Measurements and Modeling of Liquid Metal Sputtering in IIAX at UIUC

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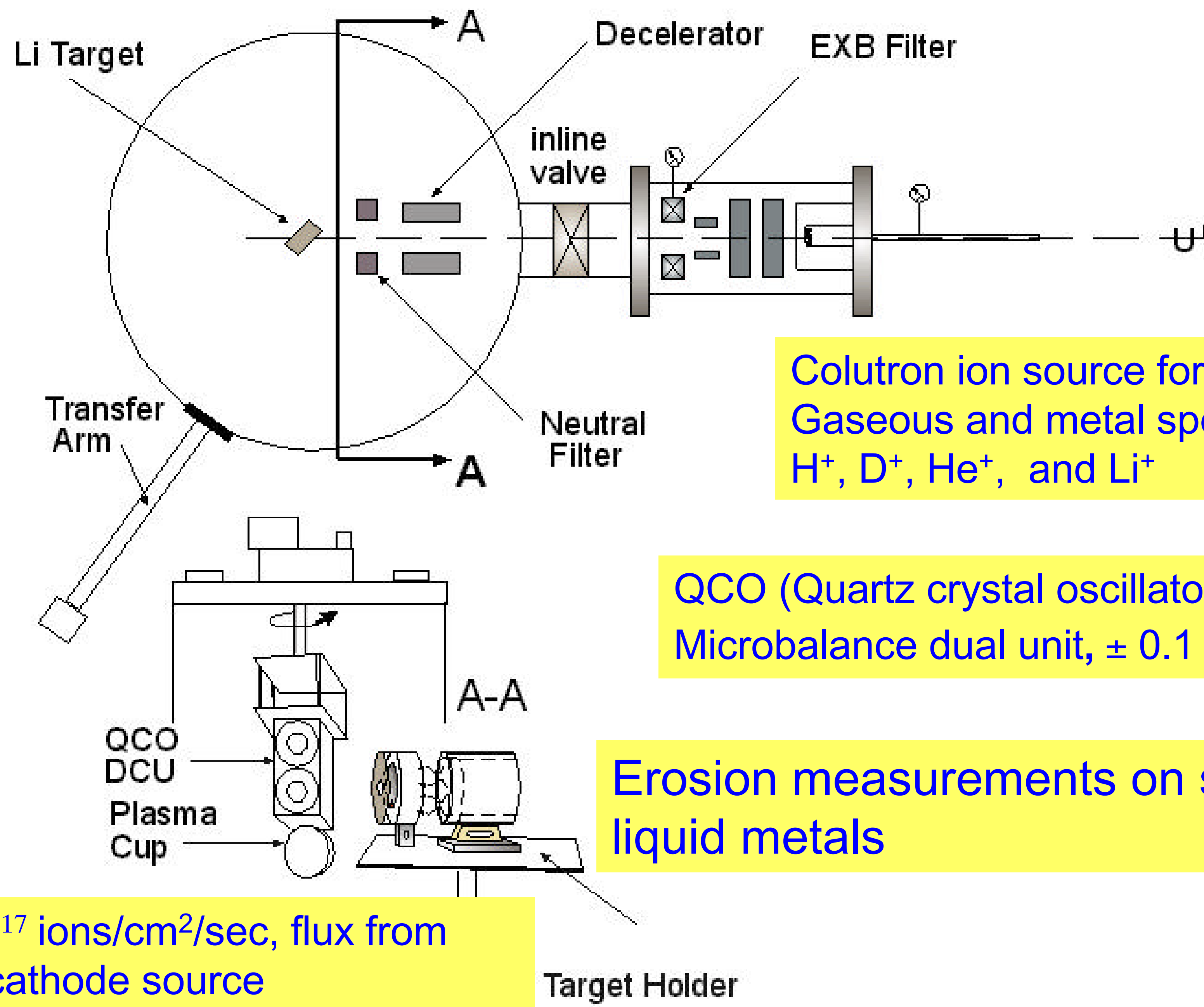
Outline of Talk

- Experimental setup of IIAX (Ion-surface Interaction Experiment)
- Lithium erosion studies in IIAX.
- Experimental results for liquid lithium and liquid tin lithium
- Temperature dependence of the Li sputtering yield from lithium and tin-lithium surfaces
- Key issues and mechanisms
- Conclusions and Future Work



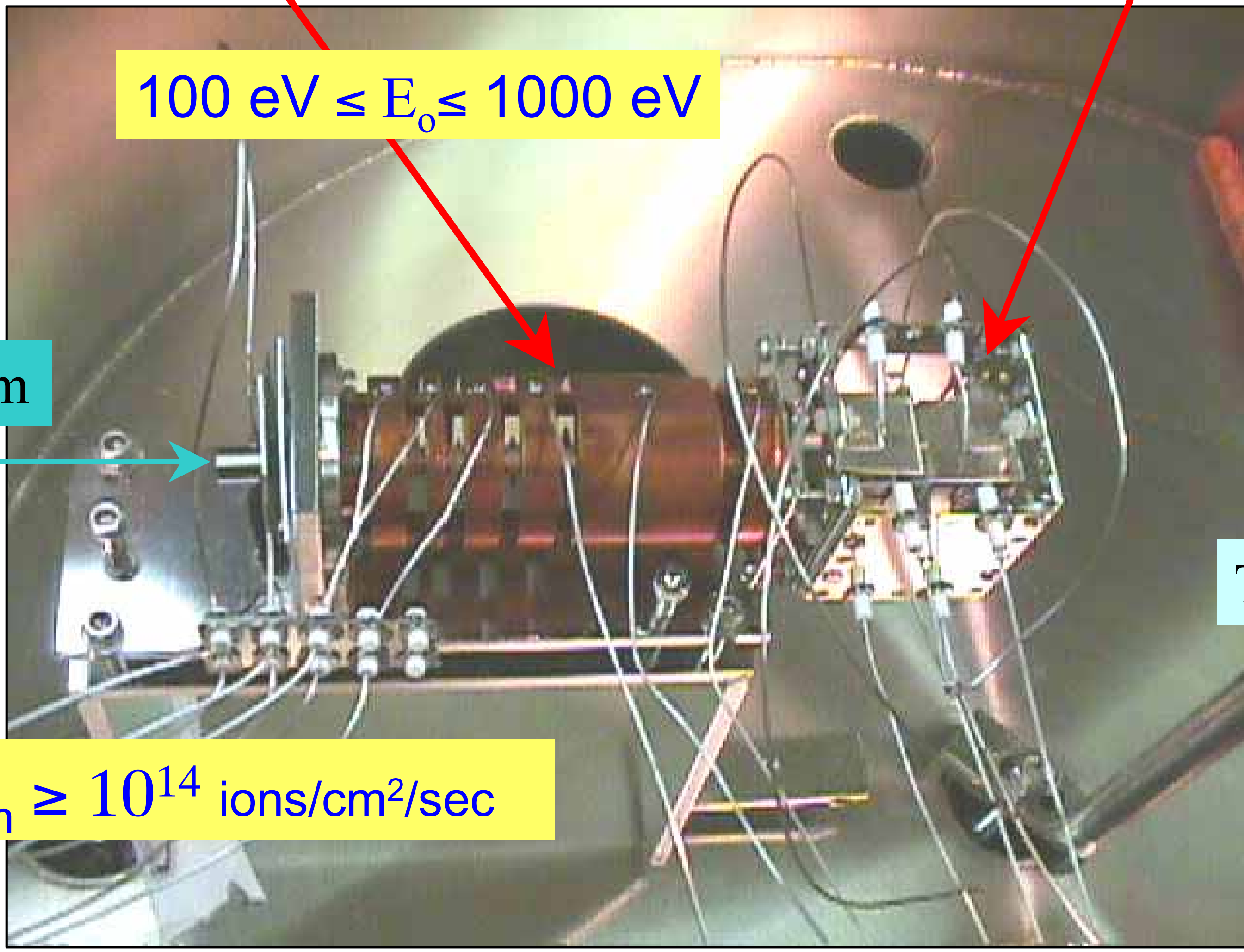
Colutron ion source for both Gaseous and metal species: H^+, D^+, He^+, and Li^+

QCO (Quartz crystal oscillator Microbalance dual unit, ± 0.1 Å)

Erosion measurements on static liquid metals

$\Gamma^+_D = 10^{17}$ ions/cm^2/sec, flux from hollow cathode source

Decelerator and Neutral Filter

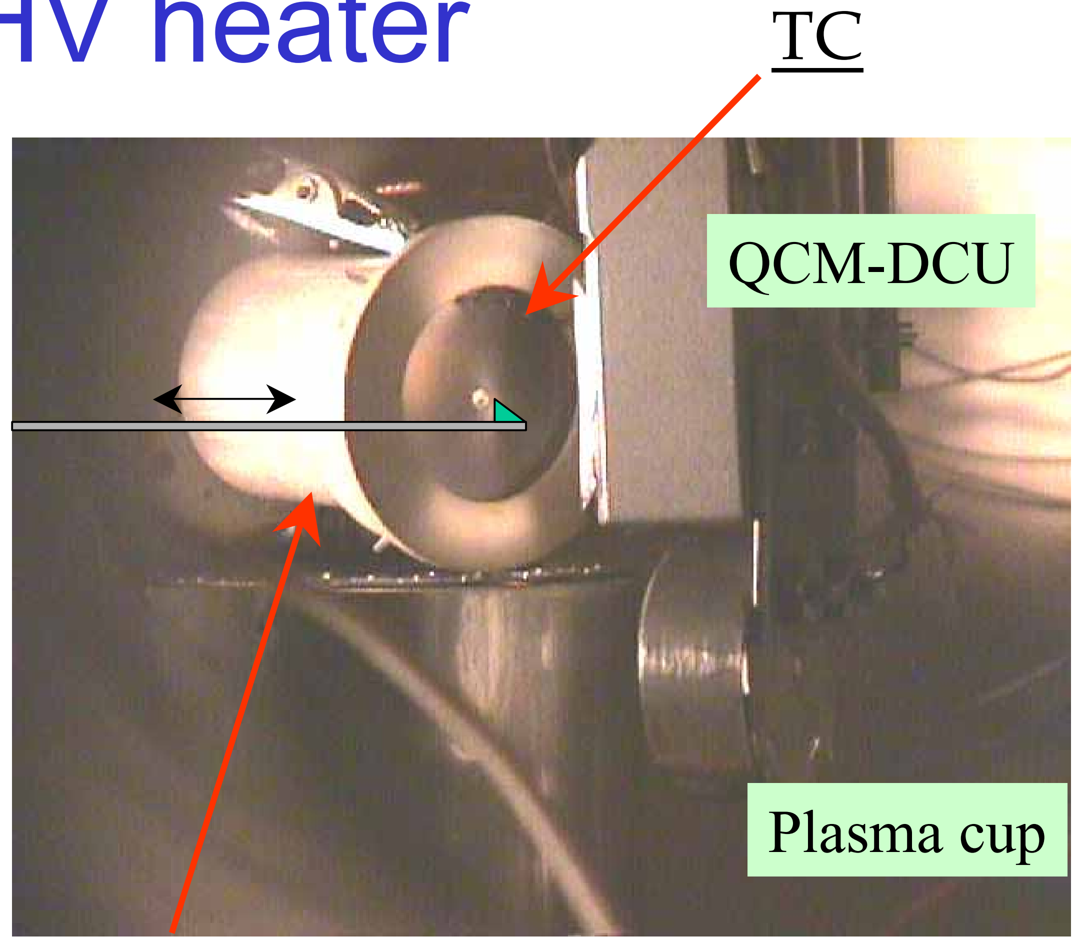


Evaporation Shield for Liquid Metal Sputtering Measurements

- Evaporation rate for Li at 200 °C:
  - calculated: 4 x 10^{11} /cm^2/sec
  - measured: 5.1 x 10^{11} /cm^2 /sec
- Evaporation rate for 0.8 Sn-Li at 380 °C:
  - calculated: 5 x 10^{12} /cm^2/sec
  - measured: 2 x 10^{12} /cm^2 /sec
- Sputtering rate for 50 nA beam, Y=0.1, on a spot size of 0.32 mm by 0.32 mm: 8 x 10^{13} /cm^2/sec
- To eliminate evaporation from regions not being struck by the ion beam, a thin tantalum metal sheet with a small hole is floated on top of the liquid Li or liquid 0.8 Sn-Li surface as an evaporation shield.

In-situ cleaving arm design and HV heater

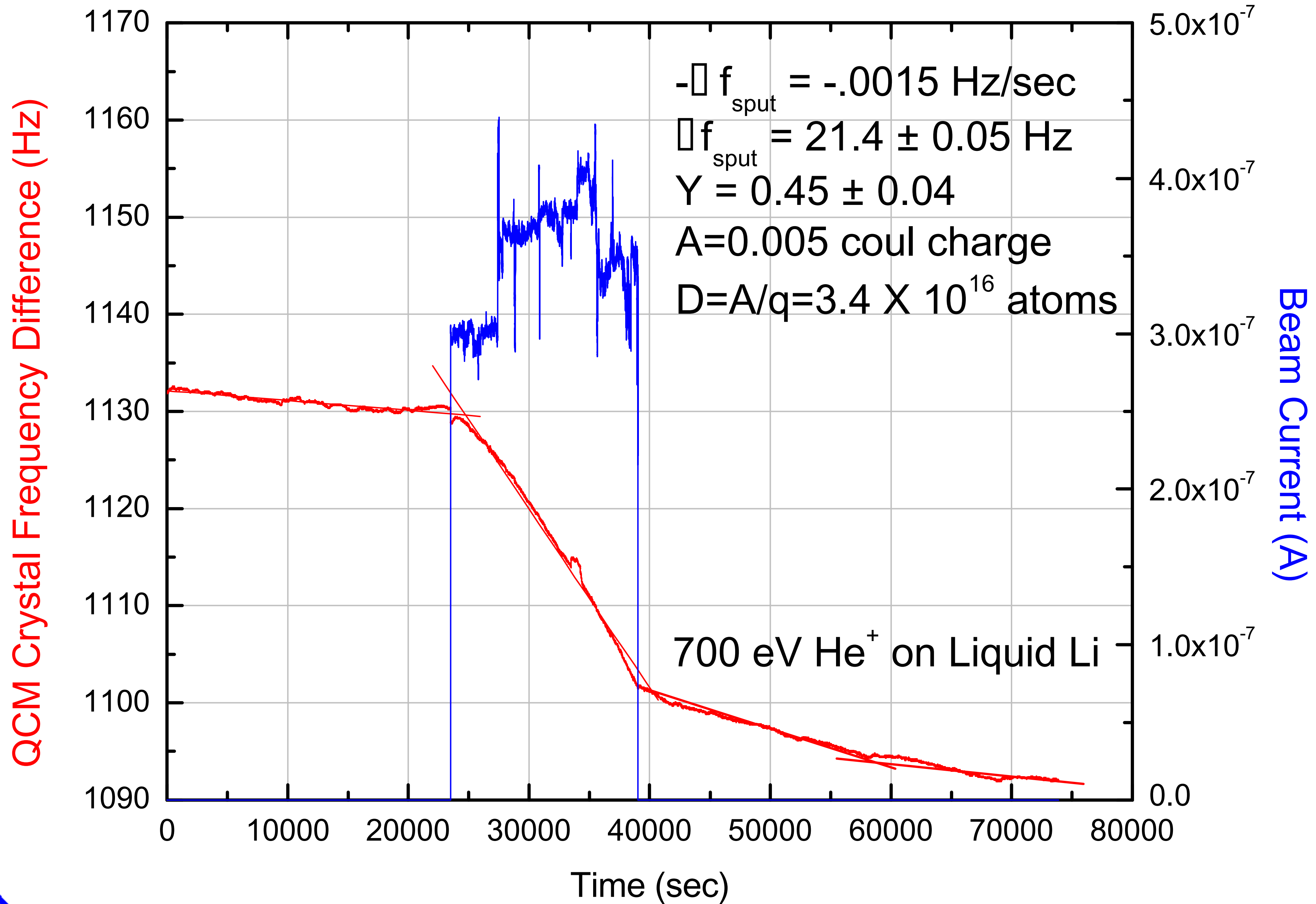
- Cleaving arm is designed to remove thin oxide layer formed on Li layer of liquid tin-lithium or liquid lithium sample
- Surface composition experiments show that Li segregates to the liquid Sn-Li surface[1]



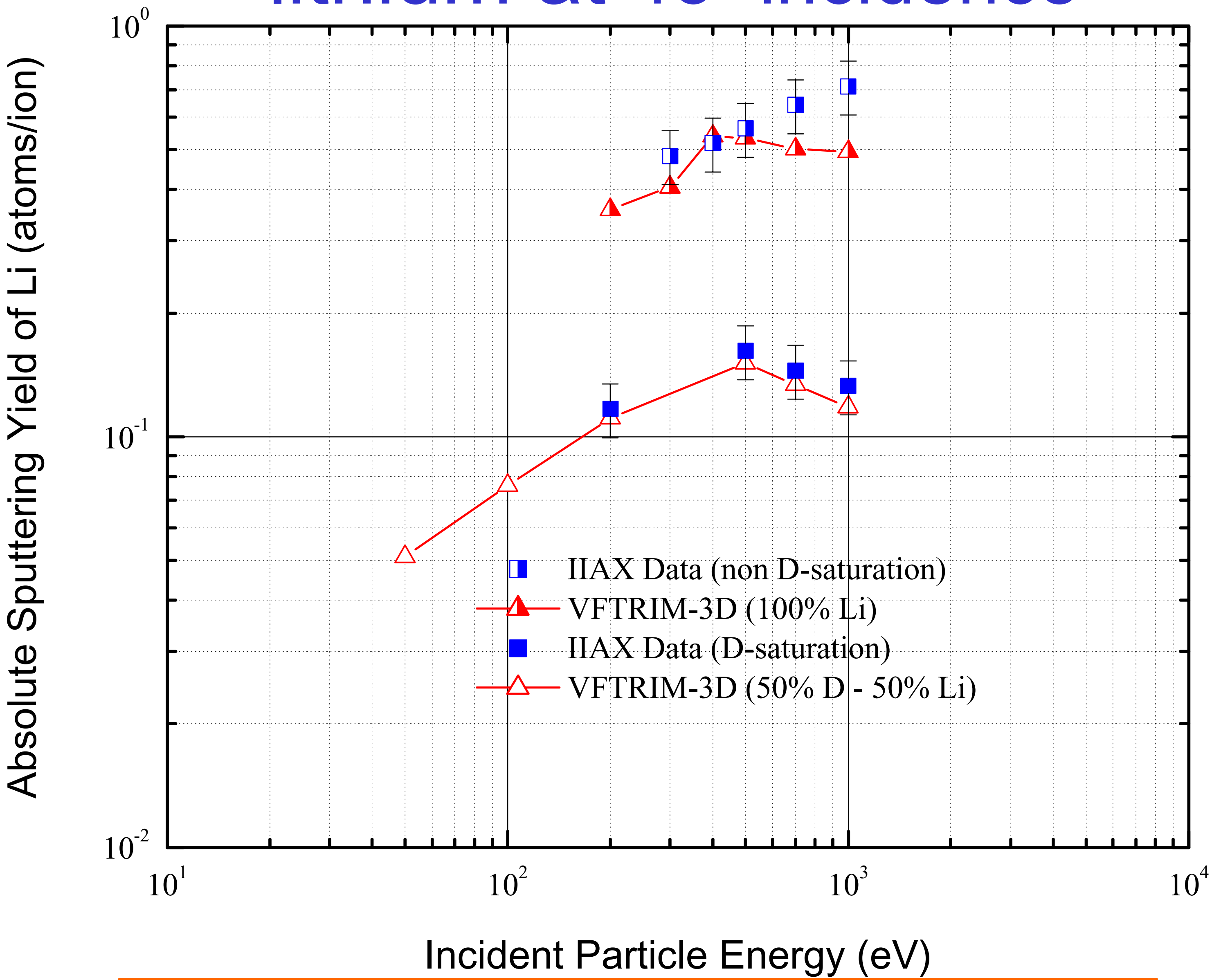
A HV heater was installed inside a BN cup.

1. R. Bastasz and W. Eckstein, J. Nucl. Mater. 290-293 (2001) 19-24

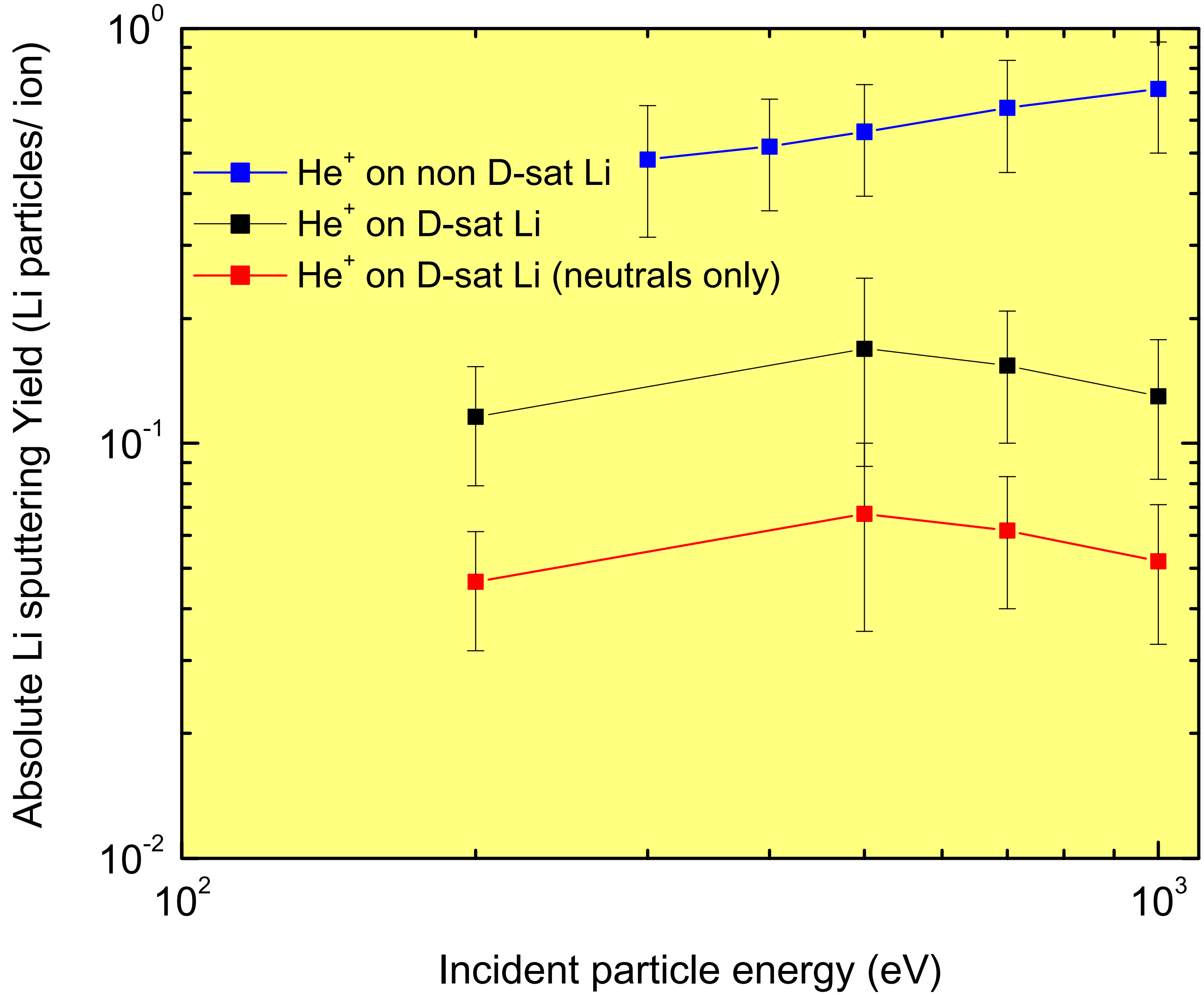
QCM Frequency Difference and Beam Current vs. Time



He$^+$ on D-saturated and non D-sat. lithium at 45° incidence

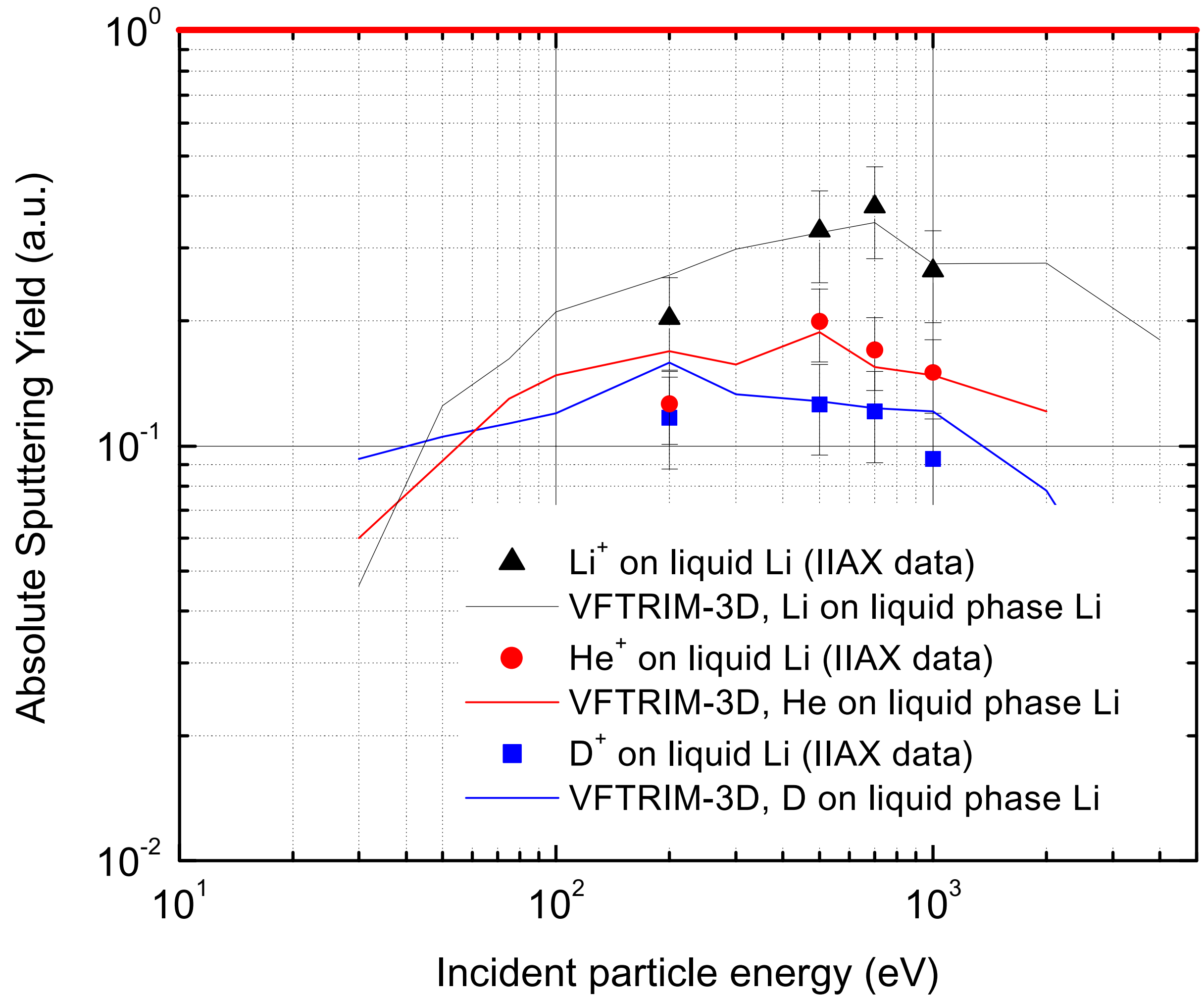


D-treatment effect on lithium sputtering



IIAX experimental and modeling data on liquid lithium erosion

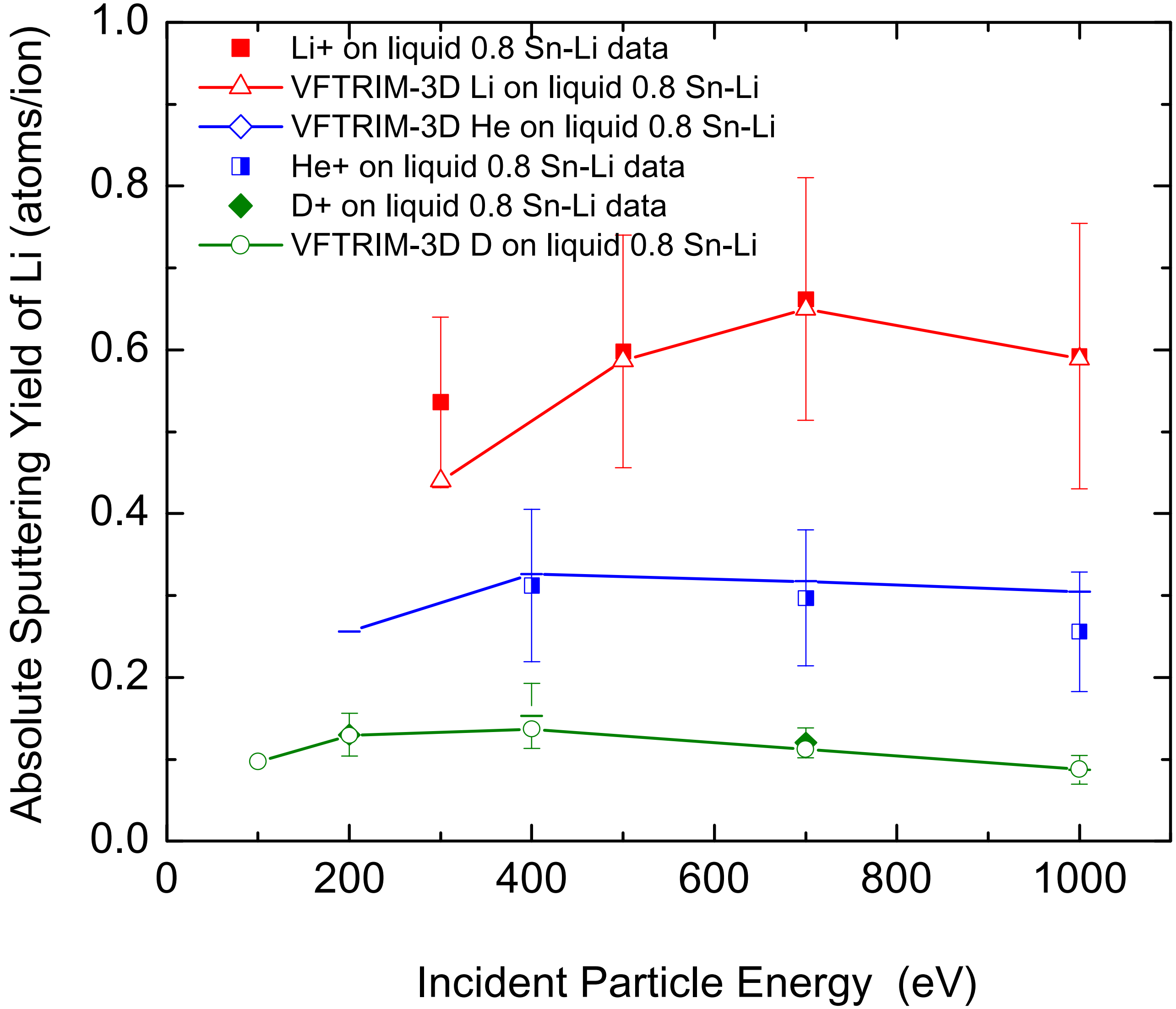
- D-treated lithium yields are well below unity
- Data taken at 45 deg. Incidence and 200 °C surface temperature



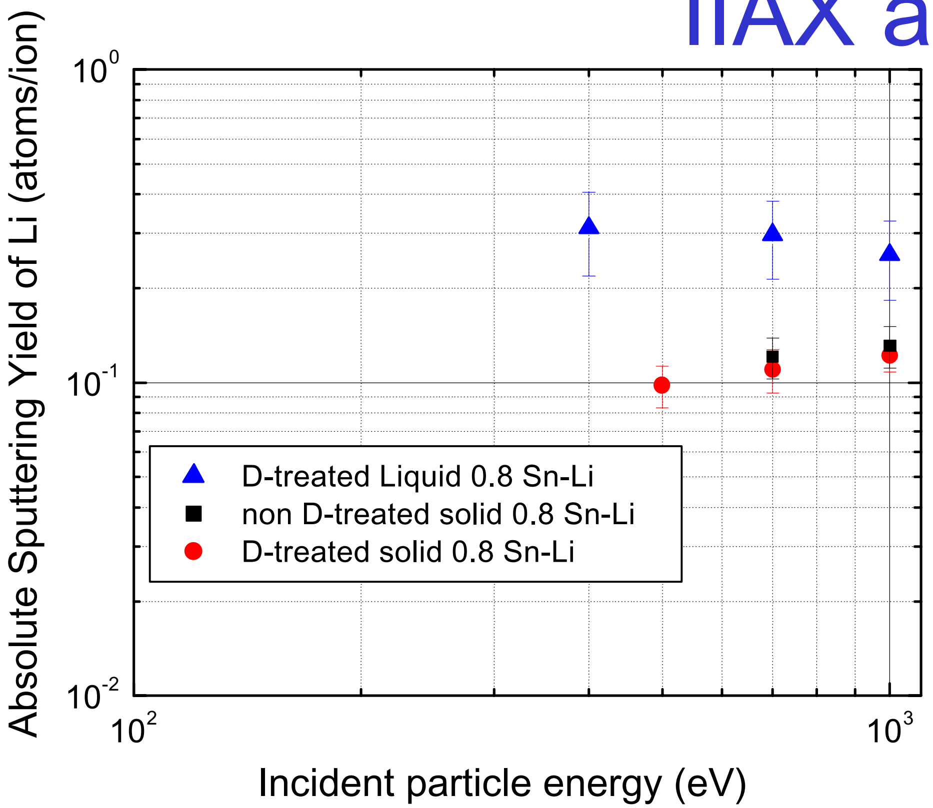
J.P. Allain, M.R. Hendricks and D.N. Ruzic, J. Nucl. Mater. 290-293 (2001) 180

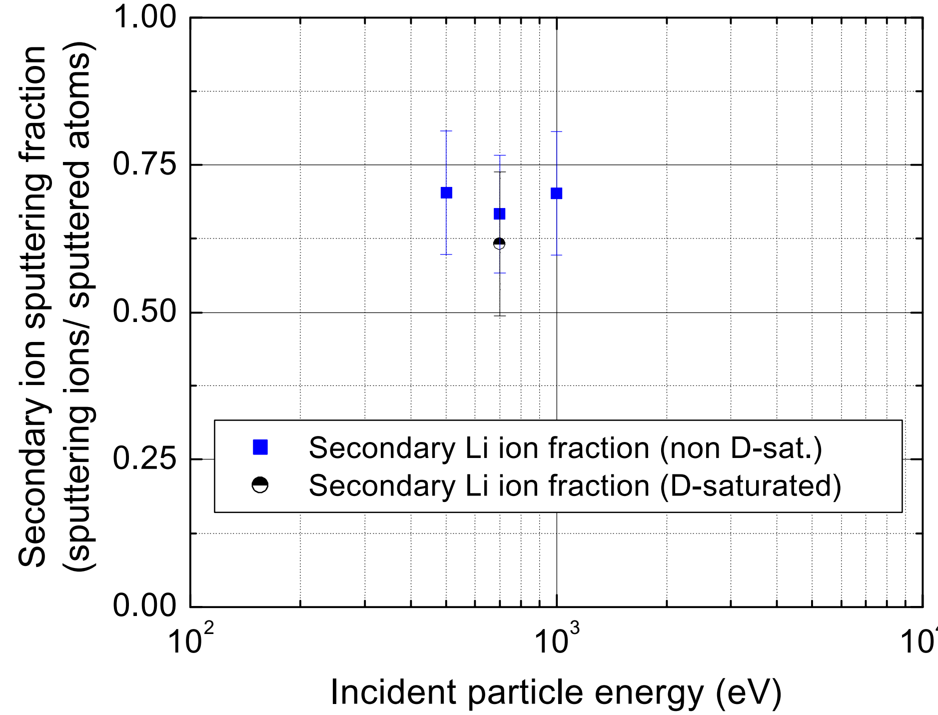
D^+, He^+ and Li^+ bombardment of liquid tin-lithium data and VFTRIM-3D simulation at 45-degree incidence.

(J.P. Allain, M.R. Hendricks, D.N. Ruzic, J.Nucl.Mater. 290-293 (2001) 33-37)



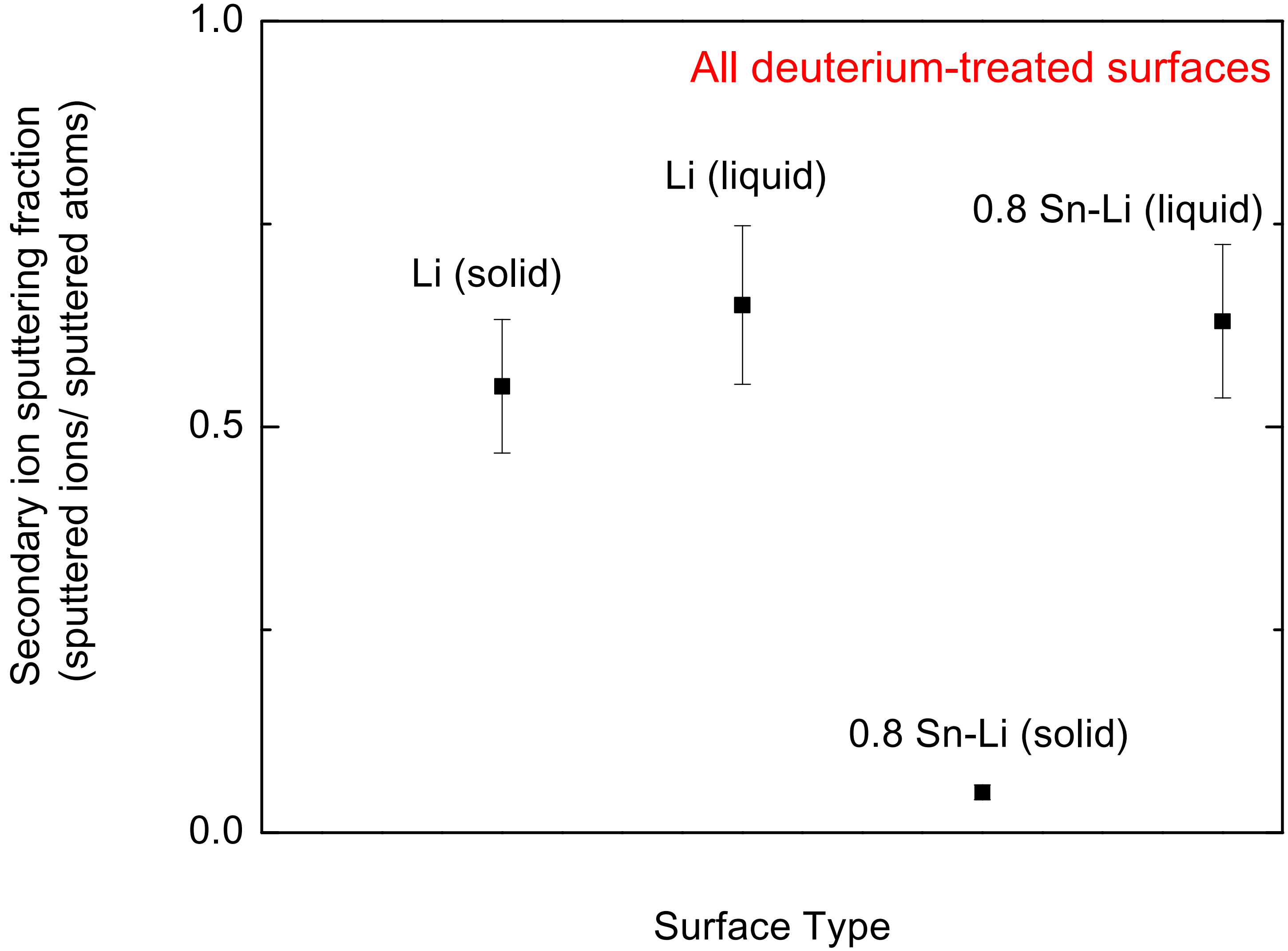
Secondary ion fraction and deuterium-saturation studies of liquid metals in IIAX at UIUC



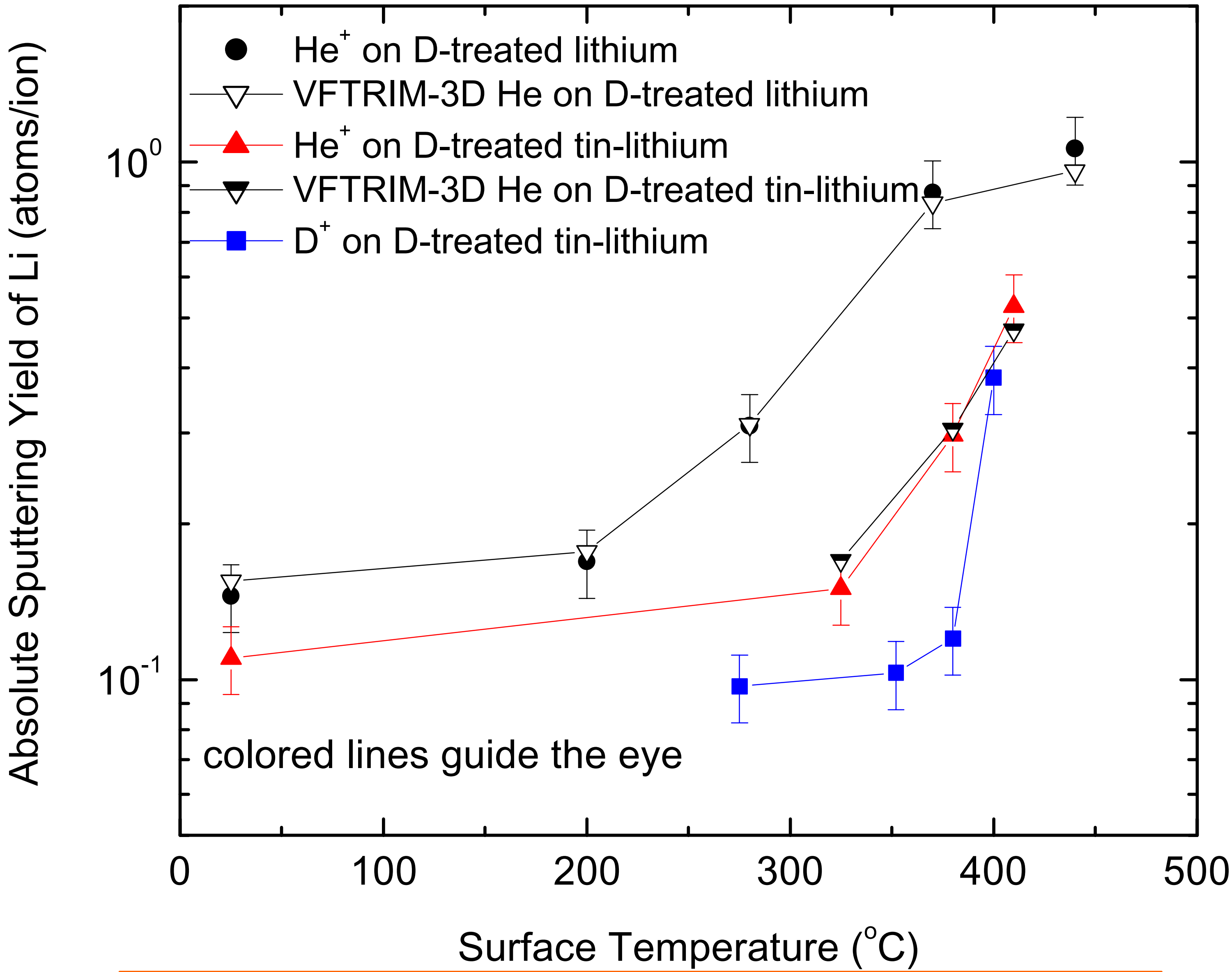


- Saturation of solid and liquid (T/Tm ~ 1) tin-lithium with D atoms results in no effect on the absolute sputtering yield of lithium.
- Ion fraction measurements show that 55-65% of sputtered atoms from D-saturated solid and liquid lithium are in an ionized state.

Secondary Ion Fraction of sputtered Li as a function of surface



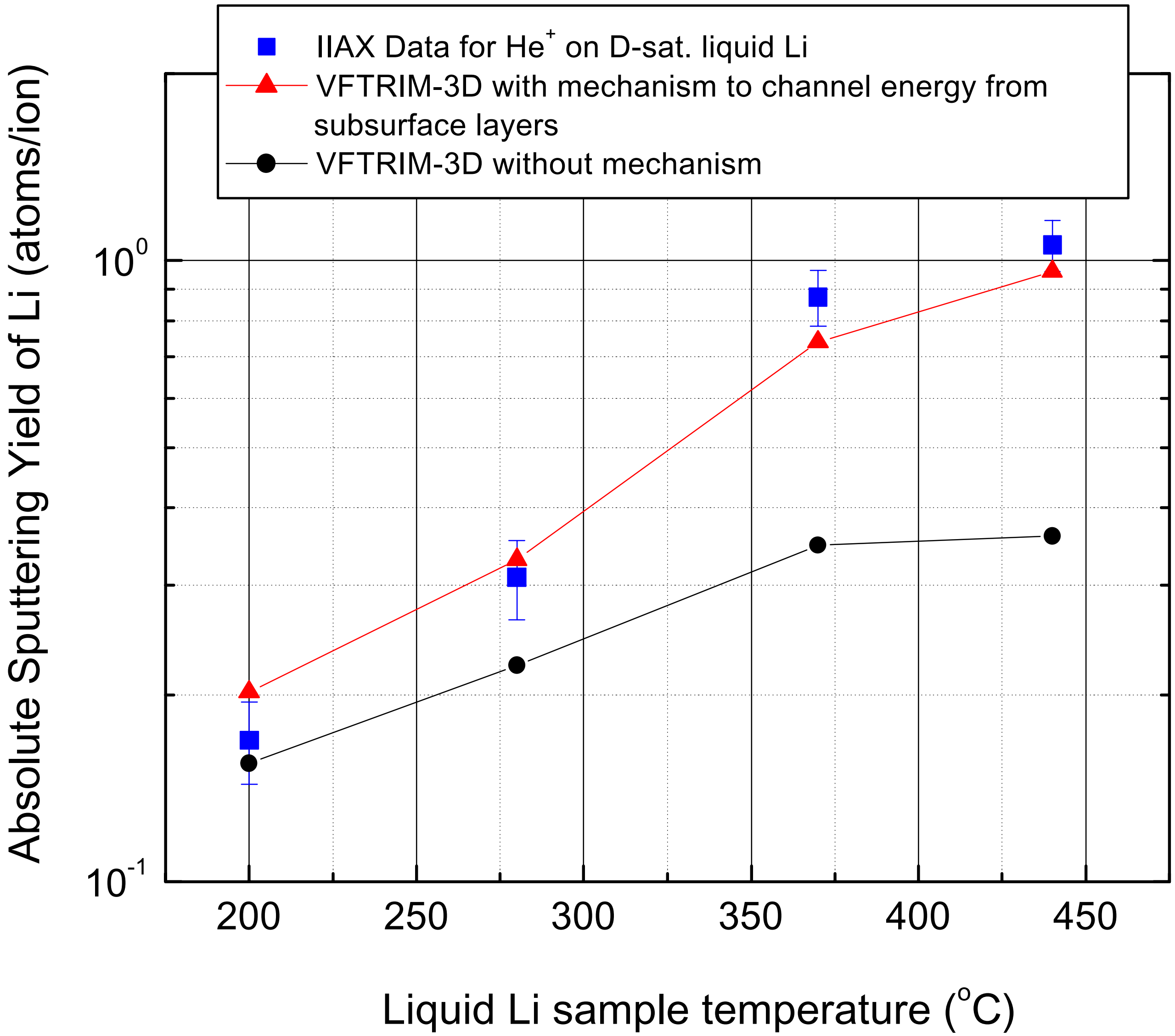
Lithium sputtering yield temperature dependence for lithium and tin-lithium targets



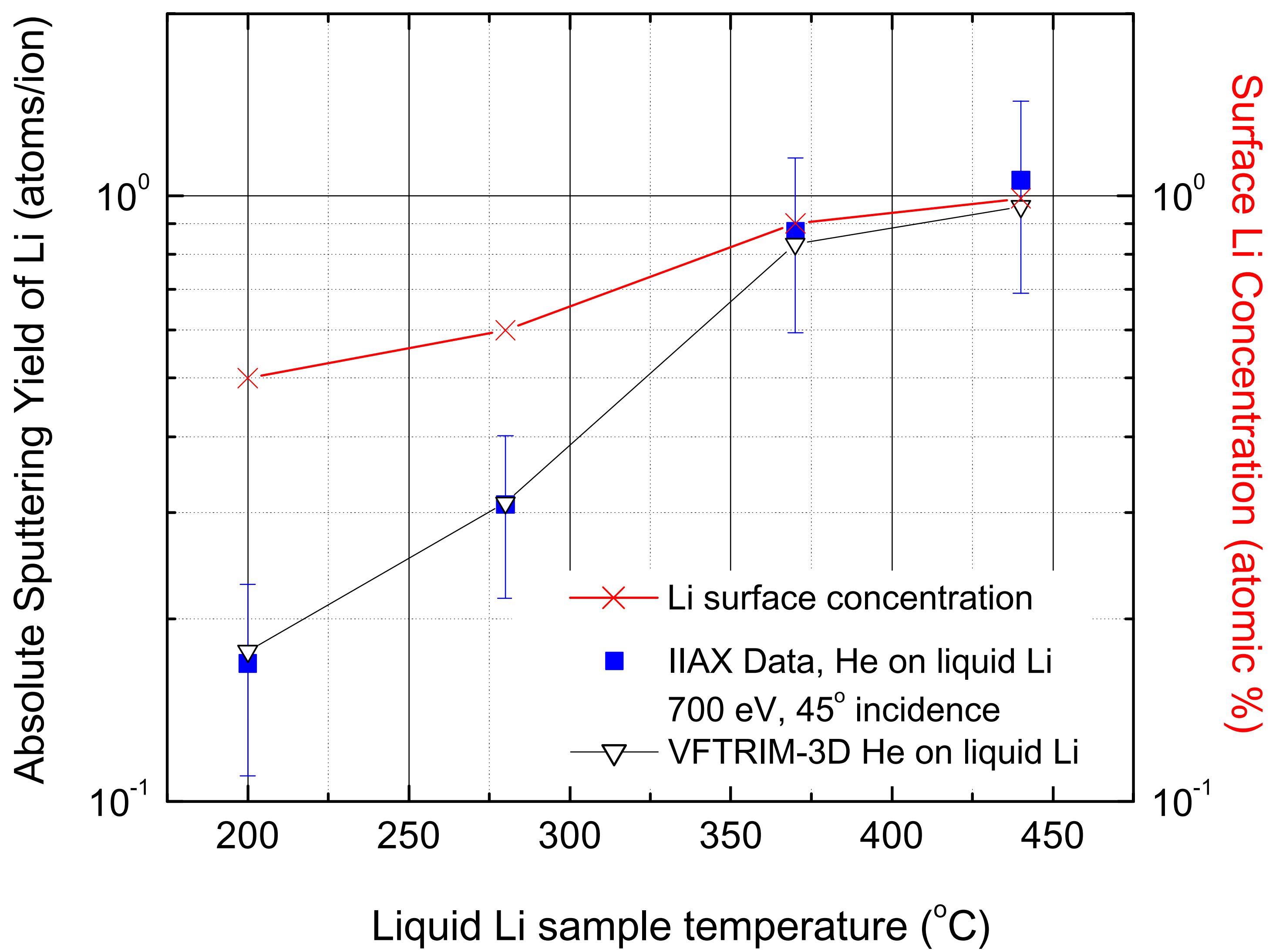
VFTRIM-3D Model for temperature dependence in IIAX liquid lithium sputtering data

- Initially began with surface model for D-treated liquid lithium (presented earlier) at 200 C.
- Deuterium surface concentration varied as T/T_m increases with mechanism for channeling energy from subsurface layers to the top surface layer.
- Surface binding energy calibrated to mean ejected energy of sputtered lithium atoms measured in PISCES-B. Assuming Thompson distribution with sbe ~ $\langle E_{sp} \rangle / 2$. **(R.P. Doerner, et al. J. Nucl. Mater. 290-293 (2001) 166-172.)**
- Density of liquid lithium layer calibrated to experimental data of temperature dependent density (~ weak T function) **(T. Iida and R.I.L. Guthrie, “The Physical Properties of Liquid Metals”, Oxford, 1988)**

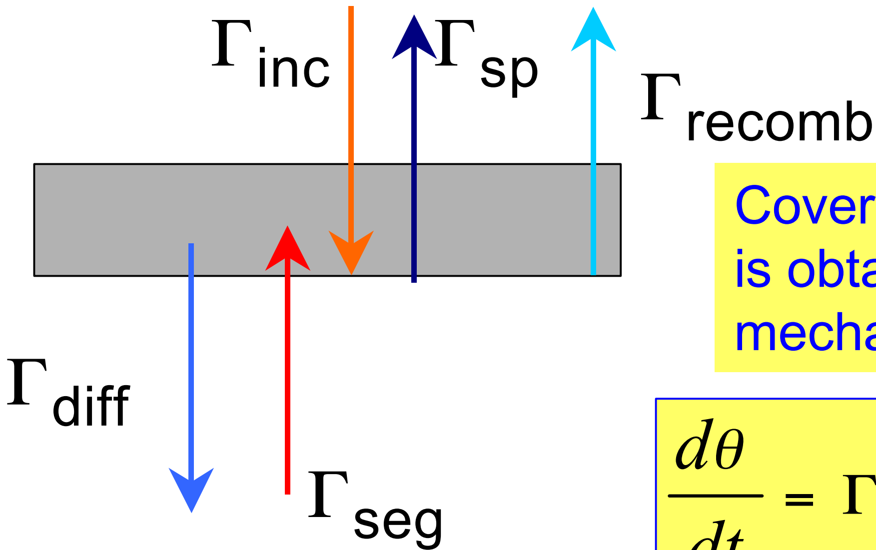
VFTRIM-3D Simulation for He^+ on D-sat. liquid Li as a function of target temperature with IIAX data



Lithium surface concentration of D-treated liquid lithium surfaces as a function of surface temperature



Model to determine deuterium surface concentration as a function of time for a particular temperature



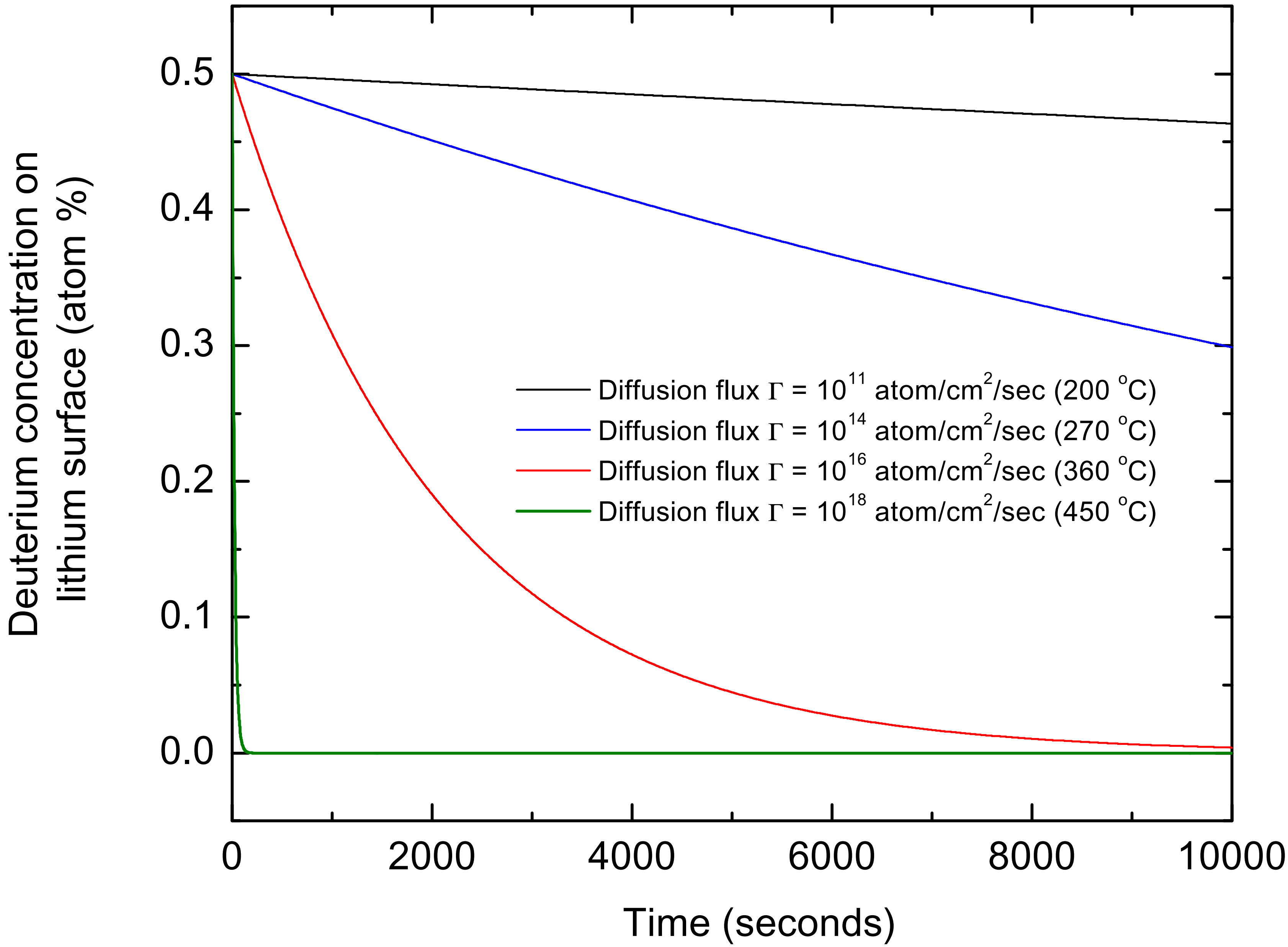
Coverage of deuterium atoms on liquid lithium is obtained accounting for all gain and loss mechanisms

$$\frac{d\theta}{dt} = \Gamma_{imp} + \Gamma_{seg} - \Gamma_{sp} - \Gamma_{diff} - \Gamma_{recomb}$$

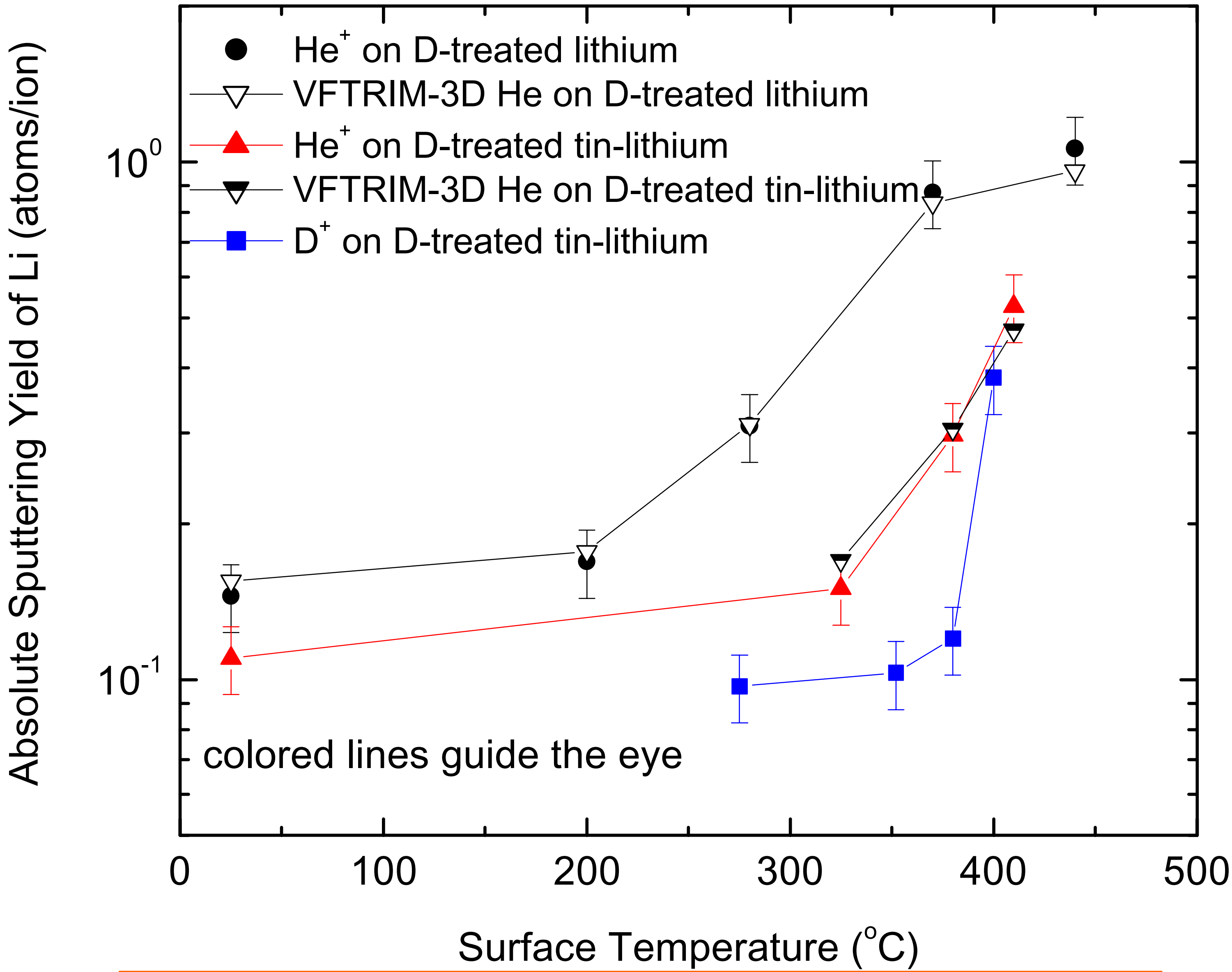
$$\theta(t) = \frac{e^{-k_1 t}\left(k_2\sqrt{k_1} + k_3 Erf\left(\sqrt{k_1 t}\right)\right)}{\sqrt{k_1}}$$

R. Bastasz and W. Eckstein, J. Nucl. Mater. 290-293 (2001) 19-24
R. Kirchheim and S. Hoffmann, Surf. Sci. 83 (1979) 296-300

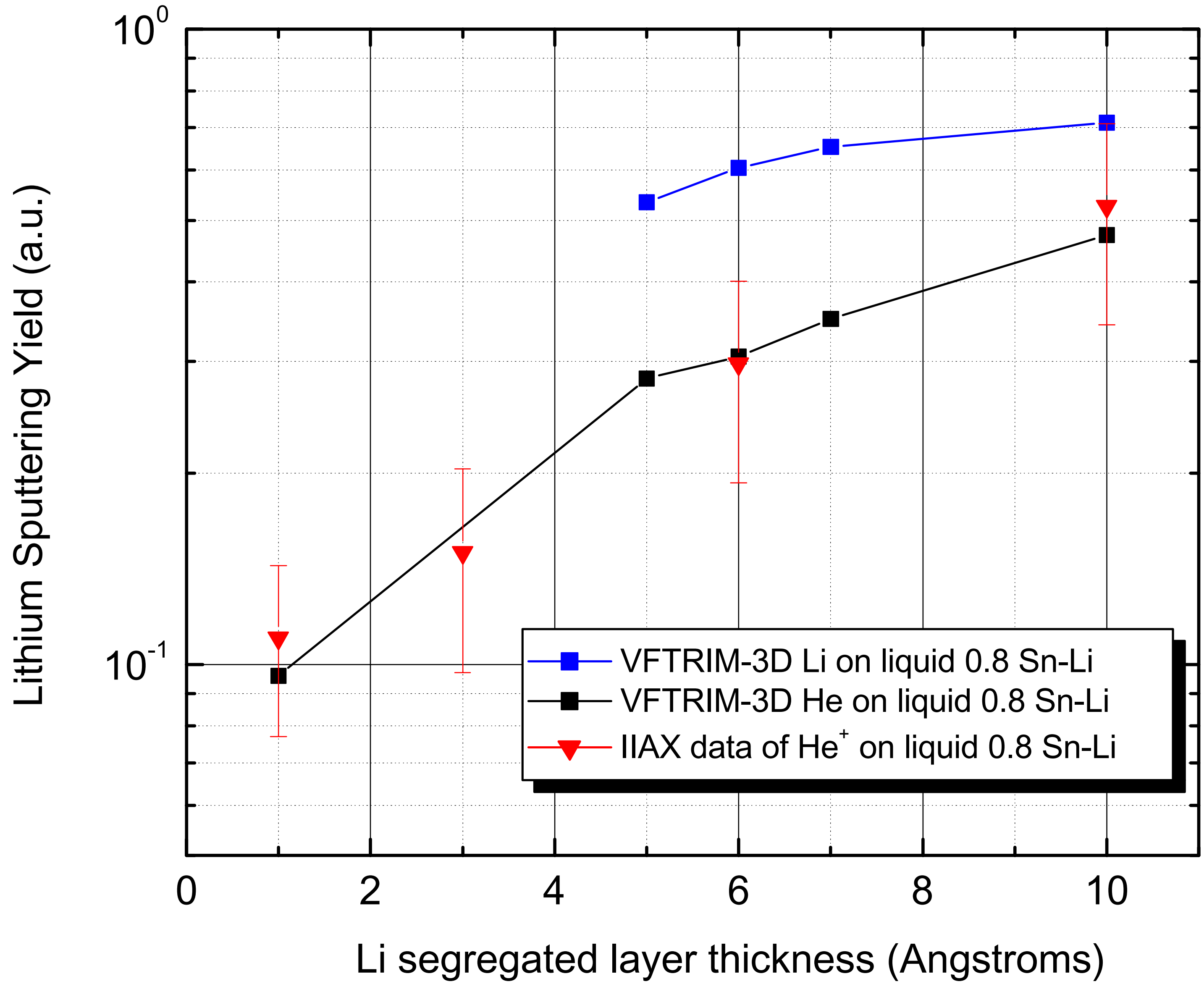
Kirchheim-Hoffmann Surface Model



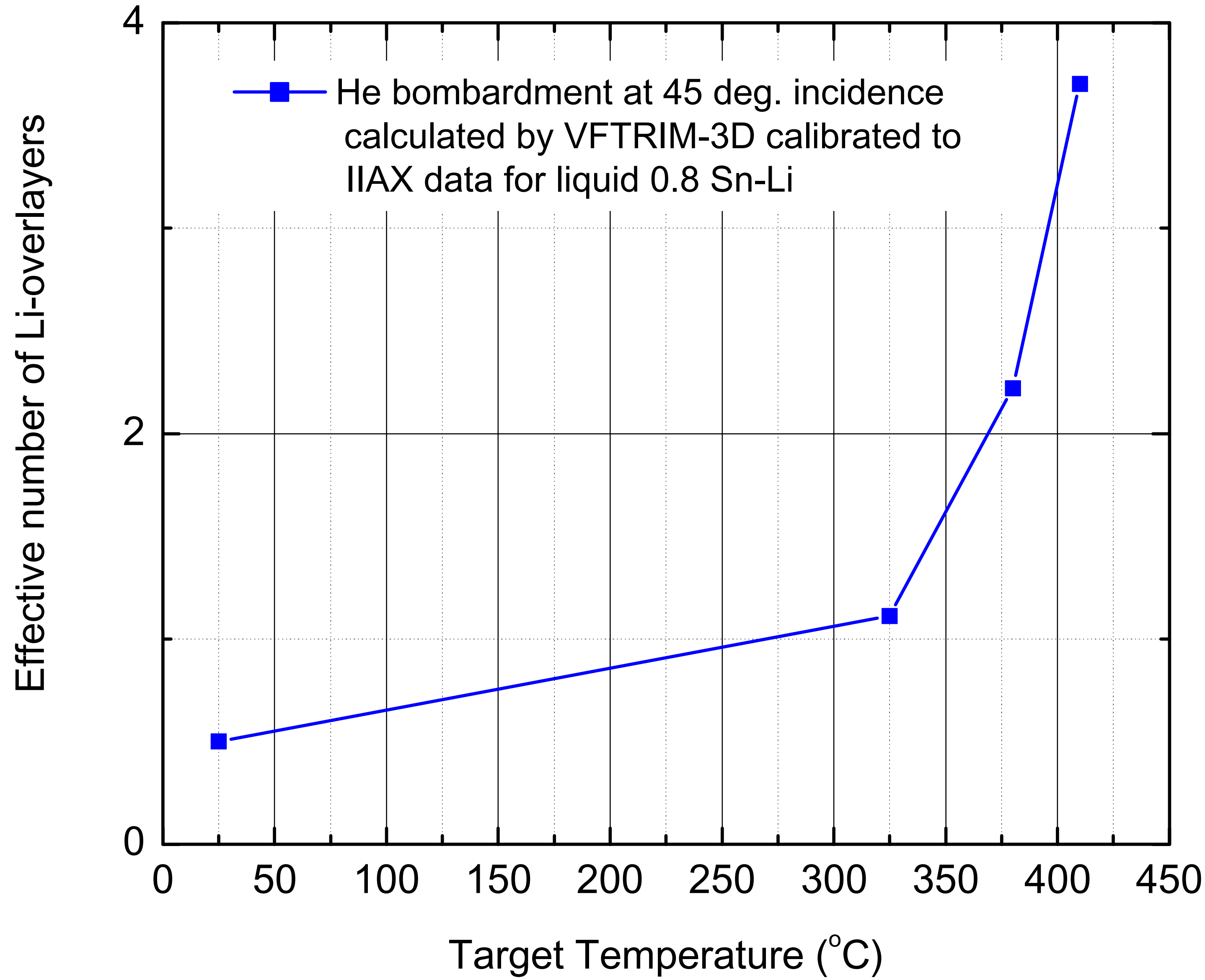
Lithium sputtering yield temperature dependence for lithium and tin-lithium targets



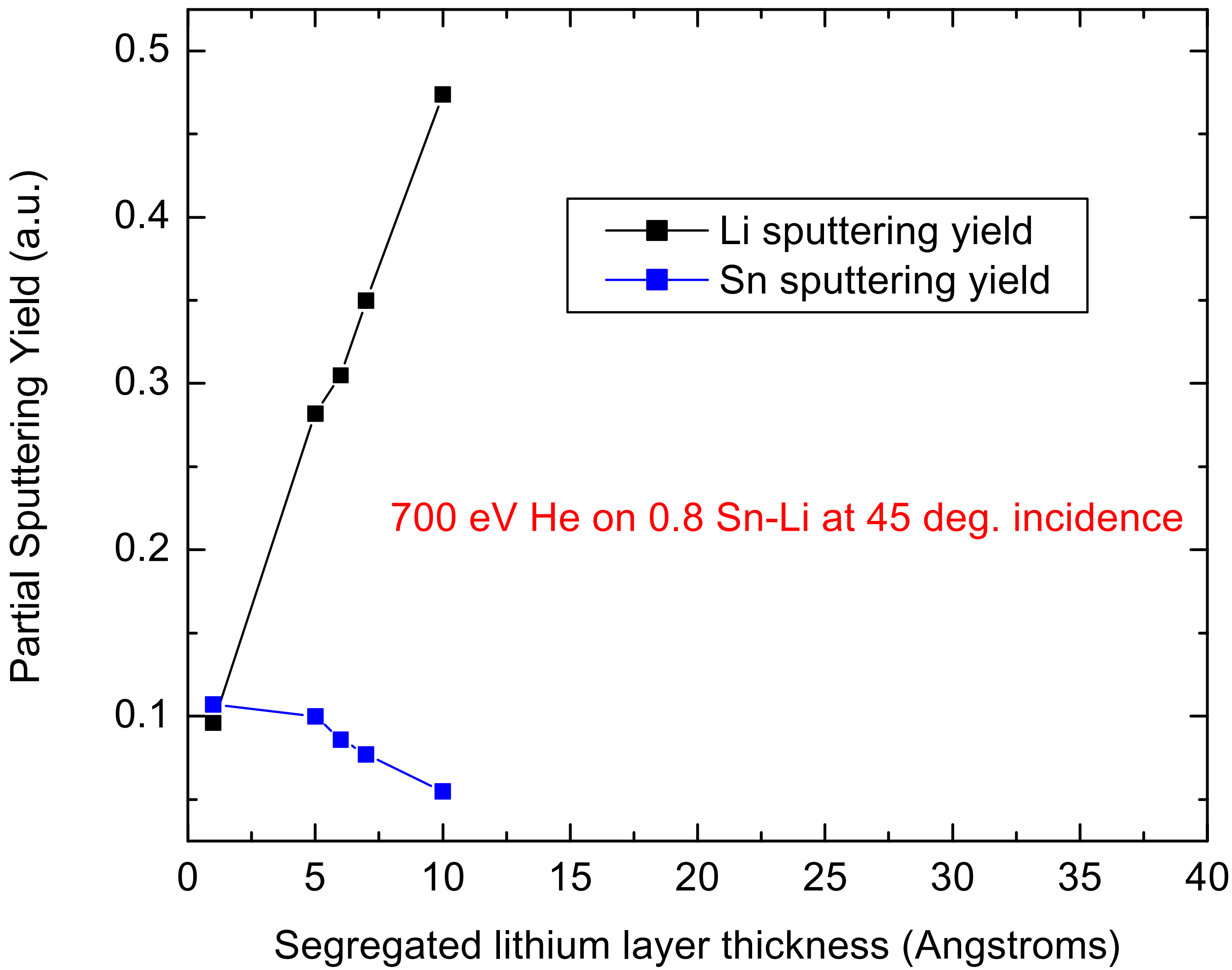
Partial lithium sputtering yield vs segregated lithium layer thickness in 0.8 Sn-Li



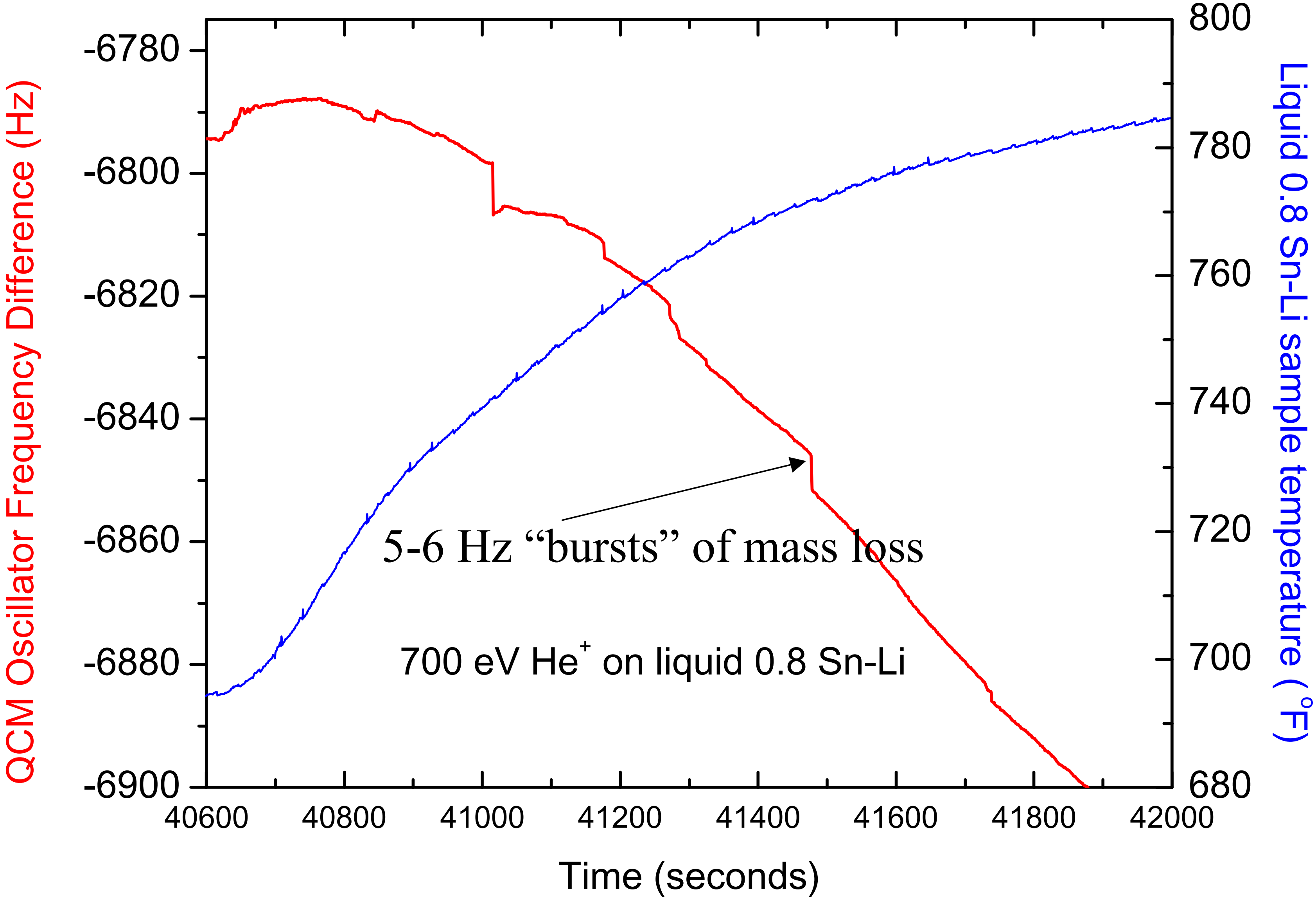
Segregated lithium layer thickness as a function of liquid 0.8 Sn-Li temperature



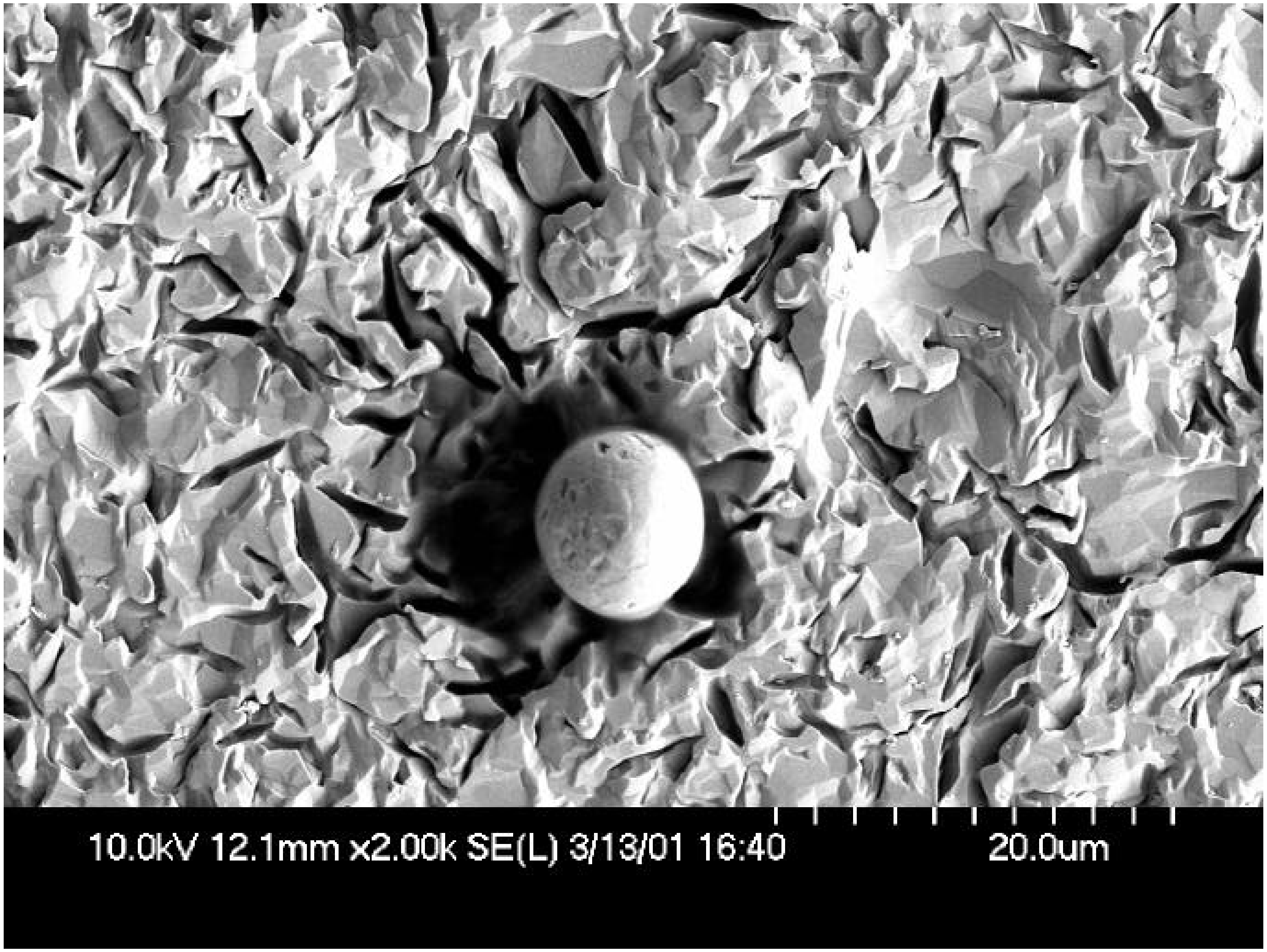
Partial sputtering yields of lithium and tin from He bombardment as a function of segregated lithium layer



QCM Frequency dependence on Liquid 0.8 Sn-Li sample temperature

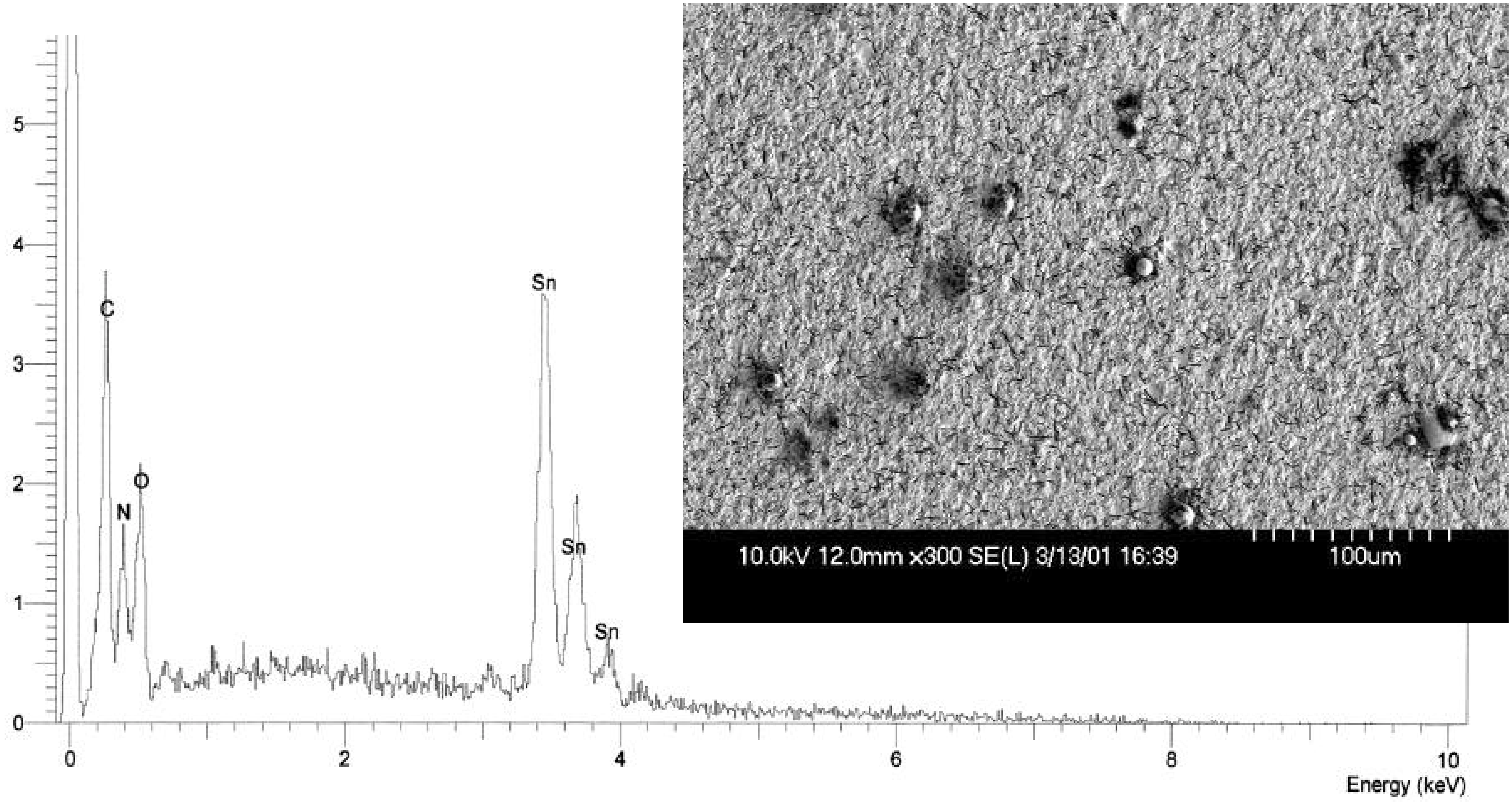


SEM micrographs of macroscopic erosion in IIAX



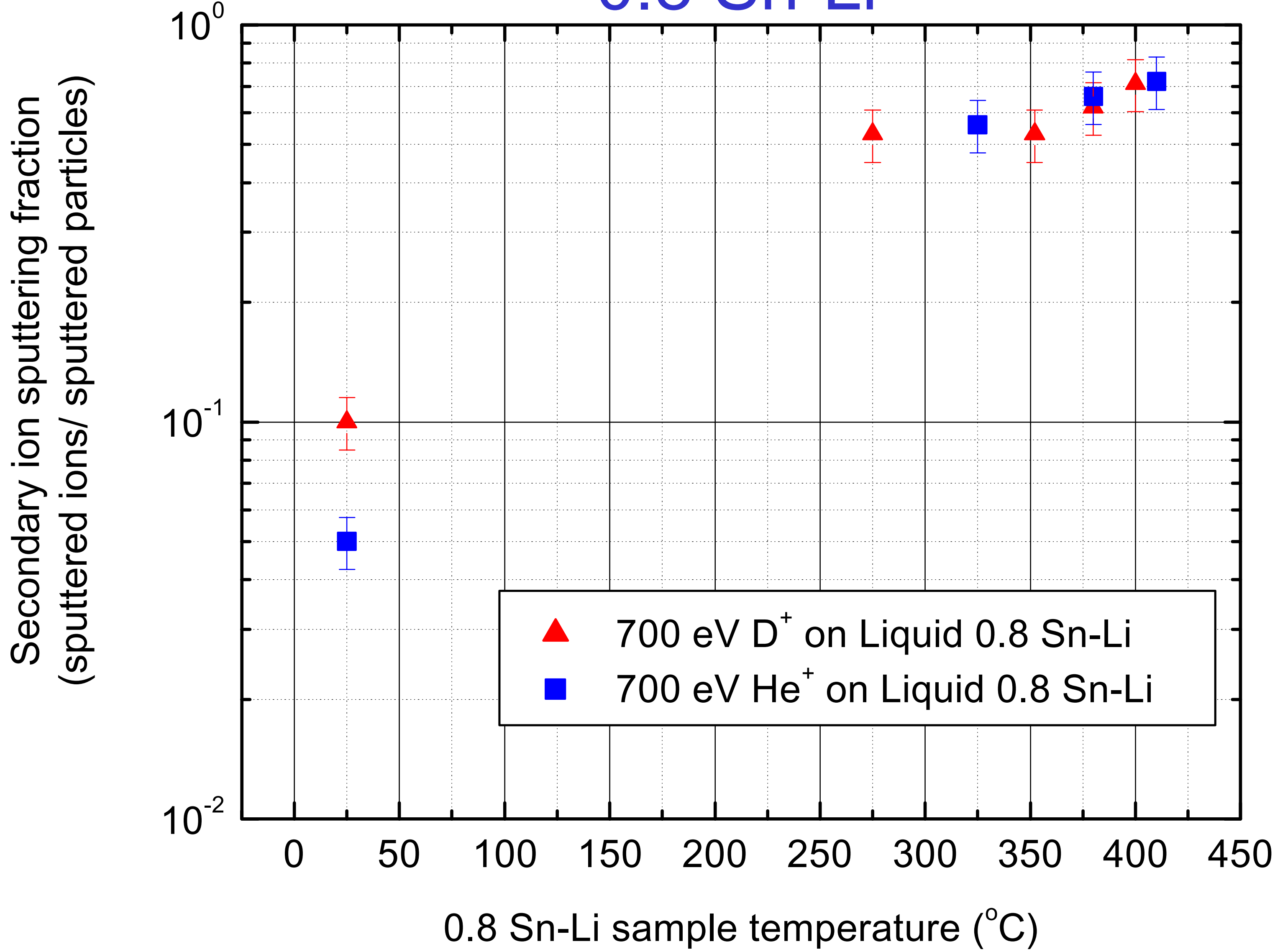
Large particulate ~ 6-7 μm is shown with crater

EDS (X-ray energy dispersive spectroscopy) confirmed that particulates are made of tin



Need to run XPS to determine amount of lithium in particulates

Secondary ion sputtering fraction (Y^{+}_{sp}) dependence on target temperature for Liquid 0.8 Sn-Li



Liquid lithium and tin-lithium dependence on temperature at low incident flux

- Low flux (~ 10^{14} ion/cm^2/sec) not expected to result in dramatic enhanced erosion for temperatures near $T/T_m \sim 1.0$.
  (P. Sigmund and M. Szymonski, Appl. Phys. A 33 (1984) 141-152)
- Erosion enhancement is a result of combined mechanisms: local enhanced evaporation (net decrease in ejected sputtered energy), segregation, diffusion and possible bubble formation
- Key: liquid surface stratification…strong temperature dependence?
- Other key: understand transition from microscopic to macroscopic erosion (static vs dynamic liquid metal surfaces)
- Finally: Identify conditioning techniques that can address enhanced erosion, i.e. can Y^{+}_{sp} increase, surface treatment (deuterium, etc…)

Important mechanisms to consider in liquid metal sputtering (not exclusive)

Surface Stratification
S.A. Rice, P. Pershan, W.P. Morgan

Bubble formation in liquid metals
L.B. Begrambekov, A. Hassanein

Thermal Sputtering: local enhanced liquid-metal evaporation
R. Kelly, M.W. Nelson, R.S. Nelson, P. Sigmund

Damage Distribution Theory: Gibbsian segregation, Radiation-enhanced diffusion, preferential sputtering
P. Sigmund, N. Lam, R. Kelly, A.R. Krauss, D.M. Gruen, J. Bohdansky, W. Eckstein, etc

Self-consistent model which addresses most of these mechanisms
HEIGHTS, A. Hassanein (Argonne National Laboratory)

Temporal and spatial scales relevant to liquid-metal sputtering

- Temporal Scales
  - Diffusion, segregation processes
  - Local temperature rise of impacted lattice region
  - Preferential sputtering
  - Desorption and recombination rates
  - Thermal spike and bubble formation
- Spatial Scales
  - Depth of origin of sputtered atoms
  - Depletion surface region
  - Range of ions
  - Thermal spike radius
  - Stratified layer thickness
  - Short-range order of liquid metal

FLIRE (Flowing Liquid Surface Illinois Retention Experiment)

- FLIRE will provide fundamental data on the retention and pumping of He, H, and other gases in flowing liquid surfaces.

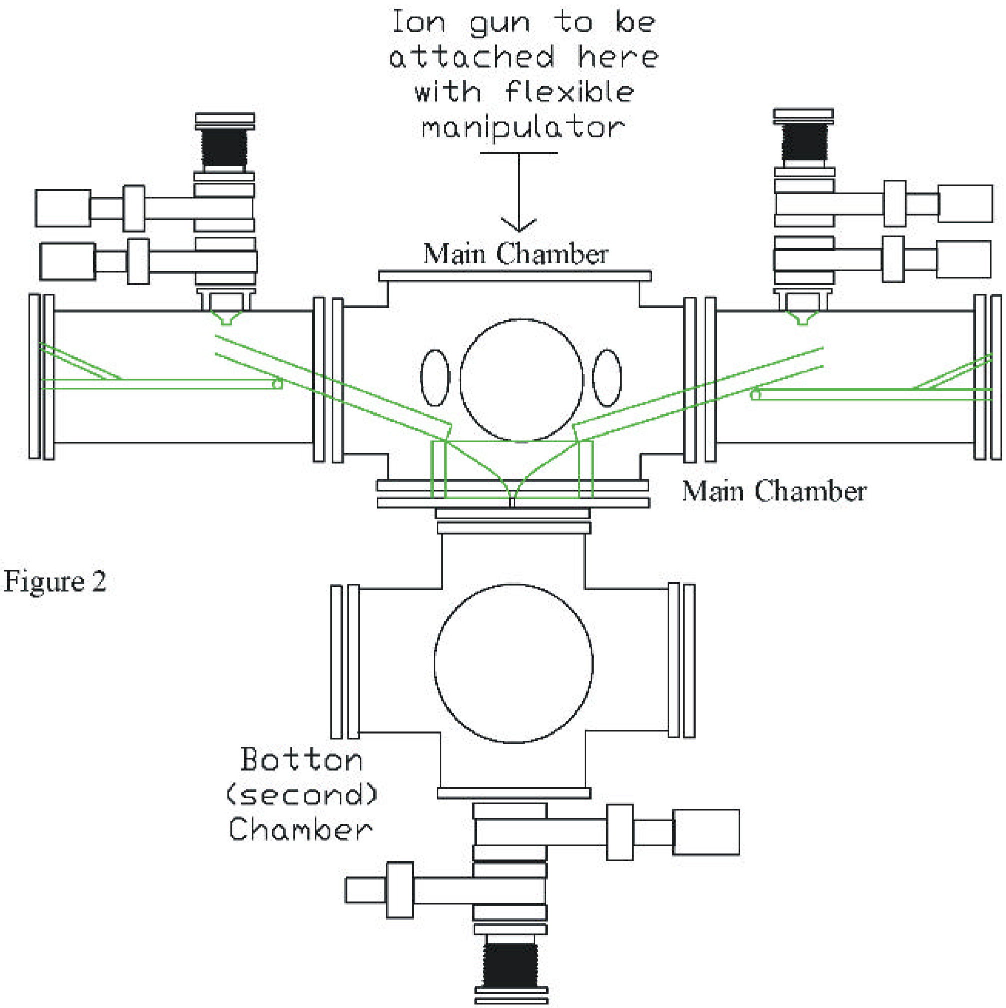
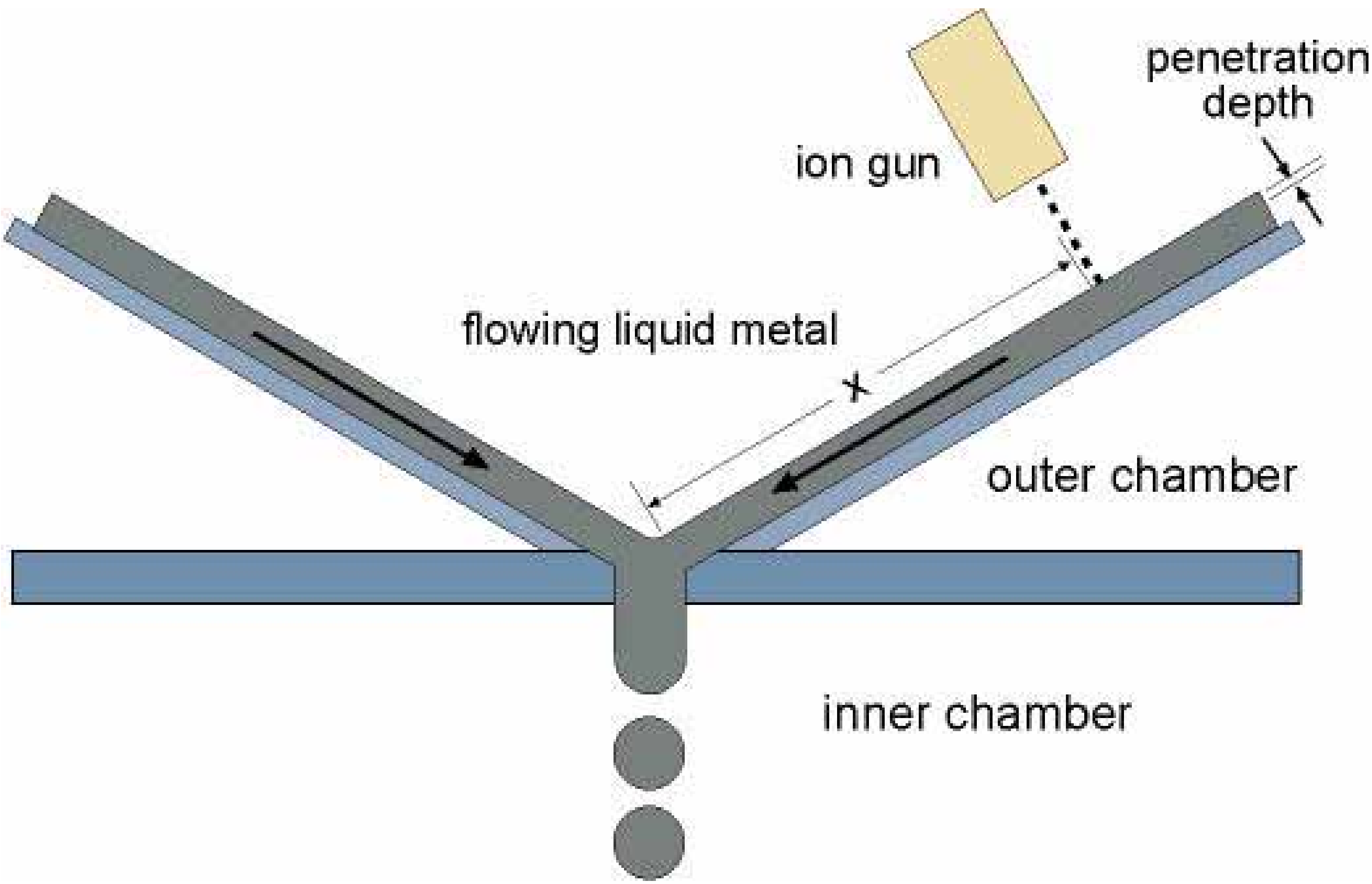


Figure 2

FLIRE concept



Conclusions

- Solid and liquid lithium sputtering yields measured in IIAX-UIUC show less-than unity self-sputtering yields.
- Measurements of liquid eutectic tin-lithium sputtering yields are larger than both measured solid and liquid pure lithium sputtering yields in IIAX.
- Ion fraction of sputtered species for liquid lithium and liquid tin-lithium are 55%-65% compared to <10% for solid phase tin-lithium.
- Deuteration of lithium surface results in a ~ 60% decrease in absolute sputtering yield of lithium.

Conclusions (cont.)

- Bombardment of liquid lithium and liquid tin-lithium show dramatic increase in absolute sputtering yield as temperature is increased.
- Sputtered ion fraction shows modest increase with sample temperature
- New models need to account for surface stratification and its effect on physical sputtering of candidate liquid metals

Future Work Plan

- Experiments to understand temperature dependence of physical sputtering yield
  - Saturation and pre-treatment effects
  - Understand: desorption, segregation and diffusion mechanisms
  - Thermal spikes
  - Search for bubble burst events
- Ion-induced secondary electron yields
- Study other candidate materials (i.e. Sn)

Acknowledgements

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