A New Method for Grain Texture Manipulation in Post-Deposition Niobium Films

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Experimental Strategy

- Employ energetic condensation (EC) to maximize film deposition density

- Use cold substrate to control surface energies during nucleation; dense amorphous film is created which possesses a much larger energy density than the re-crystallized state.

- Re-crystallization of the film surface results in a crystalline structure resembling bulk Nb. Grain growth is driven by a pulsed laser, or other forms of heating.

- Controlled heating for surface re-crystallization is provided by a pulsed UV HIPPO laser, raster-scanned over the film surface, avoiding excessive substrate heating.

- Also, since the surface processing is capable of being performed in the vacuum chamber, any chance for native oxide layer buildup, or interstitial contamination can be eliminated.

Process Stages

Produce a thick (~ 1um) Nb film on “cold” substrates, beginning with silicon, and then copper (2” coupon). 77 K < “Cold” < 300 K

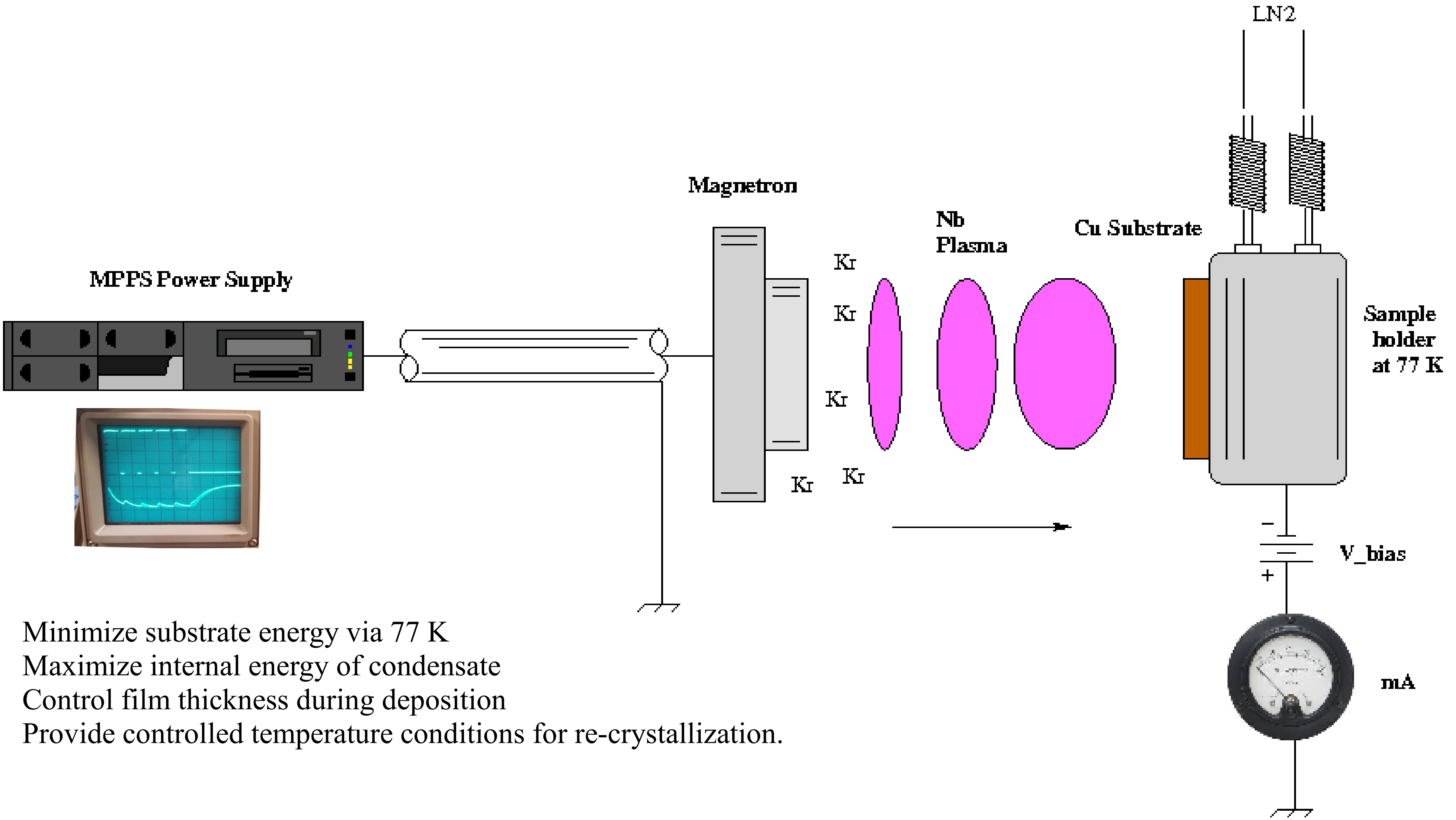
Energetic condensation (deposition), via MPPMS

- Cool substrate (LN2)
- Cold substrate discourages (abnormal) grain growth during the deposition of thin fim
- Establish graded Nb/Cu interface via high energy (> kV) ions for initial deposition (stitching)
- Deposit thick (1 um) nearly-amorphous film via EC; minimize voids, maximize energy density

HIPPO-Laser induced re-crystallization

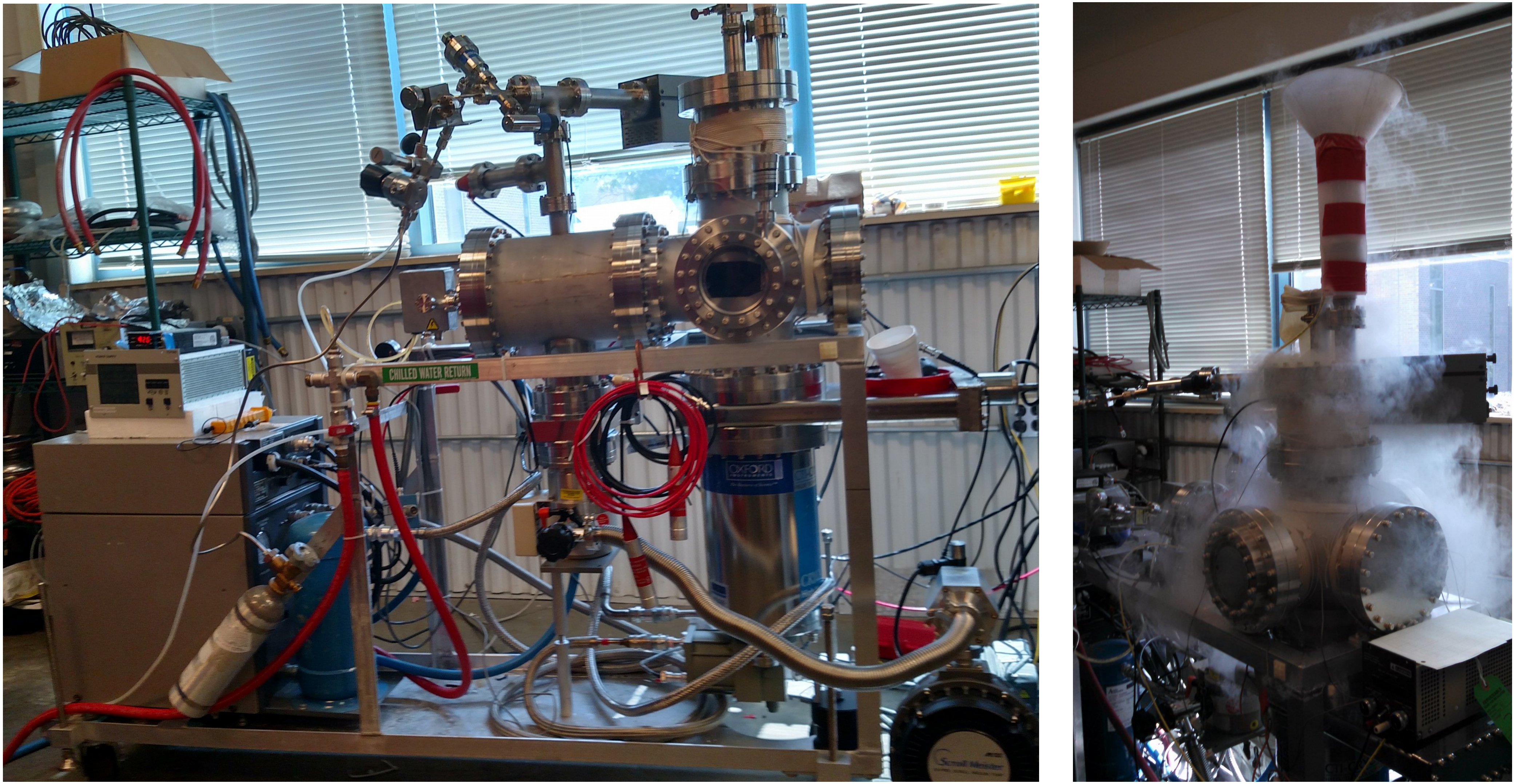
- Precise fluence delivery accurately drives grain growth
- Short pulses produce local heat at RF surface for re-crystallization.
- Exploit short pulse timing and Nb thermal diffusivity to avoid overheating substrate.

Deposition Schematic



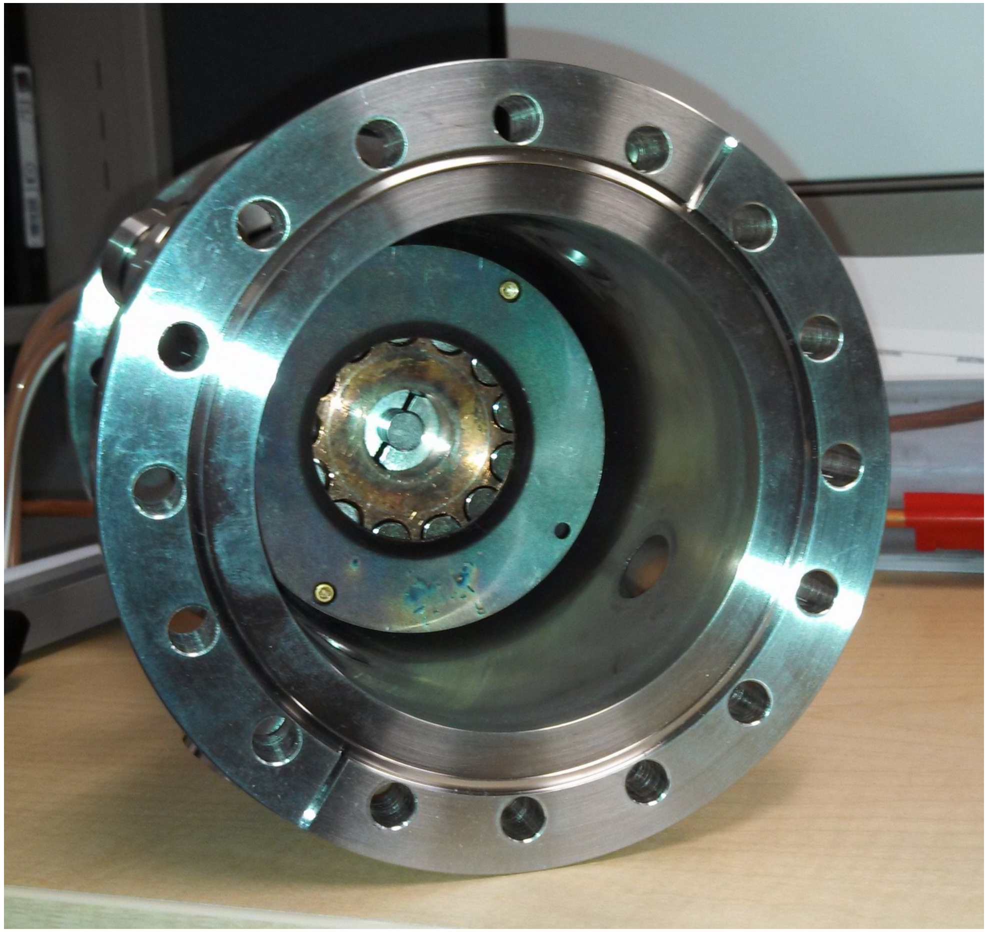
- Minimize substrate energy via 77 K
- Maximize internal energy of condensate
- Control film thickness during deposition
- Provide controlled temperature conditions for re-crystallization.

Deposition Chamber



Deposition Chamber (cont.)

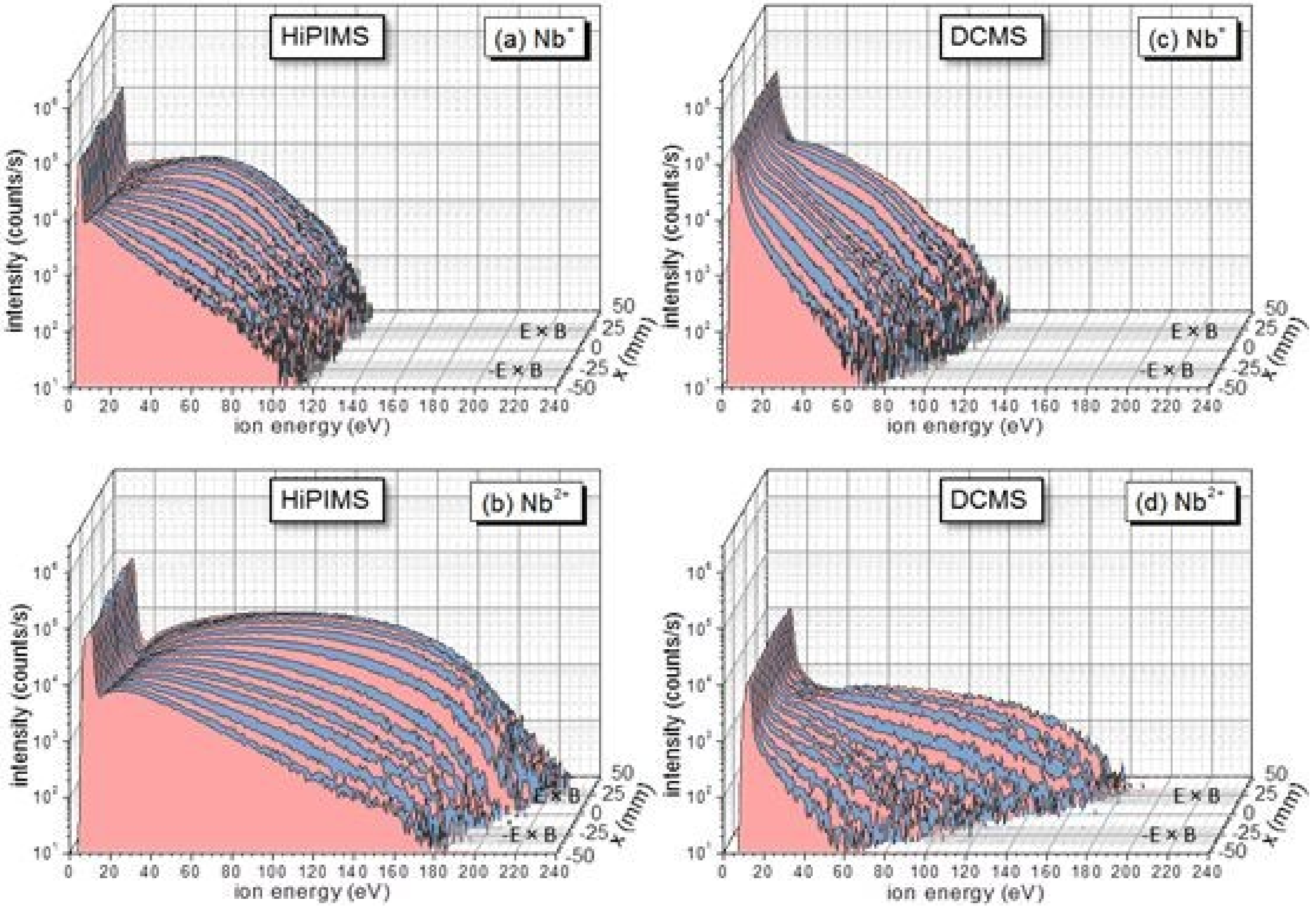
Magnetron Housing (2” Aja)

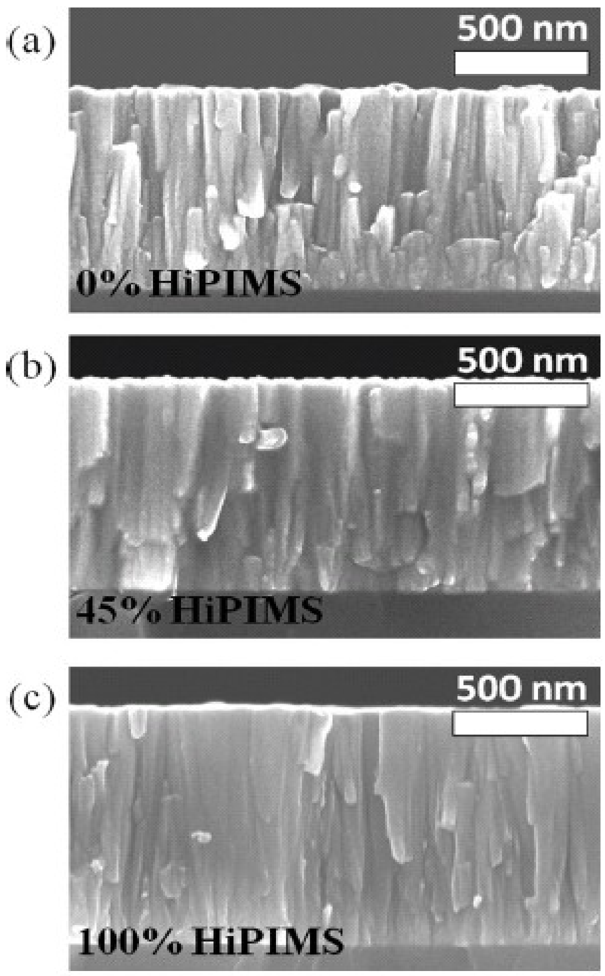


Sample Holder (insulated)

HiPIMS

(High-Power Impulse Magnetron Sputtering)





Films created with 0% (a), 45% (b), and 100% (c) HiPIMS. Density is clearly evident for 100%.

As an alternative to DC magnetron sputtering, the unit is pulsed with large energy (>10x), but with low duty factor (~1%). The ion ratio is quite extreme, and sufficient for direct ion bombardment. T ~ 200us. F ~ 10 kHz. Power density is ~kW/cm^2.

Anders, A., “Discharge Physics of High Power Impulse Magnetron Sputtering,” *Proc. Of 12th Inte Conf. On Plasma Surf. Eng.*, **Vol. 205**, No. 2, 2011.

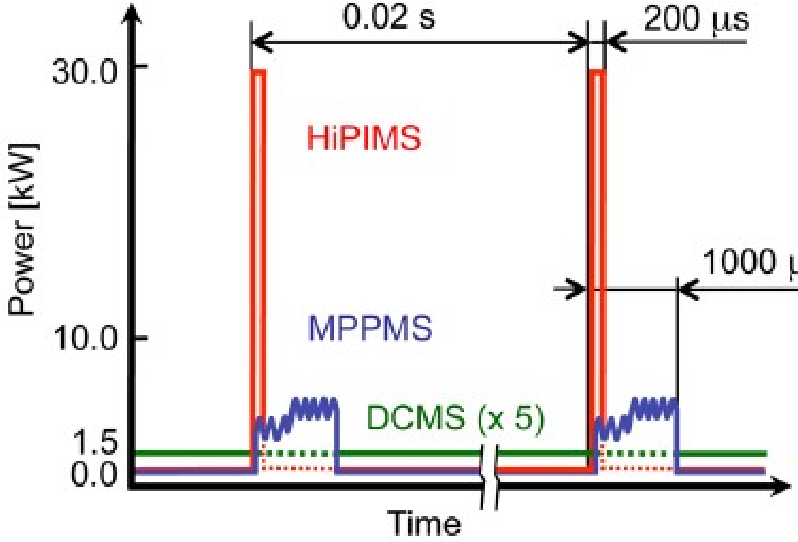
Modulated Pulse Power Magnetron Sputtering

MPPMS is an attractive alternative to HiPIMS

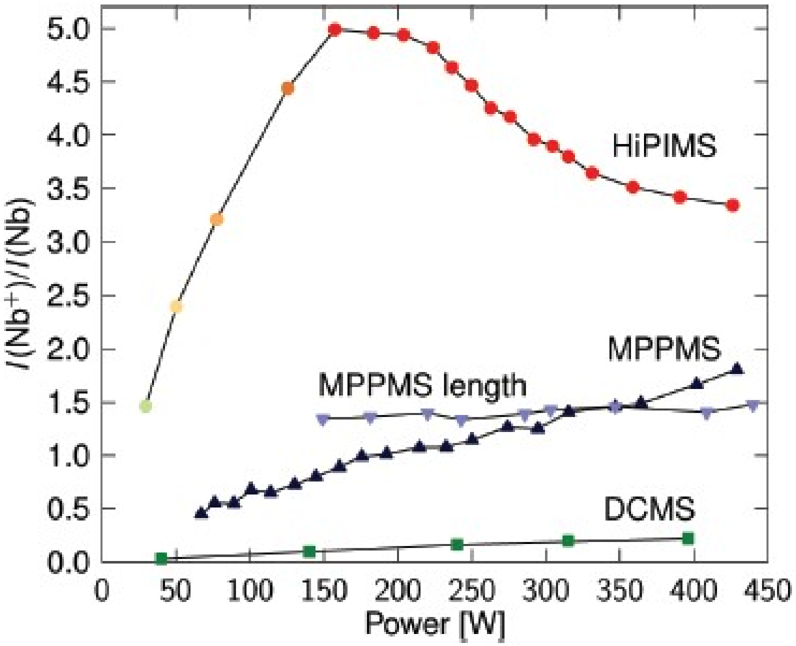
Micropulse = 100 uS HiPIMS pulse

Macropulse = up to dozens of micropulses

Ignition + extinguish + re-ignition stimulates ion-rich plasma of **well-defined energies.**

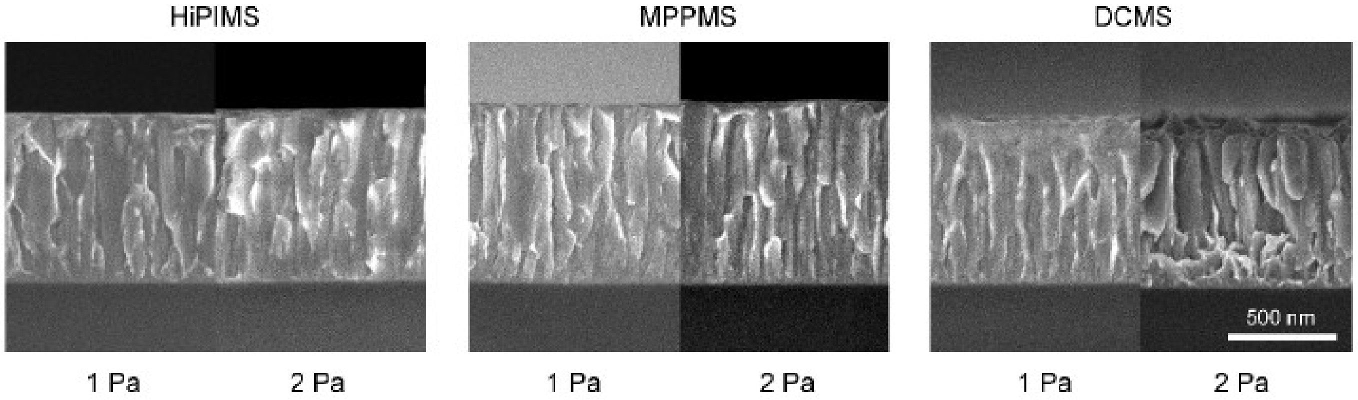


Relative Nb ion production for HiPIMS, MPPMS, and DCMS, obtained by optical spectra (Hala, et al.)

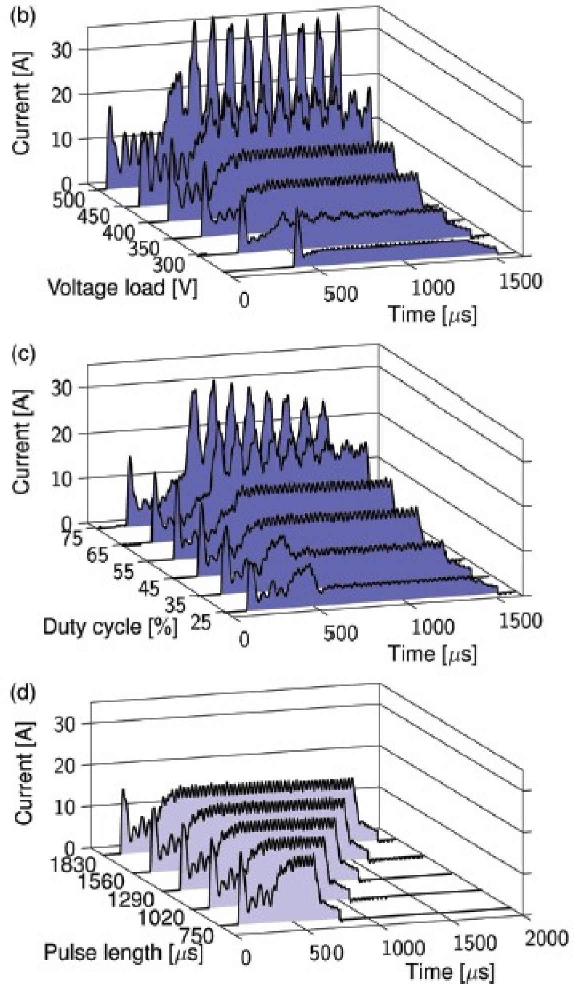


Hala, M., J. Capek, O. Zabeida, J. Klemberg-Sapieha, L. Martinu, "Pulse Management in High Power Pulsed Magnetron Sputtering of Niobium," *Surf. Coat. Tech.*, **Vol. 206**, No. 19-20, pp. 4186-4193, May, 2012.

MPPMS for Nb (cont.)



	HiPIMS	MPPMS	MPPMS length	DCMS
Type of power regulation	Cathode voltage	Voltage load	Pulse length	Cathode voltage
Cathode voltage [V]	550 – 1600	470 – 670	650 (1 Pa) / 500 (2 Pa)	290 – 390
Pulse duration [μs]	200	1500	880 – 1830	—
Average power [W]	45 – 345	100 – 345	105 – 320	38 – 310
Peak power density [W/cm^2]	700 – 3200	140 – 540	400	3 – 25



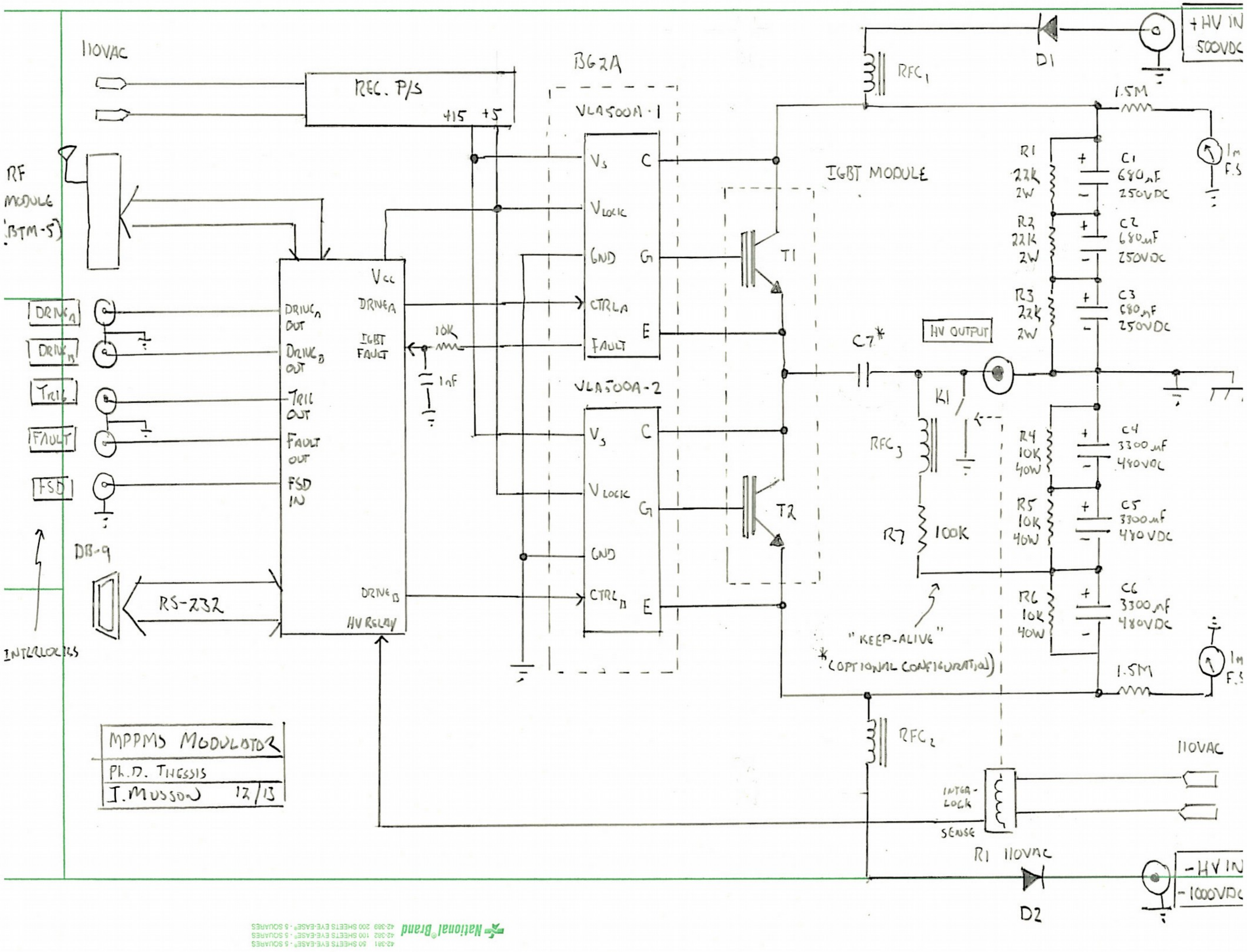
Parametric comparison of EC methods, with resulting Nb film SEMs.

MPPMS films strongly resemble HiPIMS, while also exploiting a higher growth rate (via duty factor).

Prototypical pulse structure obtained by parametric study……

Hala, M., J. Capek, O. Zabeida, J. Klemberg-Sapieha, L. Martinu, "Pulse Management in High Power Pulsed Magnetron Sputtering of Niobium," *Surf. Coat. Tech.*, **Vol. 206**, No. 19-20, pp. 4186-4193, May, 2012.

500 J MPPMS Modulator (IGBT, FPGA)

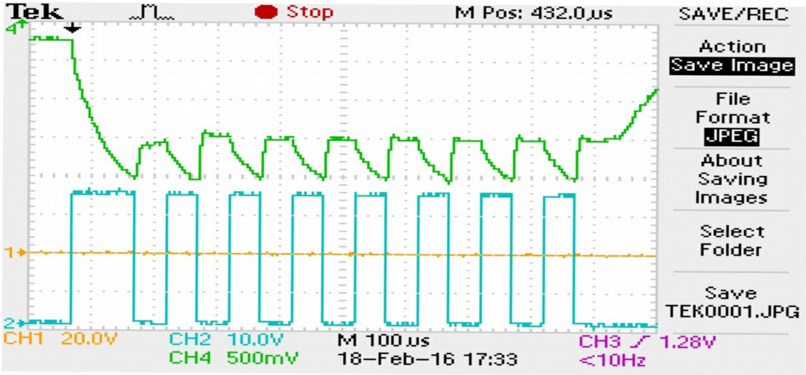


1200 V @ 1500 A / pulse

77 K Deposition Formula.....Si Substrate

Kr pressure = ~10 Torr (1 Pa)....Niobium's nearest inert neighbor

Each macropulse = 8 micropulses:



First pulse = 100 us

50 us for subsequent pulses

MPPMS parameters held close to Hala, et al., template.

$V = 650$ V

$P_{ave} = 300$ W

$F_{rep} = 94$ Hz

$P_{pk} = 7$ kW / macro

$W_{pulse} = 3$ J / macro

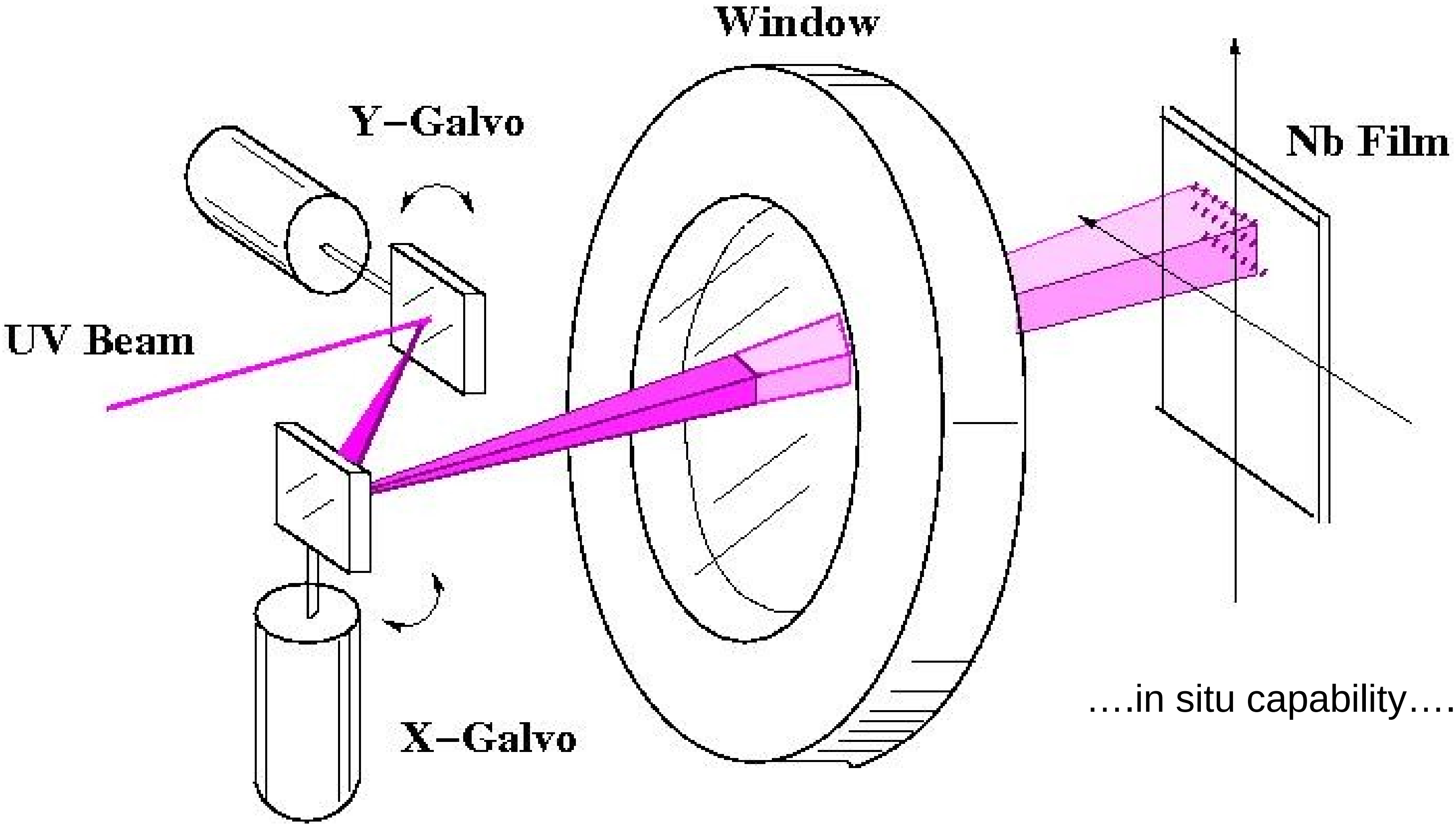
$I_{ion} = 35$ mA

Starting pressure = 1.7 10^{-7} Torr

Deposition time = 5 hrs

Measured thickness = 765 nm (SEM)

Raster-Scanned 355 nm HIPPO Laser

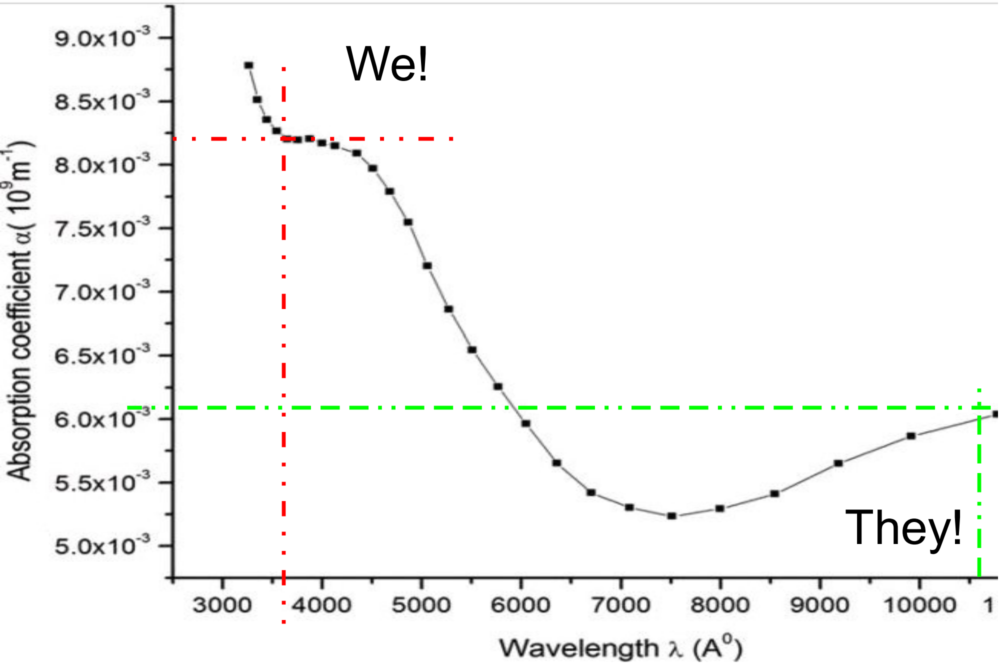


….in situ capability….

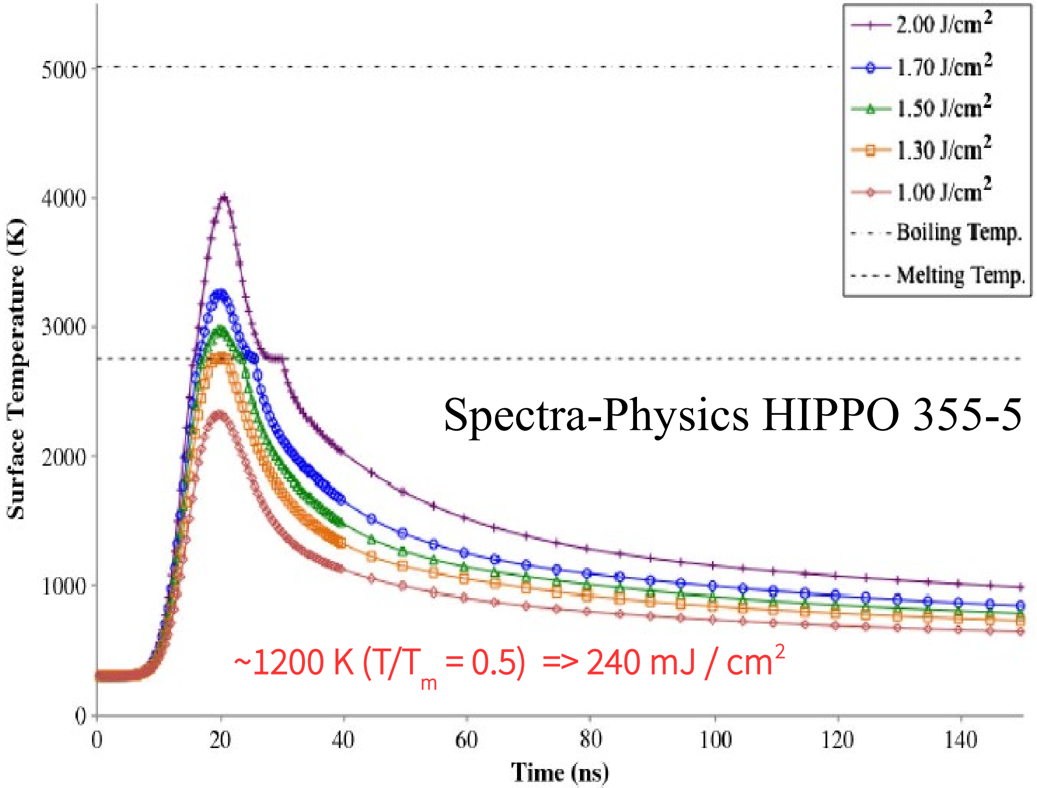
Thermal Profile Estimation

Singaravelu, et al., have performed bulk niobium annealing ….. we have a starting point!

Absorption vs Wavelength, niobium



Simulation of 8 ns, 1064 nm Fluence Pulses



Spectra-Physics HIPPO 355-5

~1200 K ($T/T_m = 0.5$) => 240 mJ / cm^2

Singaravelu, S., J. Klopf, C. Xu, G. Krafft, M. Kelley, "Smoothing of Niobium Superconducting Radio Frequency Cavity Surfaces by Laser Melt Process,"

Thermal Profile Estimation (cont.)

Spot diameter ~ 100 um

1-D model OK for now..... 100 um >> $\sqrt{D_{Nb}\tau} \approx 700\,nm$ $\longrightarrow$ $\frac{\partial T}{\partial t} = D\frac{\partial^2 T}{\partial x^2} + \frac{Q(x,t)}{c\rho}$

Laser pulse ~12 ns

Repetition rate ~ 50 kHz

T_{dwell} = 20 us

Bechtel Square Laser Pulse Model*

$$\Delta T(0,t) = 0,\ t<0$$

$$\Delta T(0,t) = \frac{2I_m(1-R)}{K}\frac{(Dt)^{1/2}}{\pi^{1/2}},\ 0 \le t \leqslant \tau$$

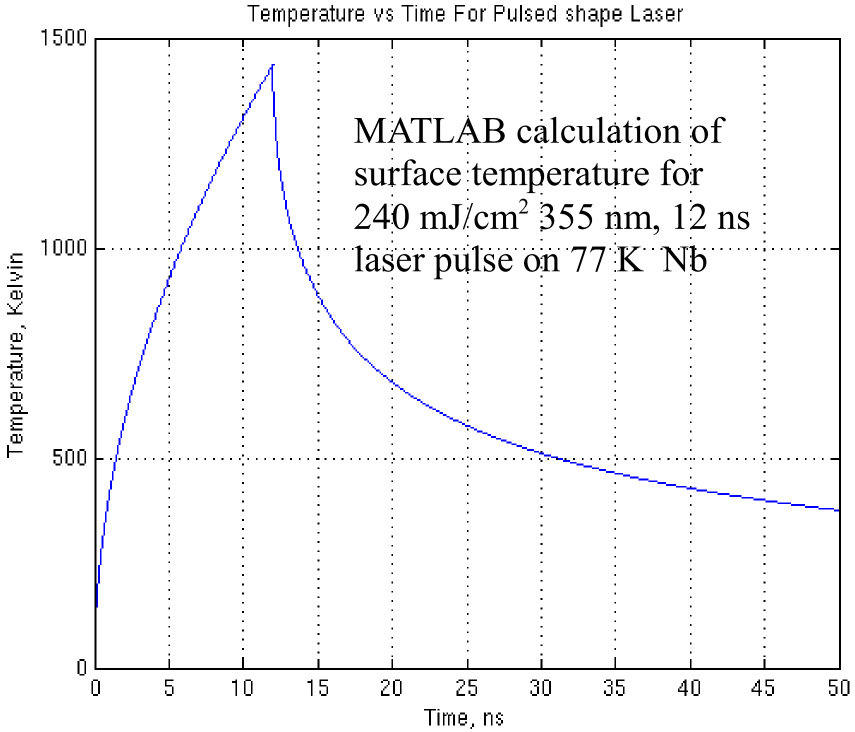
$$\Delta T(0,t) = \frac{2I_m(1-R)}{K}\frac{(Dt)^{1/2}}{\pi^{1/2}}\left[\left(\frac{t}{\tau}\right)^{1/2} - \left(\frac{t}{\tau}-1\right)^{1/2}\right],\ t>\tau$$

K = initial temperature

D = thermal diffusivity

R = reflectivity

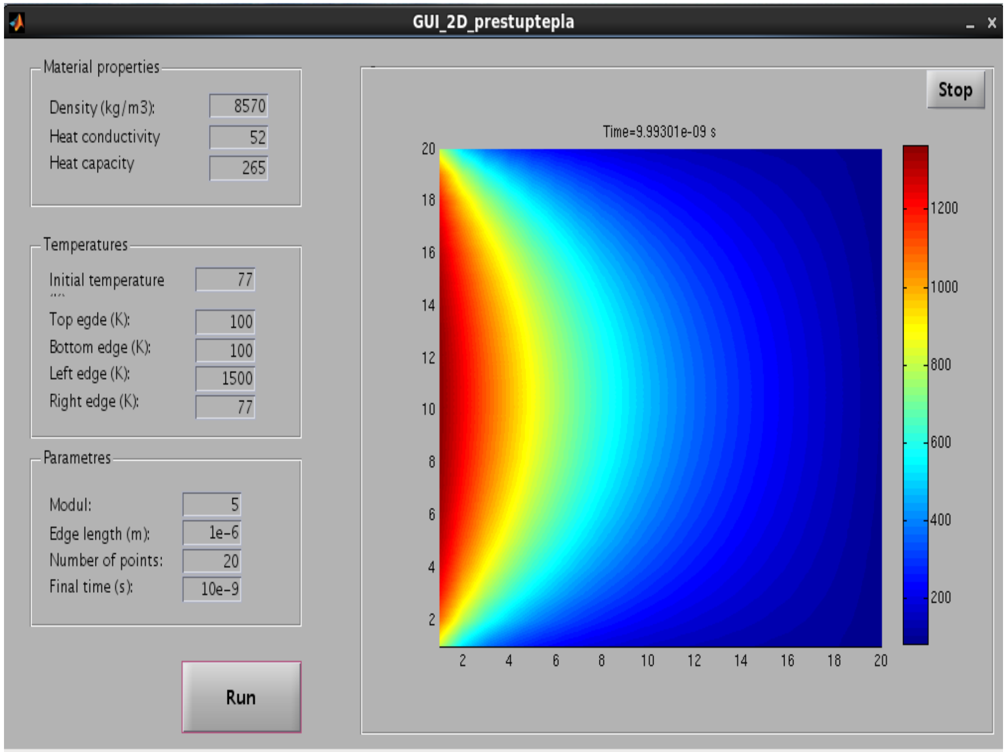
I = irradiance



MATLAB calculation of surface temperature for 240 mJ/cm^2 355 nm, 12 ns laser pulse on 77 K Nb

*Bechtel, J. H., “Heating of Solid Targets with Laser Pulses,” *Journ. Appl. Sci.*, **Vol. 46**, No. 4, April, 1975.

Scan Rate Calculation



Material properties

Density (kg/m3): 8570

Heat conductivity 52

Heat capacity 265

Temperatures

Initial temperature 77

Top egde (K): 100

Bottom edge (K): 100

Left edge (K): 1500

Right edge (K): 77

Parametres

Modul: 5

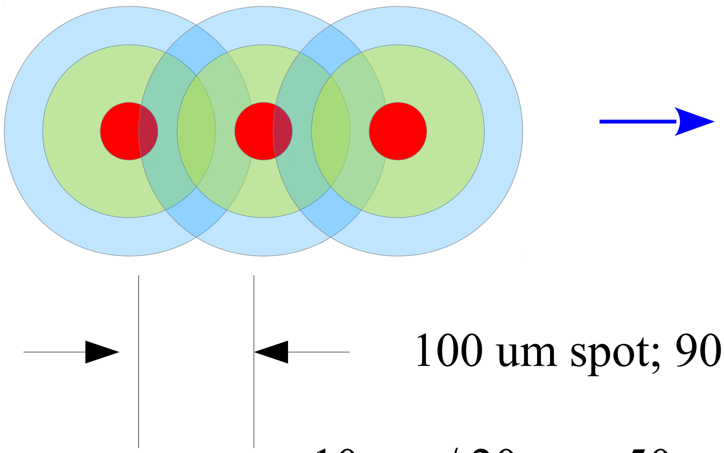
Edge length (m): 1e-6

Number of points: 20

Final time (s): 10e-9

MATLAB Model courtesy of Dominik Gibala.*

Raster-scanning for precise fluence control



100 um spot; 90% overlap

10 um / 20 us = 50 cm /s

~ 20 ms / cm (25 Hz / cm)

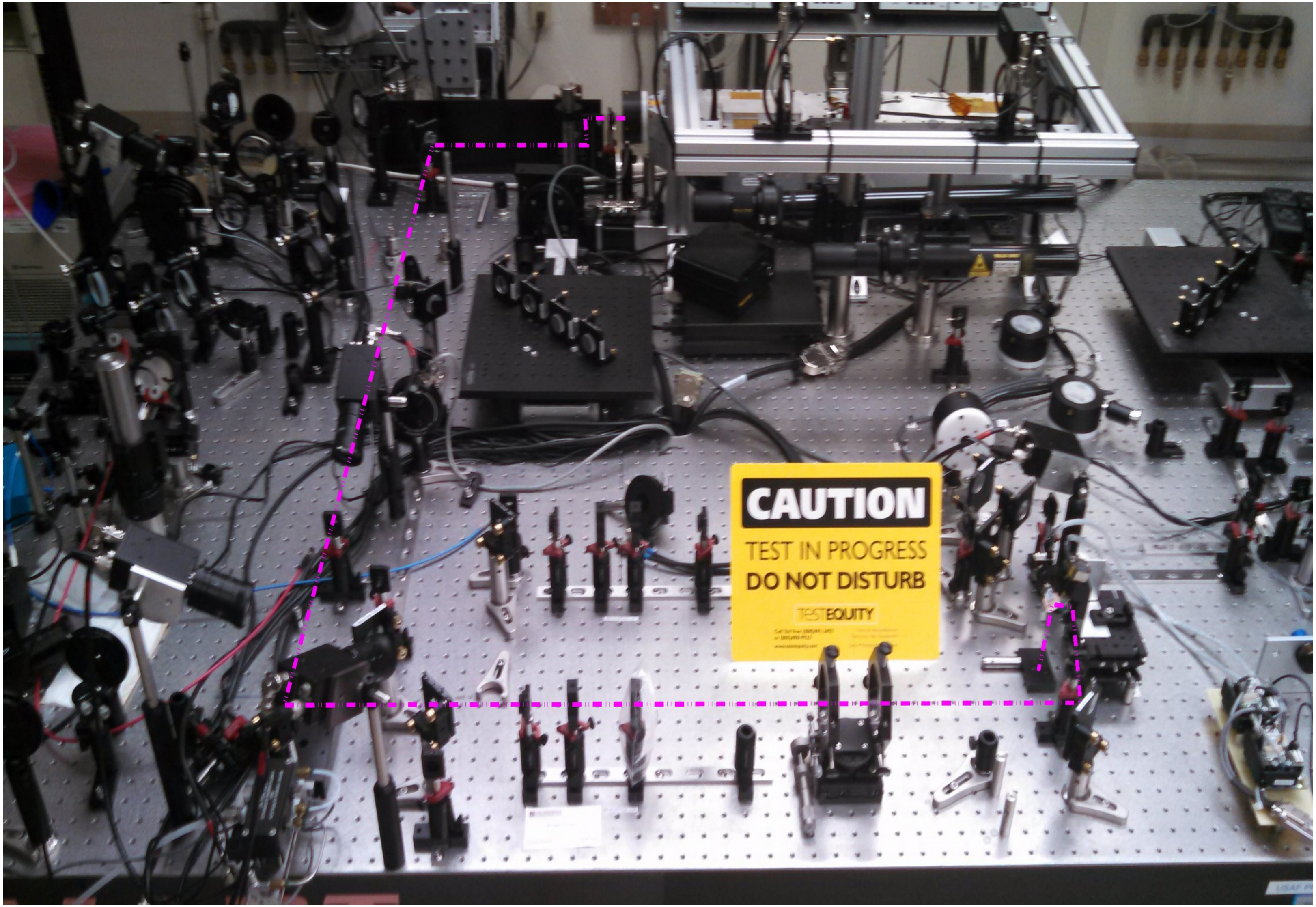
~ 125 Hz on 2” Cu coupon

12 ns pulses raise local surface area to ~ 1200C

Substrate is at 77 K, and protected from surface temperature

*http://www.mathworks.com/matlabcentral/fileexchange/35068-gui-2d-heat-transfer

Spectra Physics HIPPO Laser (5W @ 355 nm)



Anticipated Analyses

XRD

- Grain Size (bulk)
- Compression strain

SEM

- Film thickness
- Surface texture, roughness

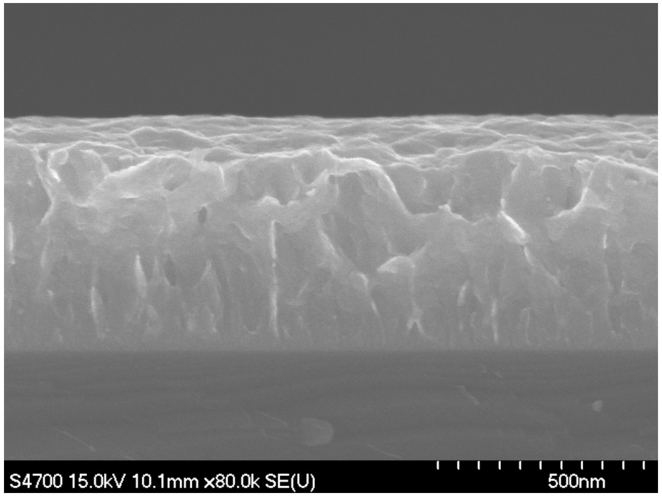
TEM

- Evidence of ion stitching
- Re-crystallization, depth, crystal structure

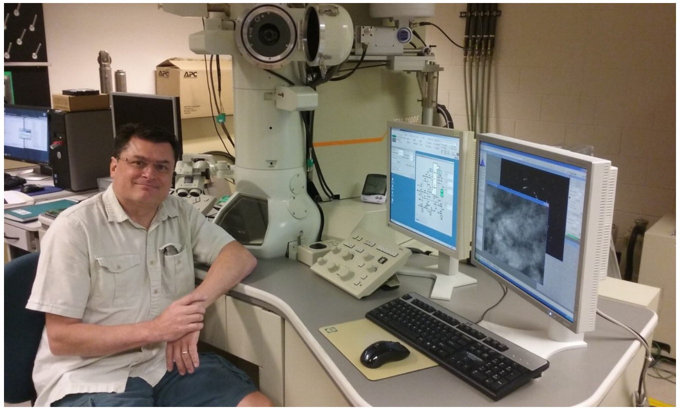
EBSD

- Grain structure (surface), orientation
- Strain

Tc, Surface Impedance Characterization Cavity*

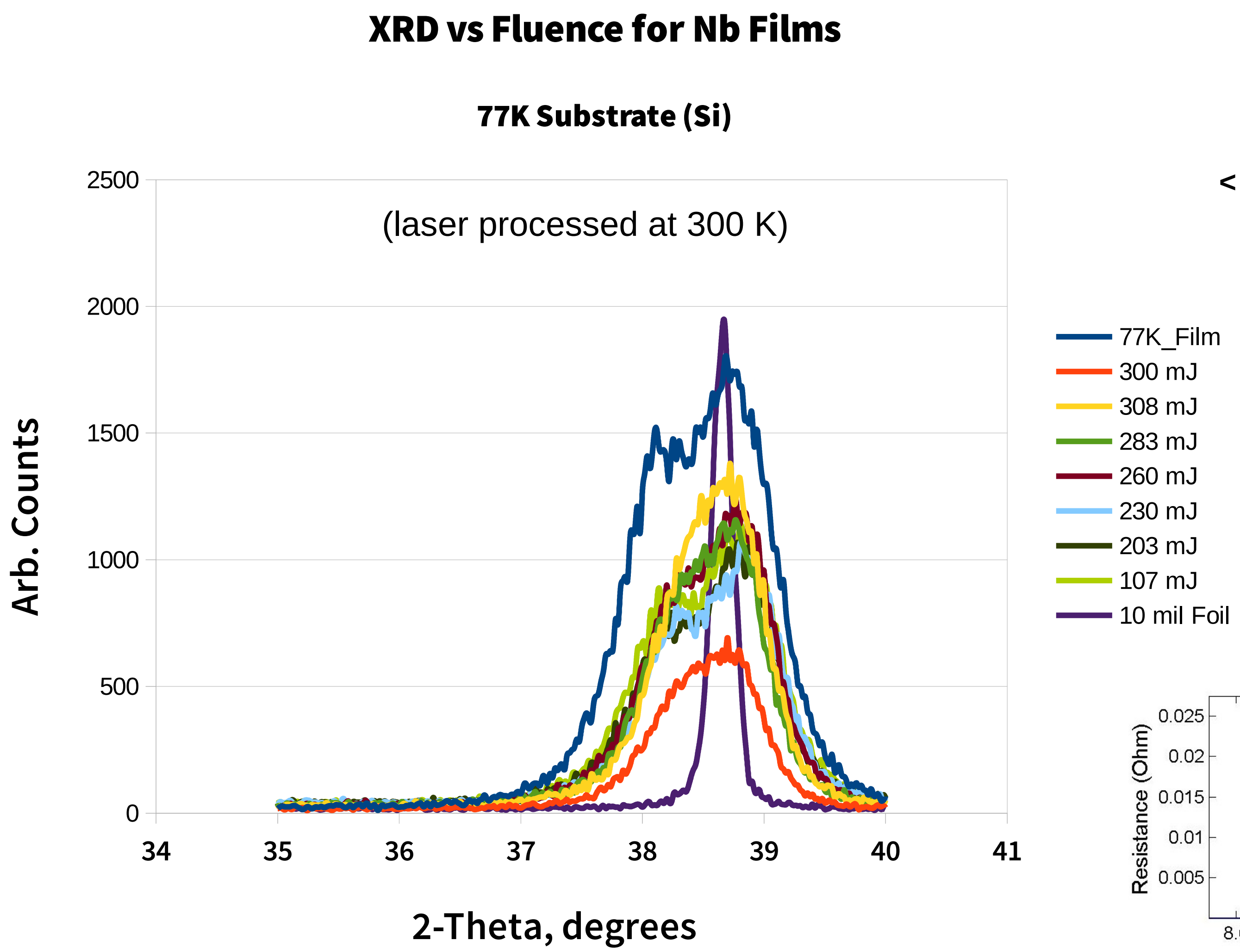


Nb film from EC, 77K Si substrate



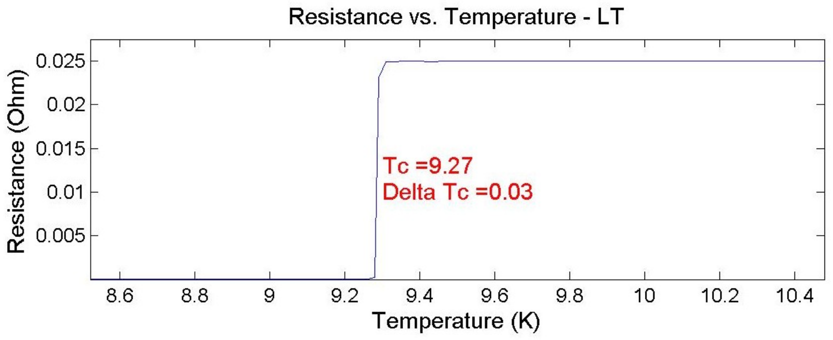
*Xiao, B., C. Reese, H. Phillips, R. Geng, H. Wang, F. Marhauser, M. Kelley, "Radio Frequency Surface Impedance Characterization System for Superconducting Samples at 7.5 GHz," *Rev. Sci. Inst.*, **Vol. 82**, 056104, 2011.

XRD, T_c Analysis of First Films (Scherrer)



< 500 mJ!

Fluence		Tc	Grain Size	Strain
77K, Raw		9.37	4.4	0.025
300 mJ		9.29	8.8	0.0125
308 mJ		9.27	9.16	0.012
283 mJ		9.27	8.8	0.0124
260 mJ		9.29	8.15	0.0134
230 mJ		9.28	7.72	0.0141
203 mJ		9.29	7.59	0.0144
107 mJ		9.32	6.98	0.0157
10 mil Foil		9.27	43.98	0.0025



Conclusion

Grain texture manipulation facilitates detailed studies

MPPMS is suitable for producing high density films

“Simplistic” thin film deposition greatly improves manufacturability (minimize parameters)

Potential Grain texture control from localized heat source

In situ processing eliminates oxide layer

Ion stitching maximizes film adhesion to substrate (thermal)

Many thanks to JLAB SRF (J. Spradlin), ODU ARC, JLAB I&C Group, M Burton