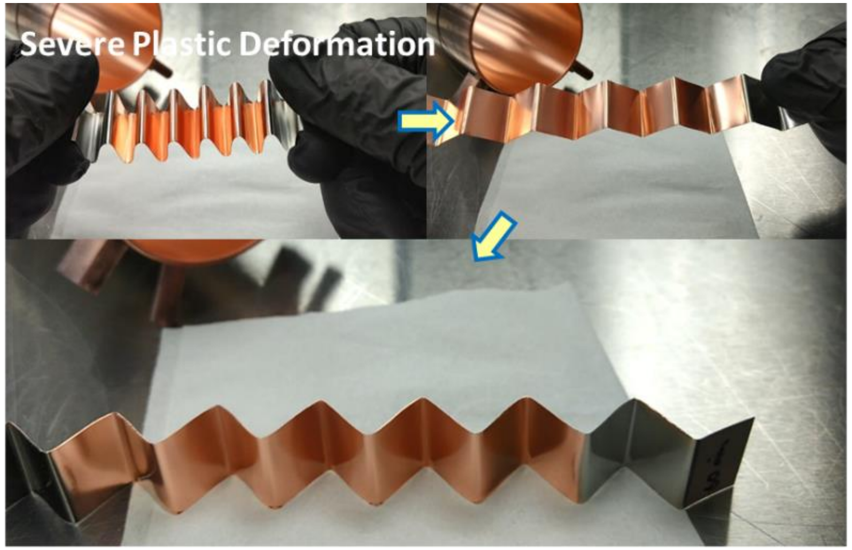
The sample holder used for this work loaded with a corrugated test coupon. (Inset) An LCLS-II bellows section.

The magnetron and test chamber used for this work.

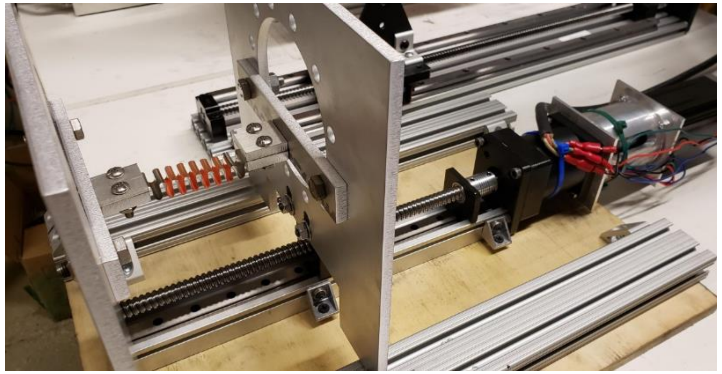
Results – Coupons and LCLS-II Bellows Sections

Adherent, thick (3—30 µm), conformal copper films were achieved on both test coupons and full bellows assemblies

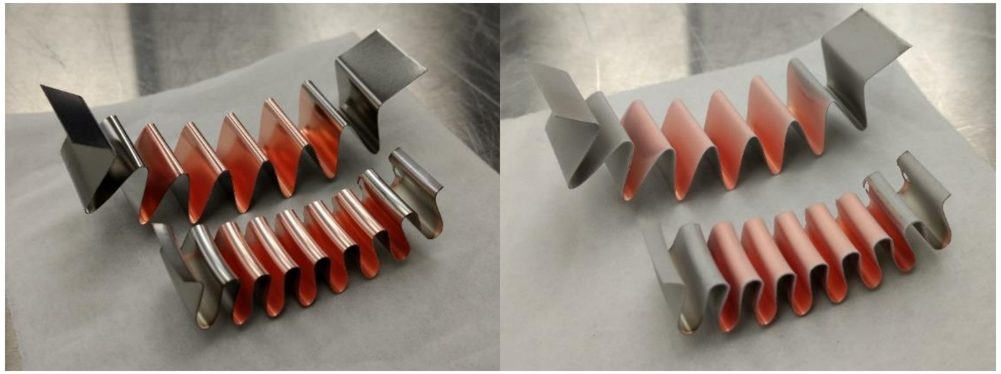
- Even after severe plastic deformation, the deposited films were not observed to buckle or delaminate
- Films did not delaminate after vacuum baking to 400 C and subsequent cooling to 77 K
- Fatigue testing had no observable effects
  - 2,000 cycles at ± 6 mm stroke, as per the LCLS-II bellows specifications
- A custom conductivity meter was used to record IACS conductivities
  - Values in the range of 70—90% IACS were typical



A test coupon with a 5 µm Cu film being plastically deformed. No buckling or delamination was observed in the Cu film.



The fatigue testing apparatus used for this work.



The test pieces after being subjected to a 400 C vacuum bake (left) and a 77 K LN2 bath (right).

Initial Results – Coated Bellows Assemblies

Depositions on full bellows assemblies were also performed

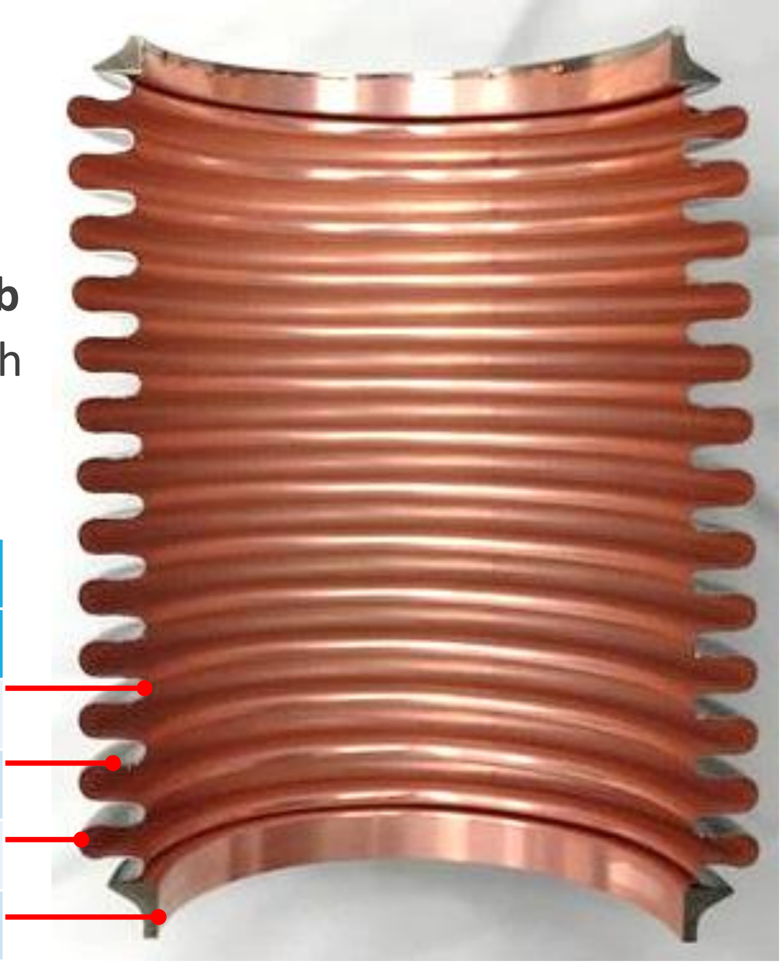
- The resulting films (~20—30 µm) were adherent, conductive, and conformal
- As with the coupons, fatigue testing resulted in no observable changes

RRR values were measured at various places along the bellows at Jefferson Lab

- Bellows were sectioned by wire-EDM and sections of film were pulled off with tweezers for RRR measurement

A Cu-coated bellows.

Location	RRR	
	4 mTorr	20 mTorr
Apex	14.33	22.93
Wall	9.53	23.42
Trough	6.07	6.74
Flat	10.35	24.86



A cross sectioned piece of a Cu-coated bellows (right). The apparent delamination at the top is where Cu was intentionally peeled off for RRR measurements. Measurements of the RRR for two processes (made at the indicated locations) are given here as well (left)

HiPIMS Parameter Optimization

The parameter optimization consisted of

- Selecting the best process from the earlier exploration
    - ‘Best’ here is a somewhat subjective mix of RRR- and film-thickness considerations
- Repeatedly expanding around that process with a series of depositions, selecting the best, and repeating

Series A—C were expansions around the previous best set of results

Series D was a set of reproducibility studies

- Used to confirm that the resulting process is repeatable, even after magnetron aging

Thus far, the optimal process conditions results in an adequate RRR

- Max/min film thickness is a little larger than we would like
- Efforts to improve that are underway with the CEBAF waveguide deposition efforts

Set Number	Main Width (us)	Peak Current (A)	Kick Voltage (V)	Pressure (mTorr)
A2	10	180	65	19
A3	10	380	55	19
A4	10	380	55	34
A5	20	380	55	19
A6	5	380	55	34
B1	10	380	120	34
B2	10	380	30	34
B3	10	540	55	34
B4	10	380	55	50
B5	15	380	55	34
B6	15	600	55	50
C1	10	380	10	34
C2	10	380	20	34
C3	10	380	30	34
C4	10	280	30	34
C5	10	440	30	34
C6	10	380	30	28
D1	10	280	30	32
D2	10	280	30	32

(Left) A photograph of a coated bellows section. (Right) A table of relevant process parameters for this optimization process.

Set Number	Main Width (us)	Peak Current (A)	Kick Voltage (V)	Pressure (mTorr)	Delamination	Thickness (Peak) (um)	Thickness (Wall) (um)	Thickness (Trough) (um)	RRR Peak	RRR Trough
Optimal	10	380	30	34	Major	42	13	20	44.04	42.68

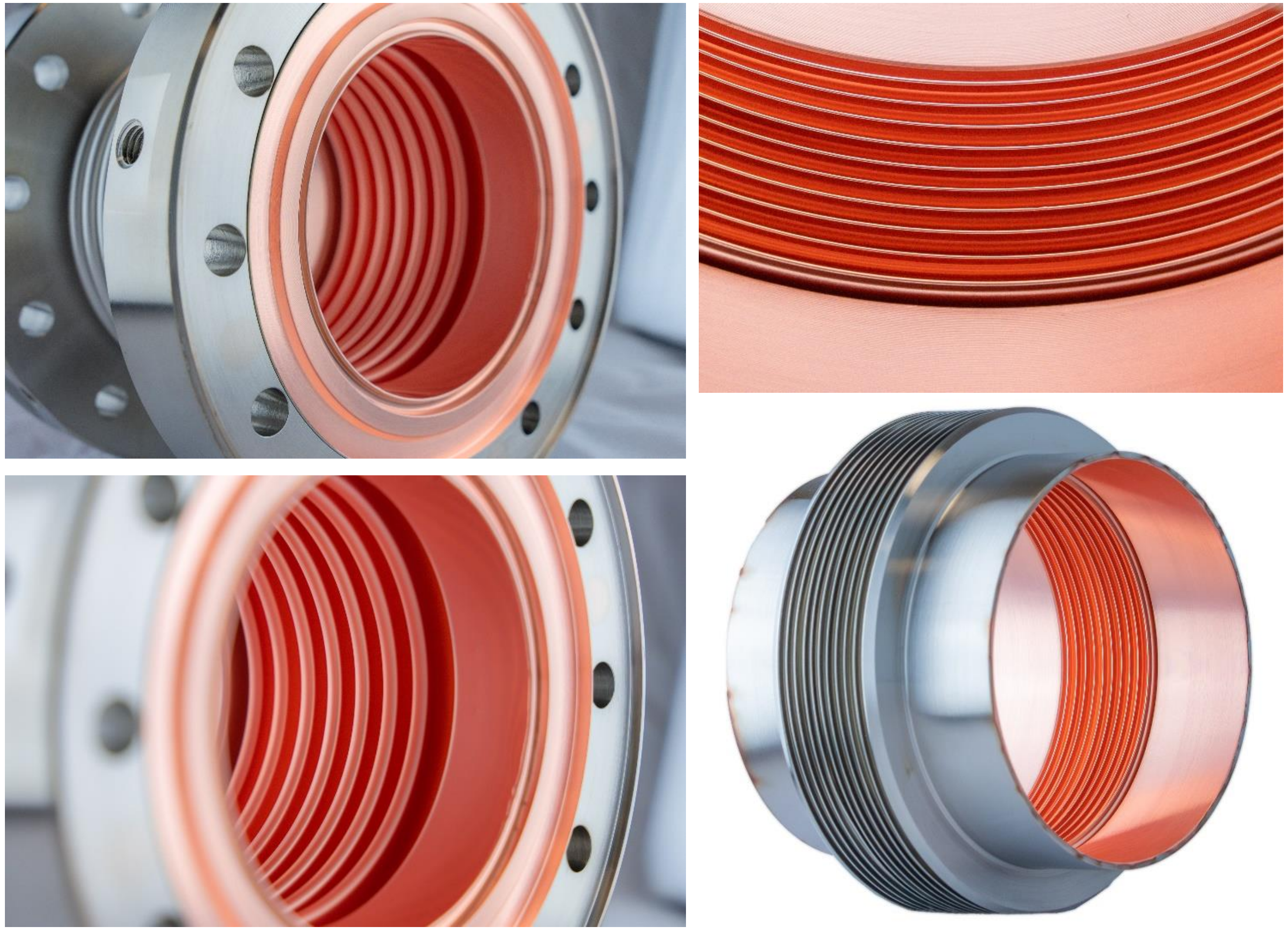
Cu-Coated Bellows

Stainless steel bellows were coated in Cu

- Optimum process conditions discussed on the previous slide were used for these depositions
- Both hydroformed and edge-welded bellows were coated
  - The edge-welded geometry makes this a more difficult deposition than for the hydroformed bellows

A rejected LCLS-II bellows (supplied by JLab) has been coated and returned to JLab for analysis

- This sample will be subjected to the same processing that an electroplated bellows would undergo



(Left) Photographs of a Cu-coated LCLS-II bellows section, which is presently being evaluated at JLab . (Right) Photographs of a Cu-coated edge-welded bellows.

A Smaller (0.5”-OD) Radial Magnetron

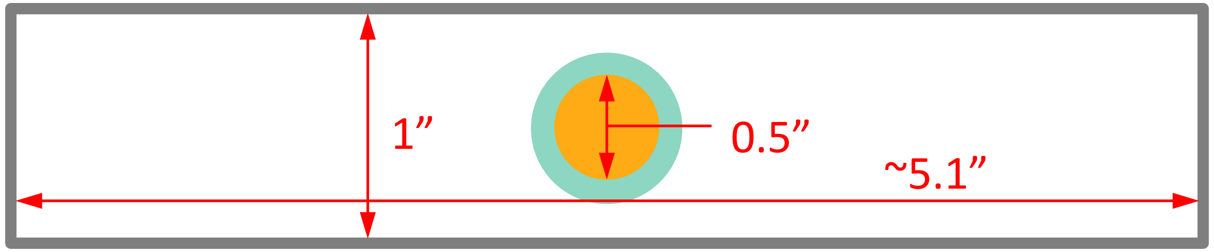
The CEBAF waveguide structure presents a much more difficult problem

- Low minimum internal clearance (1”)
- High aspect ratio (> 5:1)

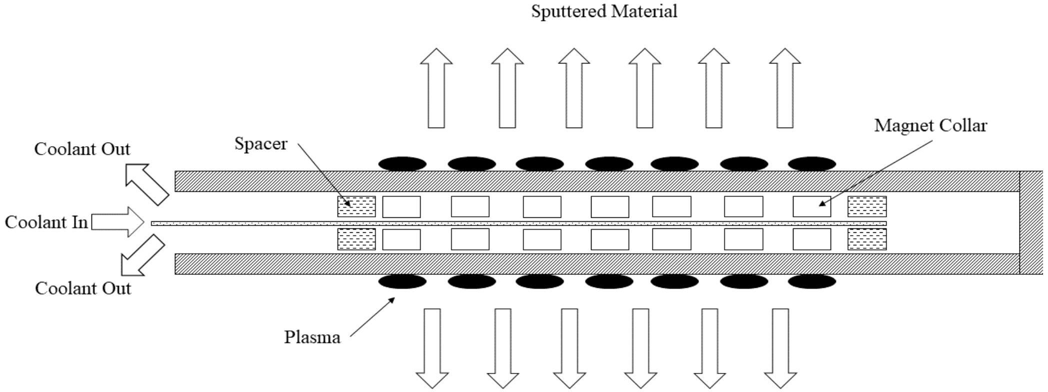
Required a new, smaller magnetron

- Same single-ended design as the 1”-OD version
- Plasma racetrack changed from a single serpentine racetrack to an array of annular (azimuthal) racetracks

We very quickly decided to make the jump to a dual-magnetron configuration to combat the challenging aspect ratio



The low internal clearance and high aspect ratio of the CEBAF waveguide structure present difficult technical challenges.



A cross-sectioned drawing of the 0.5”-OD radial magnetron design used for this work.

The prototype 0.5”-OD magnetron, which was made with an aluminum target.

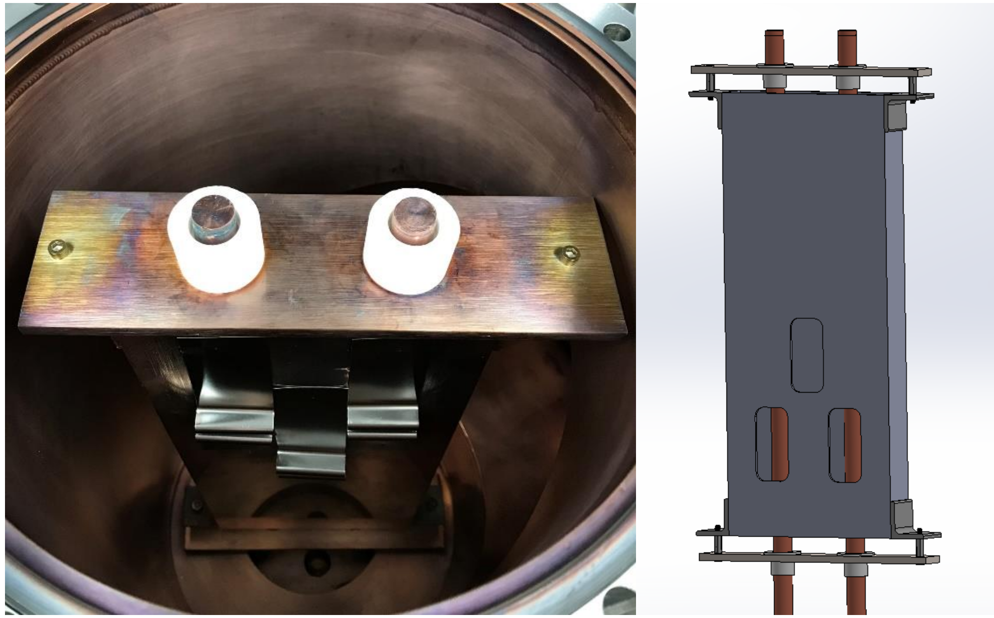
Test Reactor for Coating CEBAF Waveguides

A sample holder with dimensions representative of the CEBAF waveguide structure was constructed for this work

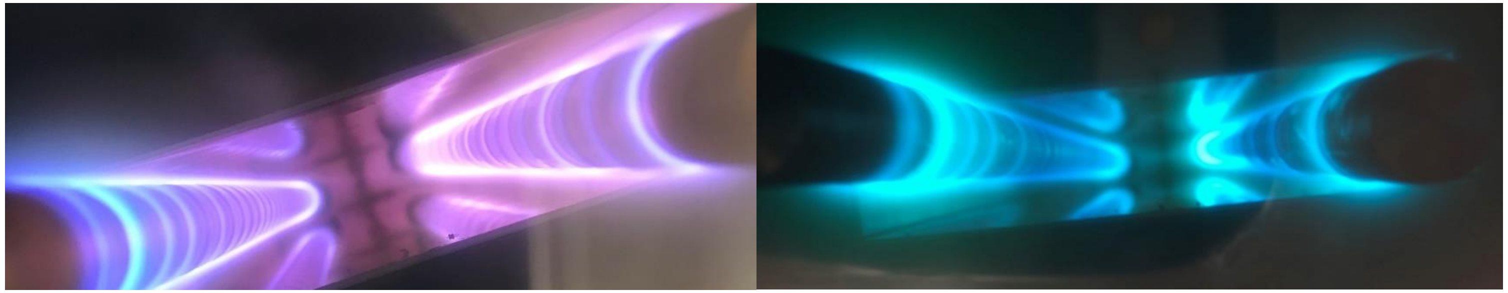
- Rectangular cutouts were included for mounting sample coupons
- Small holes were also included for mounting Si wafer pieces to serve as witness coupons for calculating local deposition rates

Fixturing was also made for holding/positioning the magnetrons themselves

- The small internal clearance requires more precision in magnetron placement than for the LCLS-II bellows



(Left) A photograph of the test-fixture used for these experiments. (Right) A CAD rendering of the same fixture.



Photographs of the dual-magnetron configuration in-operation during the plasma-etch step (left) and main deposition step (right), taken with the top of the test fixture removed.

A photograph of a coated sample coupon and Si wafer piece.

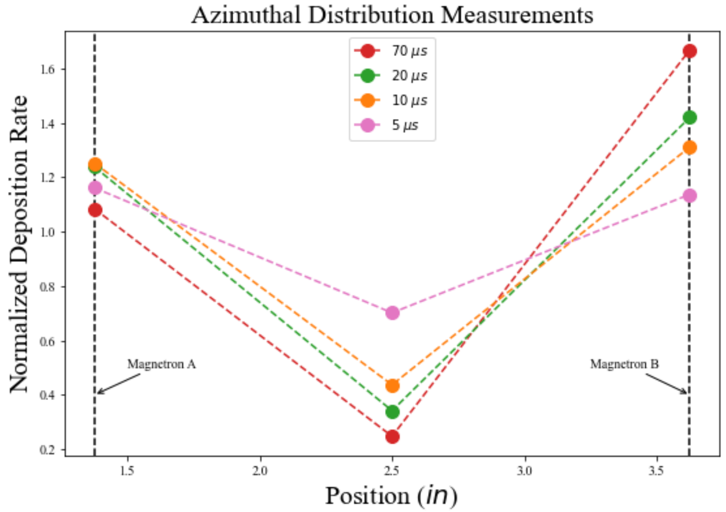
High-Aspect-Ratio Challenges

Film thickness measurements at right were taken over each magnetron and half-way between them

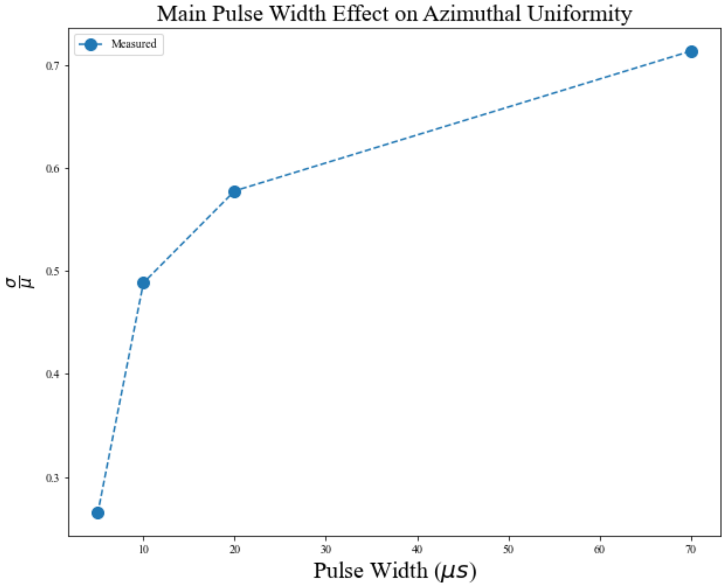
- Used as a quick evaluation of uniformity: lower peak/mid-point ratio is what we’re looking for

In general, shorter main pulses are extremely beneficial for increasing azimuthal uniformity

  - If the main pulse is too long, too much mass is deposited in the form of Cu neutrals
    - Neutrals leave the target with an azimuthally uniform distribution
- Long kick pulses (100 µs or more) are beneficial
- Moderate kick voltages (~100 V) are good
  - Too low: insufficient ionization of the in-flight Cu neutrals occurs
  - Too high: results in a large sheath region that doesn’t conformally cover the smaller features in the substrate surface



Normalized deposition rates at each magnetron and at the midpoint between the two for a variety of main-pulse widths.

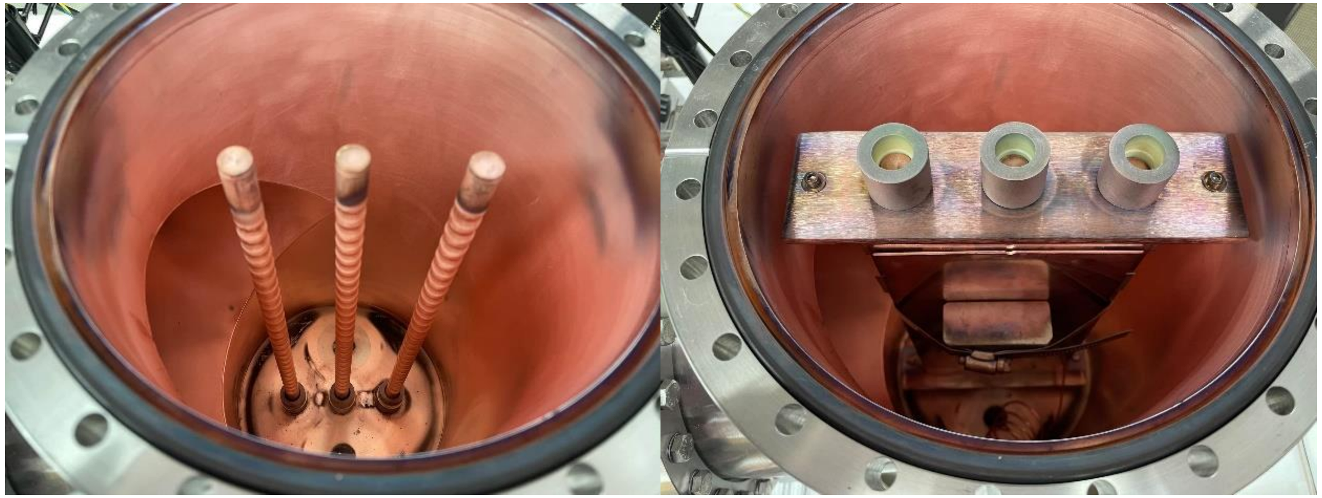


Uniformity as a function of main-pulse width.

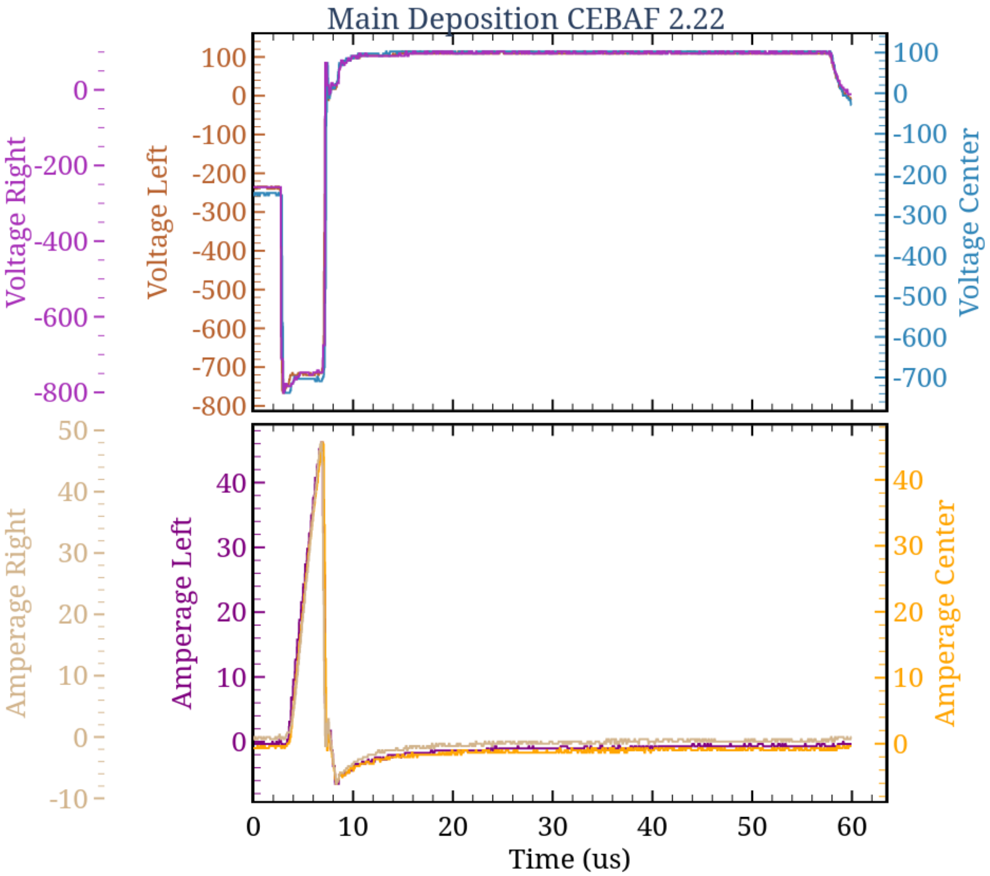
Three-Magnetron Deposition Setup

Cu is deposited onto a stainless steel strip mounted to a cutout in the sample holder

- Again, the sample holder approximates the inner dimensions of a CEBAF waveguide
  - The cutout is a strip along the entire long edge of the holder
- Pulsing of the three magnetrons is staggered to prevent current from commuting between them
  - Voltages are adjusted slightly to maintain equal peak currents



(Left) The three radial magnetrons shown with the sample holder unloaded from the chamber. (Right) The deposition chamber loaded with the sample holder and the three radial magnetrons.



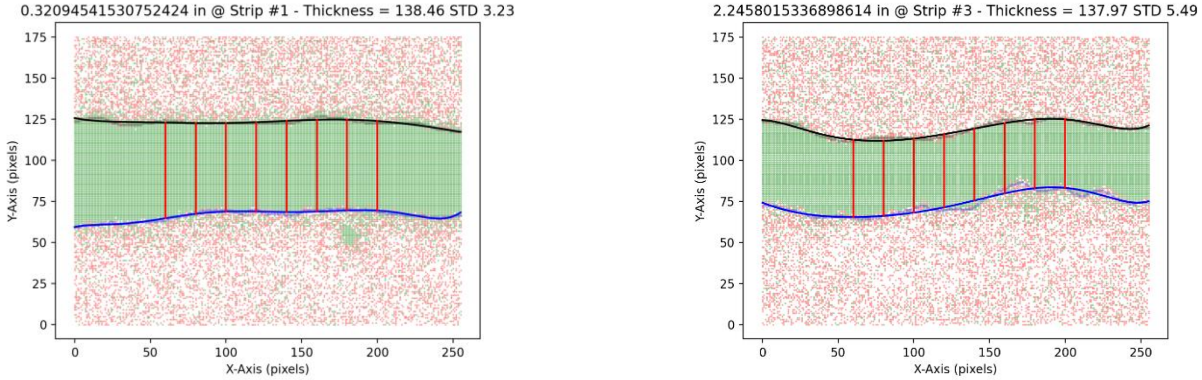
A set of voltage (top) and current (bottom) waveform for each of the three magnetrons. Each of the waveforms here are plotted relative to the t_0 trigger time for the IMPULSE unit that produced it. So, while they are shown here plotted on top of one another, in reality they are evenly spaced out in time.

Sample Analysis: SEM-EDS

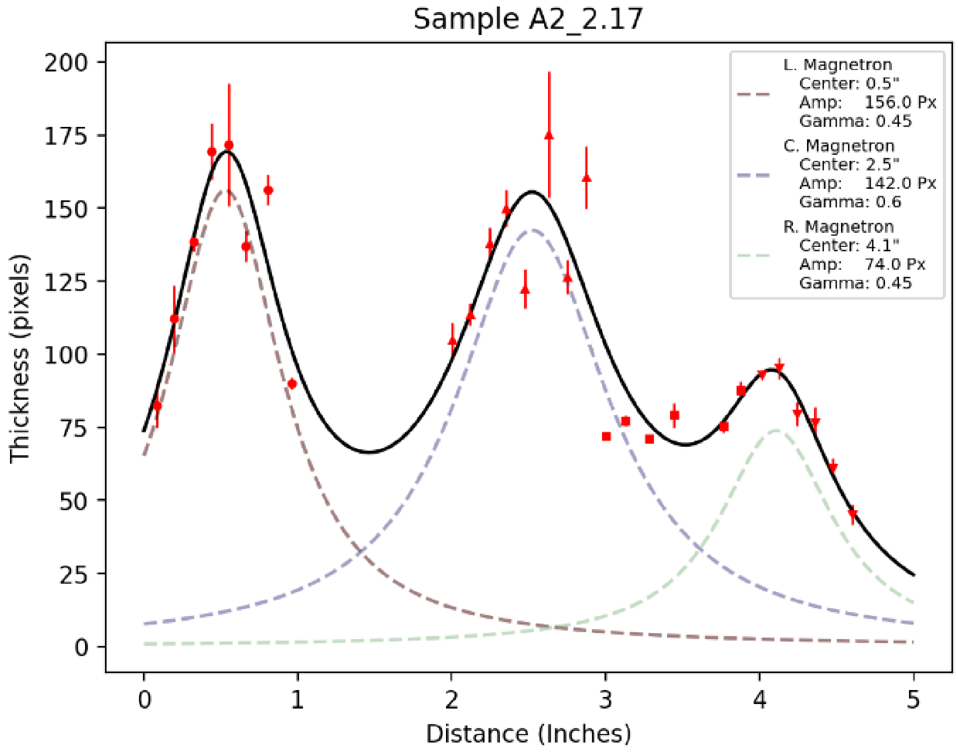
Initially attempted to use SEM-EDS to extract maps of the Cu-distribution at many places along the surface of the coupon

- Film thickness is extracted from the EDS images automatically, using an FFT-based edge-detection algorithm
- The extracted film thicknesses and their locations are used to build up a dataset showing how the film thickness varies along the length of the waveguide edge

Although initial results were promising, sample prep and repeatability were both problematic



Two EDS images showing Cu distributions, along with the detected edges (in black and blue). Red dots are a single EDS count, while green dots are locations with >1 count on the detector. The evenly spaced set of vertical red lines are merely a guide. They show the region over which film thickness is extracted.

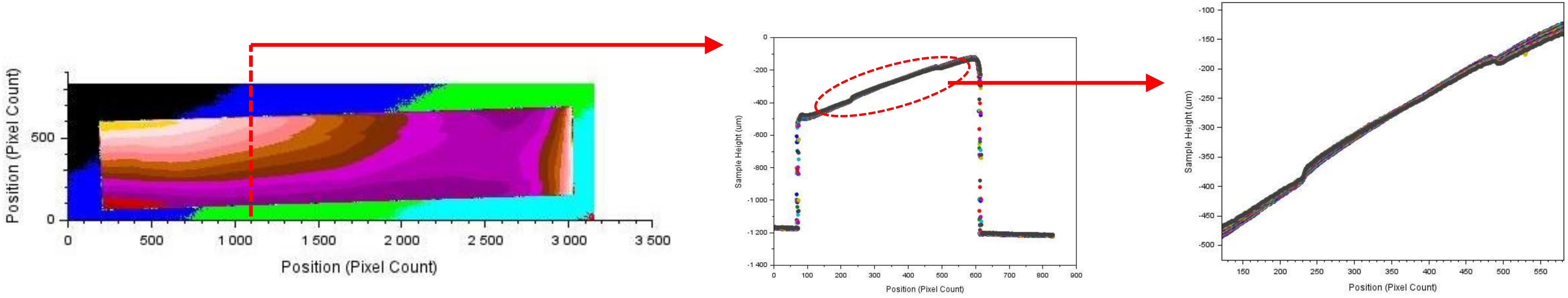


A full set of extracted film thicknesses for this sample along with a best-fit triple-Lorentzian profile (one for each magnetron).

Sample Analysis: Optical Profilometry

We can use an optical profilometer (VR-6200) instead of SEM-EDS

- The resolution is sufficient for these measurements…the Cu film can be clearly seen in the height map
- Data acquisition takes minutes
- With proper sample masking/fixturing during the dep and good fixturing during data acquisition, processing the data takes minutes as well



(Left) A false-color image showing a heightmap the scanned test coupon. The Cu film itself is just barely visible in the distortions to the color bands, especially near the top edge of the coupon. (Center) A set of vertical lineouts taken from the heightmap. The most prominent vertical features here are the edges of the coupon itself. (Right) The portion of the lineout containing the actual Cu film.

Film Thickness Extraction

Lineouts along the short dimension are used to extract film thickness values

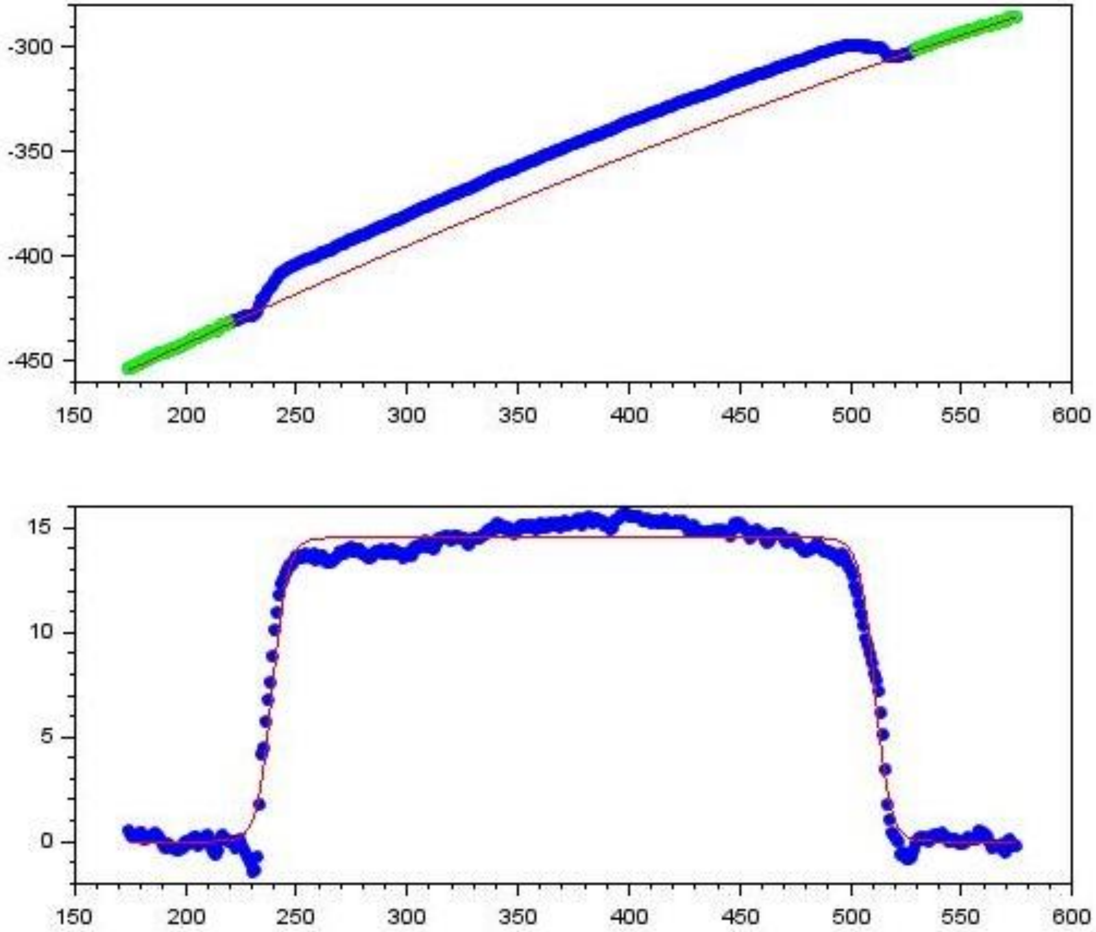
- A 2nd-order polynomial is fit to the stainless profile on either side of the Cu strip is used for background subtraction
- A dual-sigmoid function (essentially a top-hat with smooth sides) is used for fitting to the film thickness:

$$f(x) = p_1\left(\frac{1}{1+e^{-p_2(x-p_3)}} - \frac{1}{1+e^{-p_2(x-p_4)}}\right)$$

p1 is the film thickness

p2 governs the Cu-SS transition

p3 and p4 are the left and right edges of the film



(Top) A lineout taken from the heightmap of the scanned coupon. The areas in green are used for background fitting a 2^{nd} order polynomial for background subtraction. (Bottom) The data after background subtraction. The ~15-µm film is clearly visible.

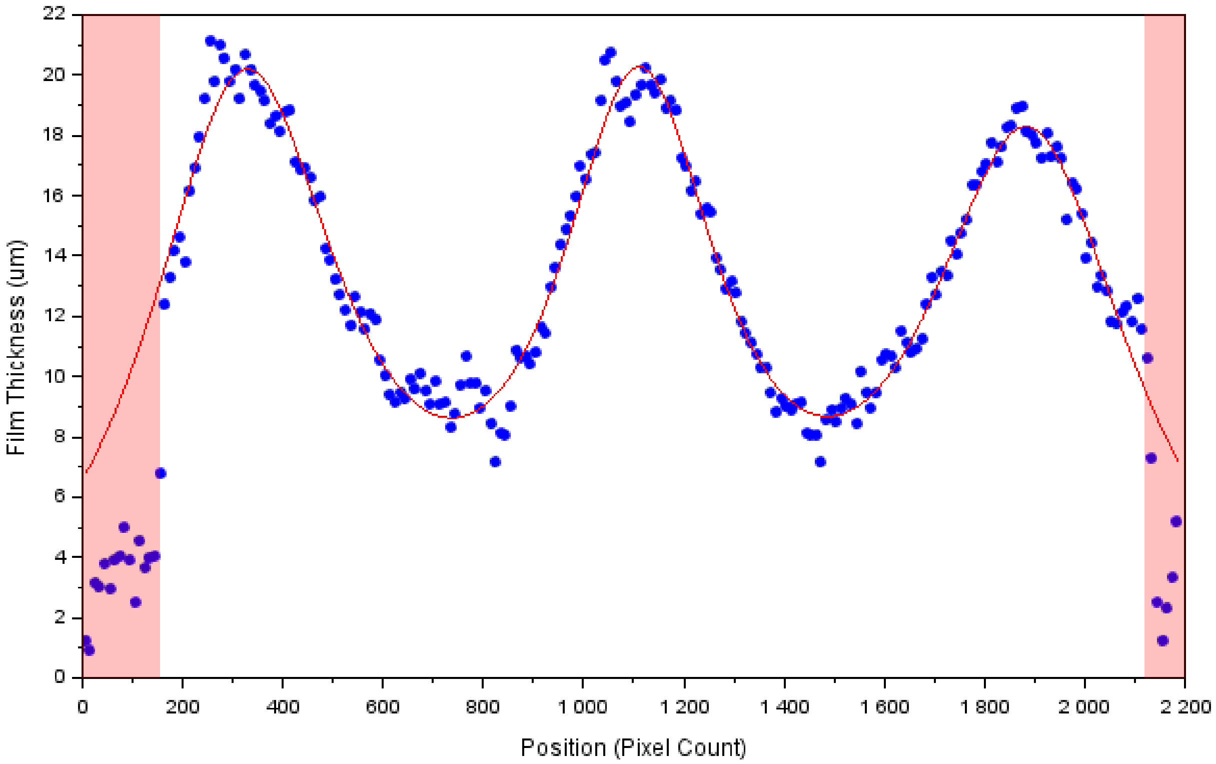
Film Thickness Profile

Film thickness values are then used to construct a profile running down the long edge of the coupon

- A function comprised of three Lorentzian functions and an offset is fit to this profile
  - The Lorentzian was chosen because it is the solution to mass flux through a line (the wall) from a point source (the magnetron) in 2-D

We'll be running a large series of depositions to determine:

- Which factors affect the relative peak heights and
- Which factors affect the conformality (i.e. the relative values of the Lorentzian amplitudes vs the offset value)



The set of extracted film thicknesses as a function of position. The areas shaded in red are outside the bounds of the shadow-mask. A small separation between the mask and the coupon allowed some Cu deposition in this region.

Conclusions, Next Steps, & Acknowledgements

Cu-on-stainless films have been achieved that are conductive, adherent, and conformal

- These films, having thicknesses of ~5—30 µm, do not delaminate, often even after severe plastic deformation of the substrate
- Conformality appears strongly correlated to main-pulse width and kick-pulse voltage

The CEBAF waveguide geometry is inherently challenging due to its high aspect ratio

- Multiple magnetrons and an investigation into further shortening the main pulse are helping to minimize the overall variation in film thickness

This same technique can be used for other applications and with other materials:

- High conductivity coatings for beampipes (e.g. in BNL EIC beamline components)
- Nb for SRF applications, porous gettering materials (Ti, Zr, Mo), etc.

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