ELM – Simulating Plasma Gun Proposal

David Ruzic, Robert Stubbers, Travis Gray and Ben Masters

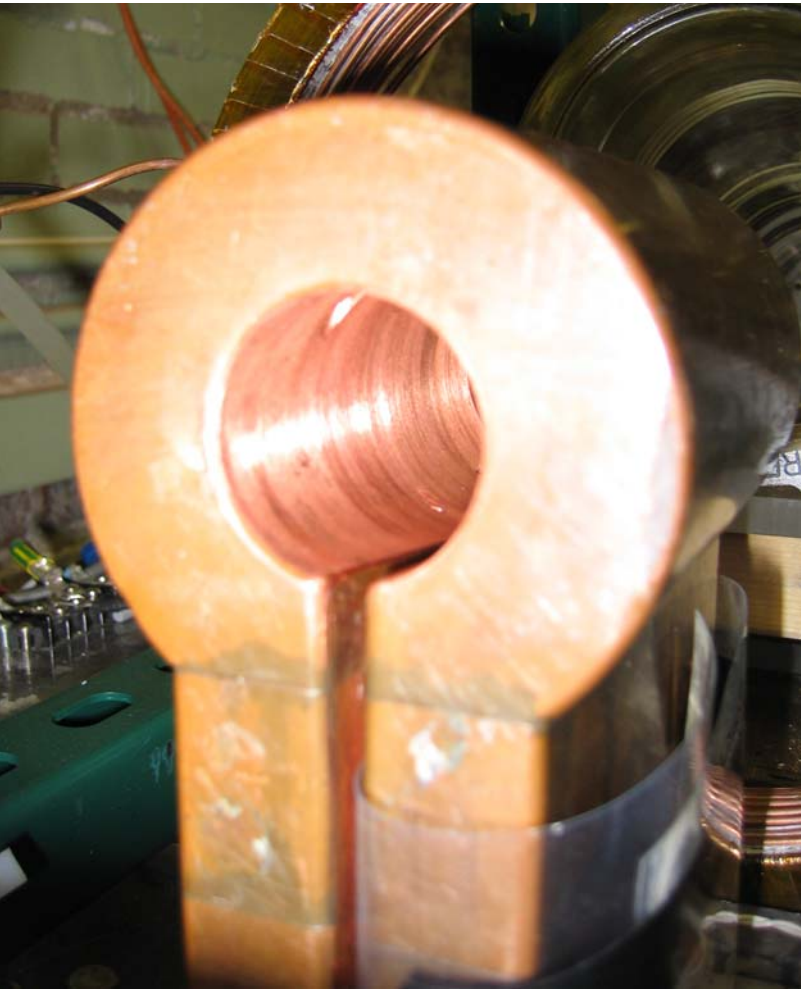
University of Illinois at Urbana Champaign

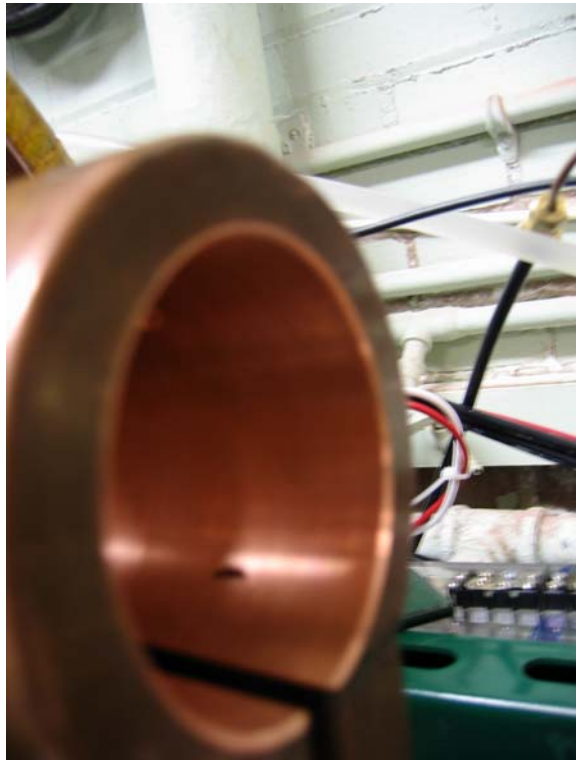
New Directions for Advanced Computer Simulations and Experiments in Fusion-Related Plasma-Surface Interactions (PSIF)
ORNL, Oak Ridge, TN; March 21-23, 2005

ELM – Simulating Plasma Gun Proposal Design Goals

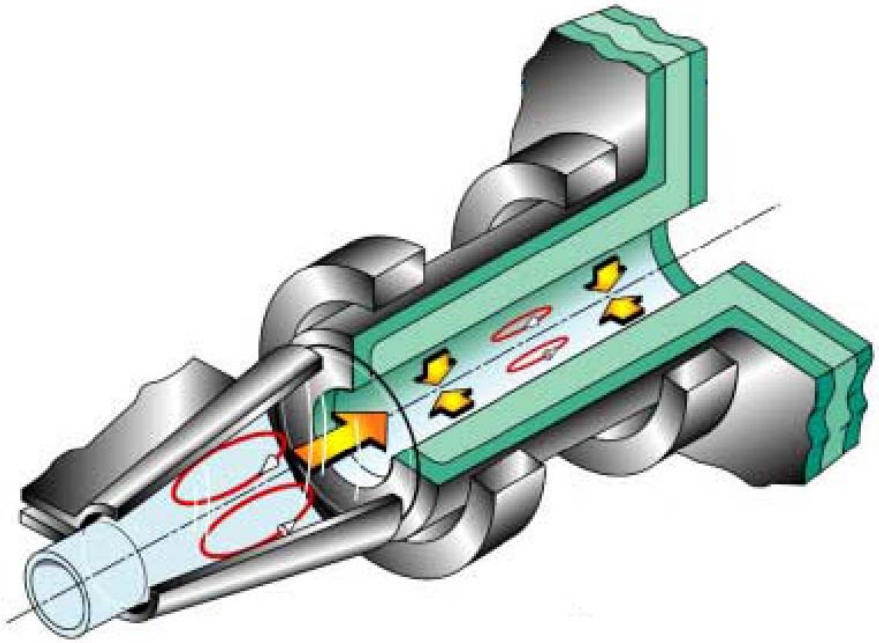
- Comparison of anticipated parameters to NSTX (short term) and ITER (long term) Type-I ELMs

ELM Parameter	ITER	NSTX	UIUC (proposed)
Power Loading	~10 MJ/m^2	< 1 MJ/m^2	1 MJ/m^2
ELM Event Frequency	~1-10 Hz	10-20 Hz	single shot
Total ELM Duration	~0.1-1 ms	~1 ms	~ 0.5 ms
Blob Subfrequency	~10-100 kHz	~10 kHz	~ 10 kHz
Plasma Temperature During ELM (~$T_{pedestal}$)	1-2.5 keV	100 eV	100 eV
Plasma Density During ELM (~$n_{pedestal}$)	~10^{19} m^{-3}	~10^{19} m^{-3}	~10^{19} m^{-3}
Magnetic Field Strength At Divertor (~B_t	~1-5 T	~0.5 T	0.4 T

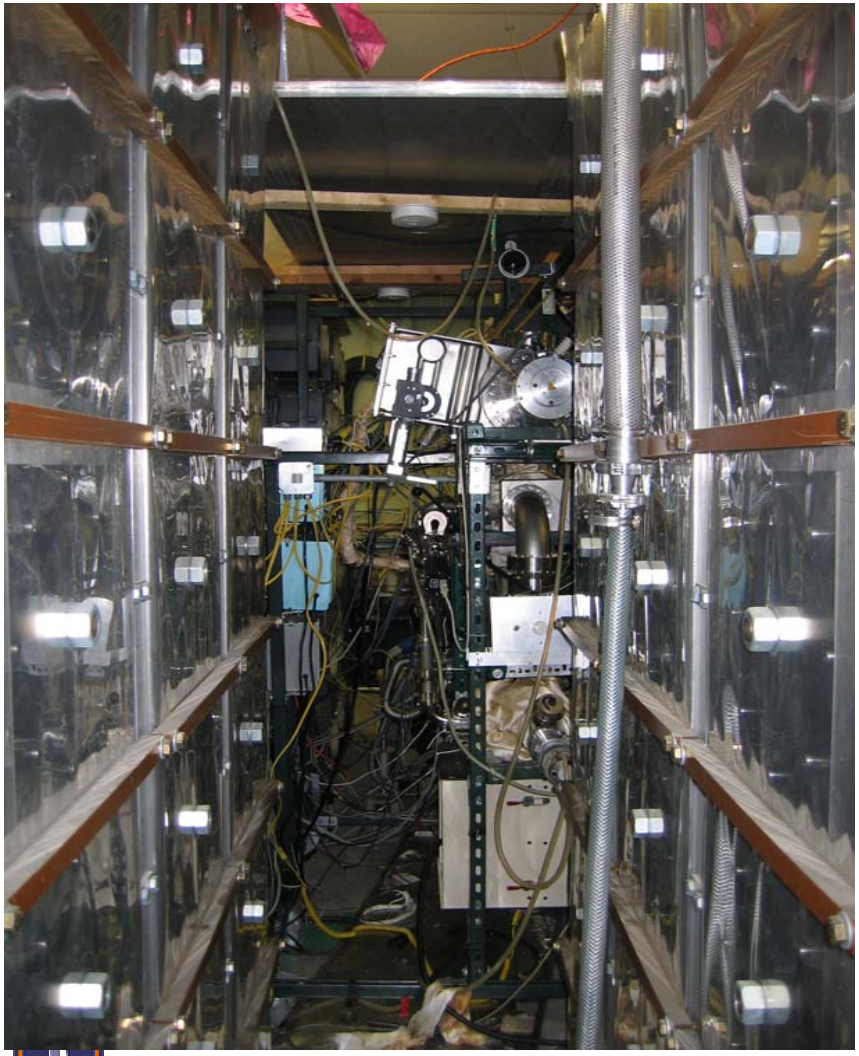




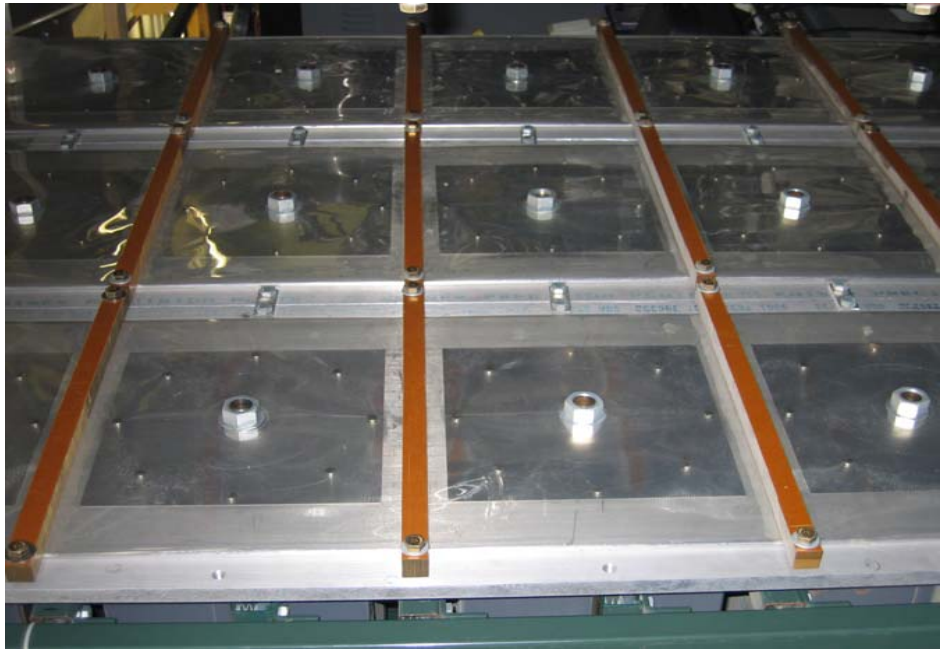
Conical Cross-Section similar to FRC formation / translation

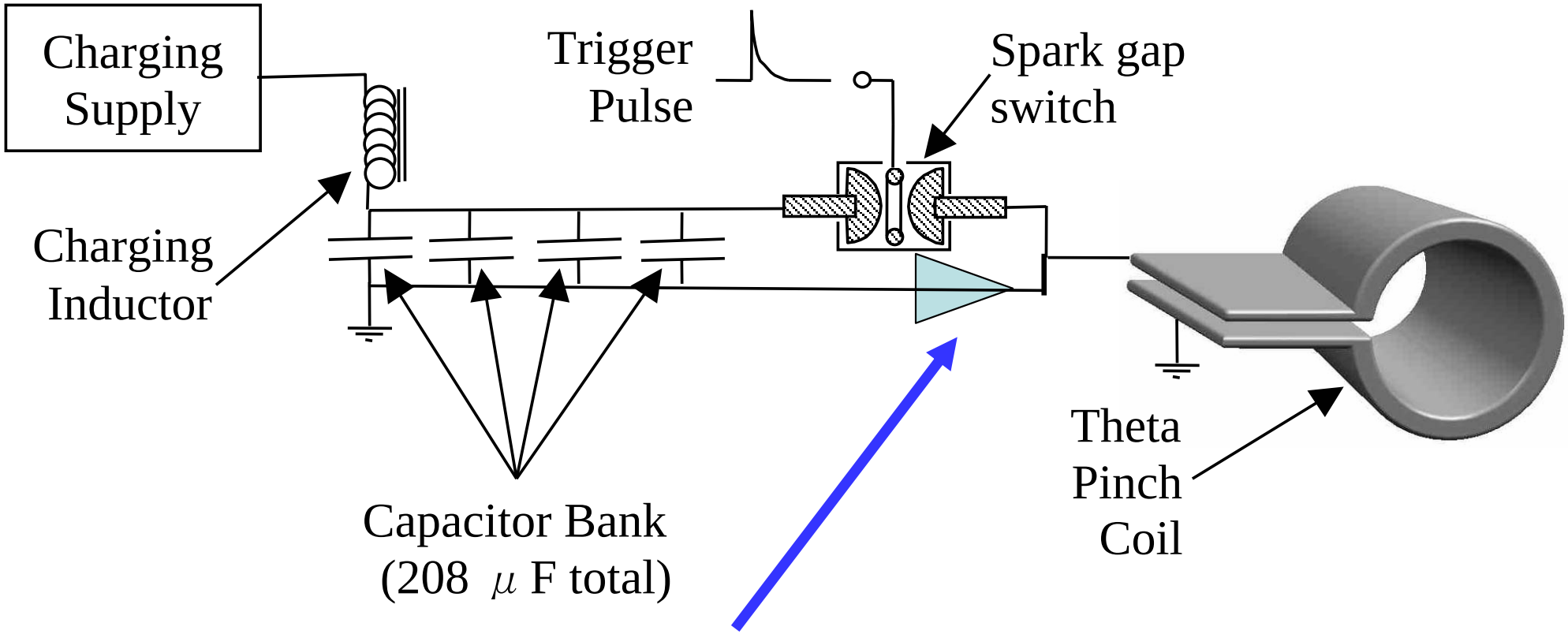


Energy Available



- 250 kJ Capacitor Bank in existence at Illinois of low inductance
- high voltage



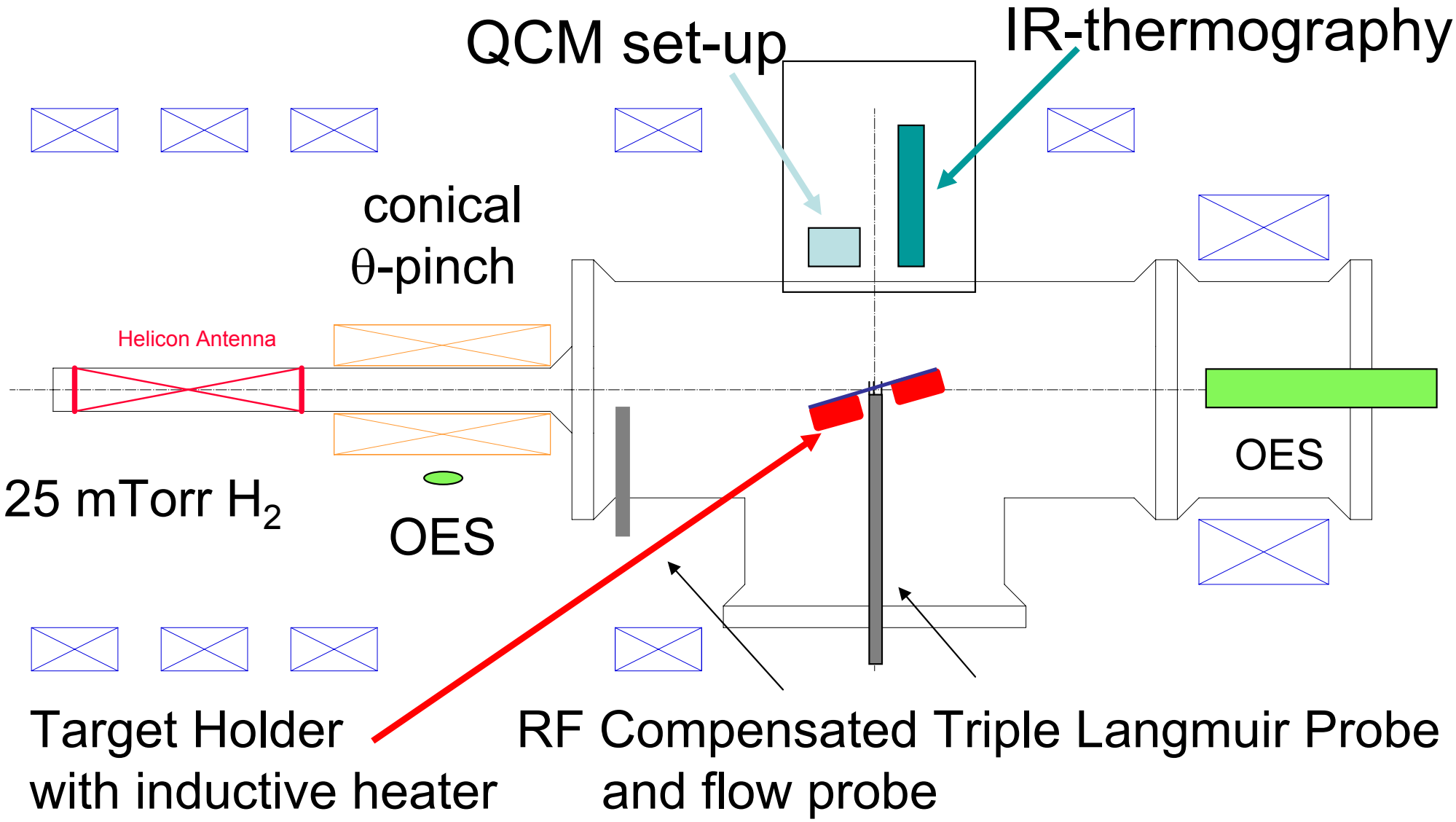


Need high-powered diode to allow trigger from one capacitor group to lie on top of trigger of next capacitor group.

ESP-Gun Features

- Will use pulsed fields to get relevant field strengths – ten times higher than present.
- Target will be at grazing incidence to field and angle can be varied – as in a fusion device.
  - Target bias not used to get energy deposition
- Inductive heater to control initial temperature
  - Plasma not used to control temperature

ESP-gun Diagram



MD and Hybrid-MD Simulations of Plasma-Surface Interactions

David Ruzic

M.D. Coventry, H. Qiu, D.A. Alman

Plasma-Material Interaction Research Group

University of Illinois at Urbana-Champaign

New Directions for Advanced Computer Simulations and Experiments in Fusion-Related Plasma-Surface Interactions (PSIF)

ORNL, Oak Ridge, TN; March 21-23, 2005

Outline

- ***Molecular dynamics*** work @ UIUC
  - Previous focus on H_xC_y simulation for carbon machines (D.A. Alman)
  - Current work:
    - Li self-bombardment (Li-Li interacting)
    - H, D → pure Li (H-Li and Li-Li interacting)
    - H, D → Li / LiD (H-Li, Li-Li, and H-H interacting)
  - Temperature-dependence investigation
  - Kinetic information extracted from MD
- ***Monte Carlo simulations:*** VFTRIM used as a baseline simulation
- Experimental indications of temperature-induced sputtering yield enhancement in solid and liquid materials
- What can MD tell us about the temperature-dependent PSI details that are conventionally missed?
- ***MD-MC hybrid concept***
  - Goals
  - Computational Techniques
  - Results
- ***Future work***

Motivation

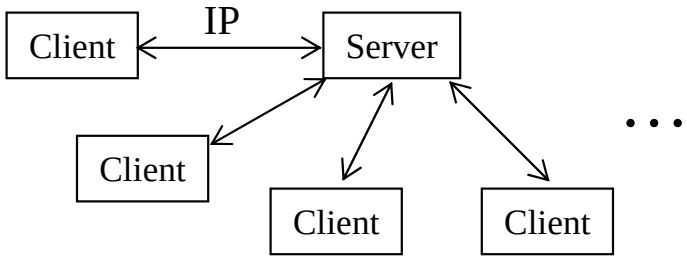
- Modeling of candidate PFC's under ion irradiation help predict material responses to fusion machine conditions
- Comparison to experimental results to validate and benchmark models
- Detailed analysis of results and computer "experiments" lead to the improvement of the fundamental understanding of the processes involved (sputtering, reflection, etc.)

Molecular dynamics effort at Illinois

- Principal code employed: *MolDyn*
  - Designed for covalent systems (specifically C, Si, and H -containing materials) using the Brenner potential & modified by D.A. Alman for modeling solid and liquid lithium
  - Random impact location used for each flight
  - Runs continuously on our research group's distributed computing system (20+ processors) maximizes available computing resources without necessitating change of platform

Client-Server Implementation

- Server
  - On one central machine
  - Distributes work to clients
  - Compiles results



- Client
  - Installed on all PCs in the research group (20+)
  - Procedure
    - (1) Contacts server to get next task to do
    - (2) Contacts server to get required input files
    - (3) Runs the MolDyn code
    - (4) Returns the result to the server

loop

- Controlled as a loop by a Java code, based on an open source client-server model
- Runs in background, uses otherwise idle CPU cycles

MolDyn computational details

- Classical molecular dynamics code
  - Fixed timestep Beeman integrator[1]
  - Berendsen method to control temperature[2]
- Nominal code configurations
  - (exact parameters depend on particulars of system modeled)
  - Time step: *0.1 to 0.2 fs*
  - Time scale: typically run for *500 fs*
  - Interaction length limited to cutoff distance to optimize computational time (e.g., 5.75 Å for Li-Li potential)

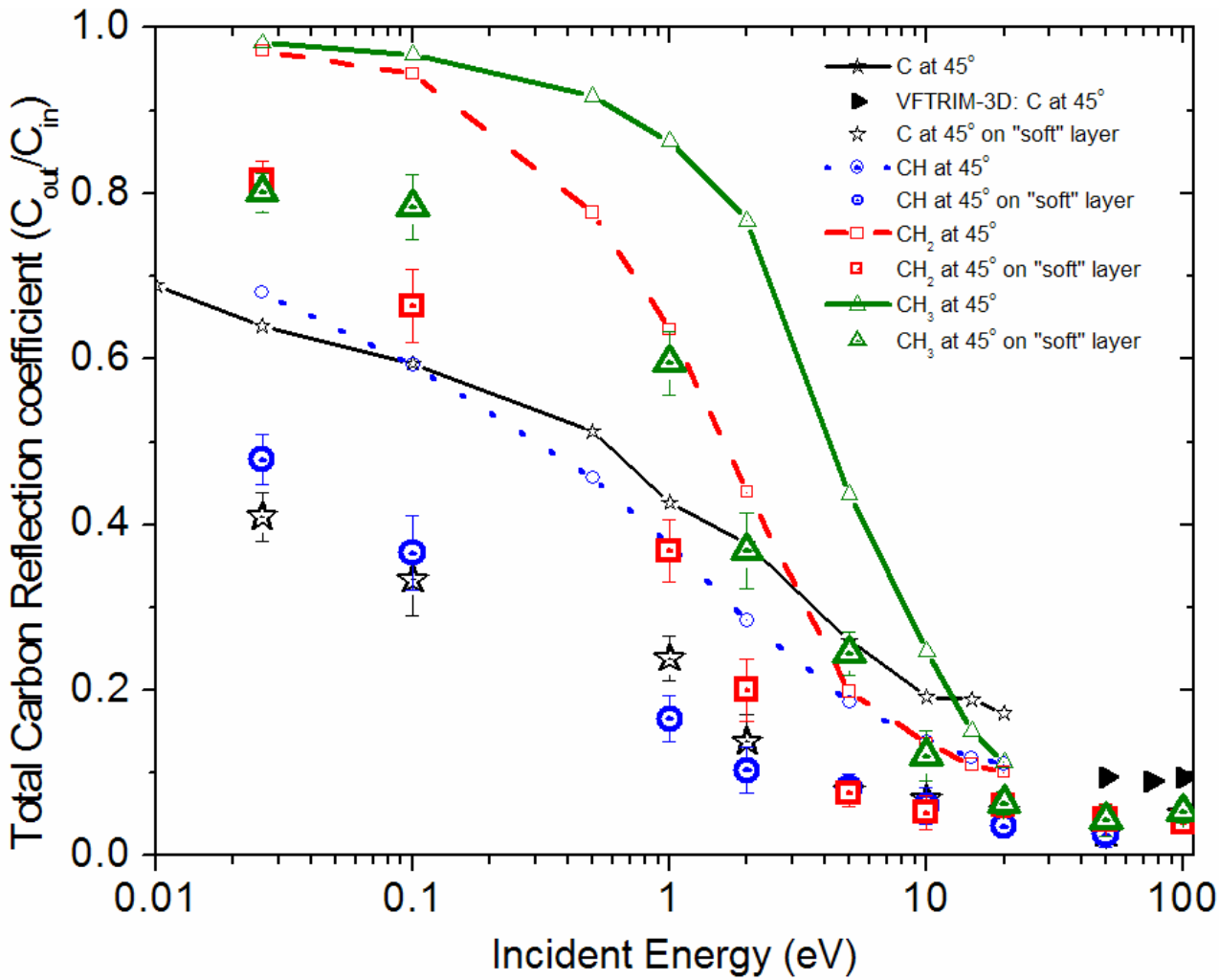
[1] D. Beeman, *J. Comp. Phys.*, 1976. **20** 130.
[2] H. J. C. Berendsen, *et al.*, *J. Chem. Phys.*, 1984. **81** 3684.

Systems examined with MD

- *Hydrocarbons*: Reflection of hydrocarbons bombarding graphitic and hydrocarbon surfaces to help explain unexpected T codeposition on the “louvers” of JET
- *Liquid lithium:* Reflection and sputtering of Li and H bombardment of pure lithium surface
- *Hydrogenated liquid lithium:* H and D reflection and sputtering from Li/LiH sample and search for expected saturation level of H-concentration

MD Modeling of Hydrocarbons: HC reflection

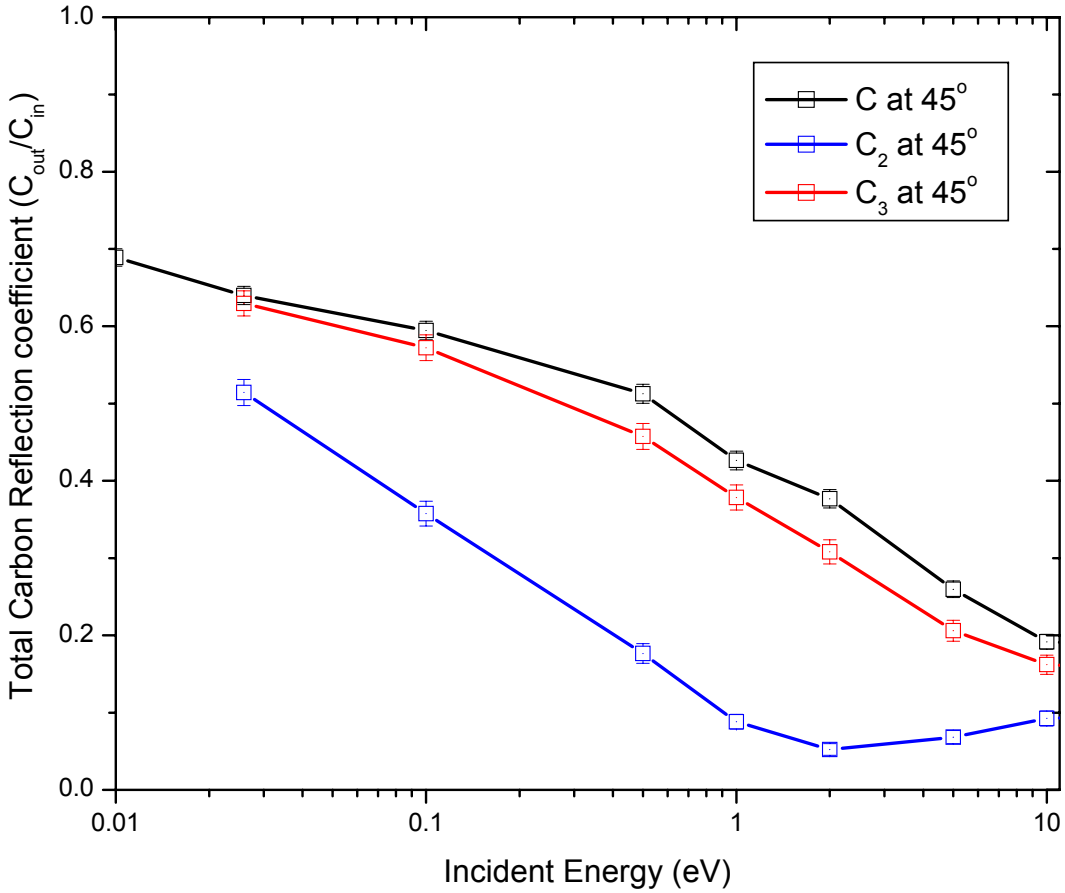
- First surface used was graphite bombarded by hydrogen until saturation (~ 0.4 H:C)
- Second surface - a “soft” H:C surface
  - Formed by deposition of thousands of hydrocarbons on a graphite surface
  - Emulates a redeposited carbon layer in a tokamak
    - Larger H:C ratio in this redeposited layer
    - Lower density
    - Carbon is less strongly bound
- Results show reflection is lower on the “soft” surface than the 0.4 H:C graphite at one energy (1 eV)



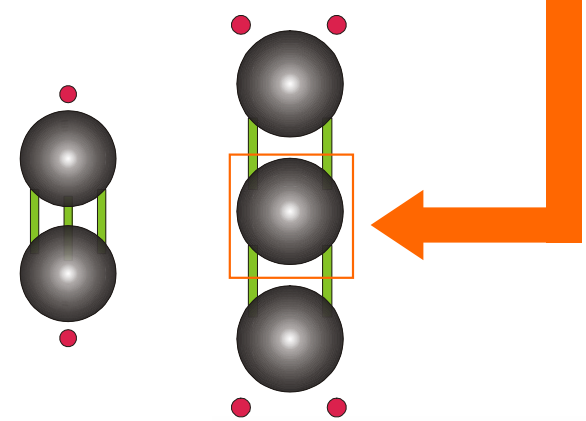
Reflection from “soft” vs. “hard” surfaces

- Why is reflection lower on the “soft” surface
  - The top layer of the “soft” surface is less dense
  - Incident atoms/molecules can penetrate more easily
  - Once inside the surface, the particles are more likely to be trapped there
  - On the “hard” surface, incident particles are more apt to have a hard collision on the very top surface and bounce off

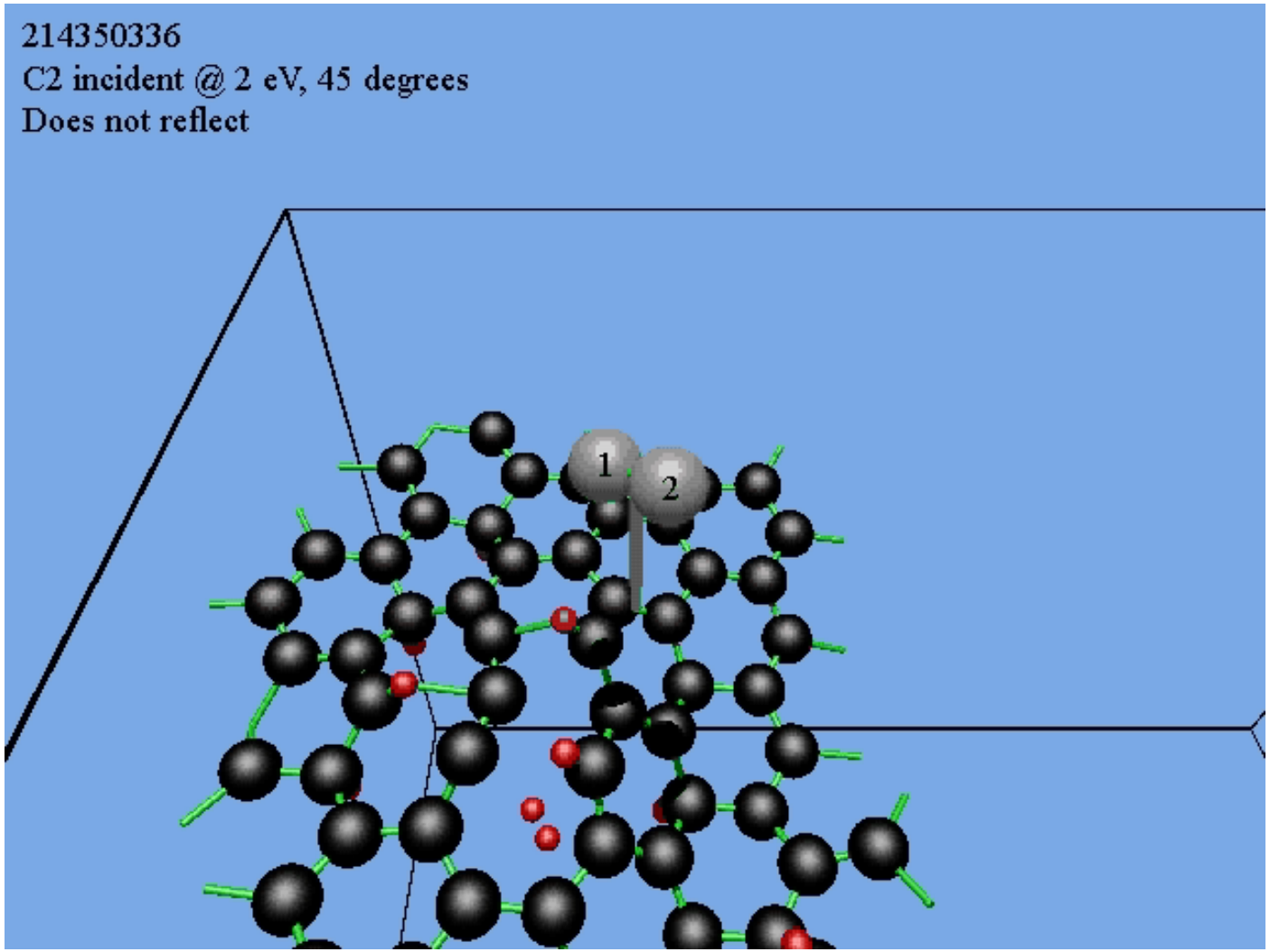
Reflection of carbon dimer and trimer molecules



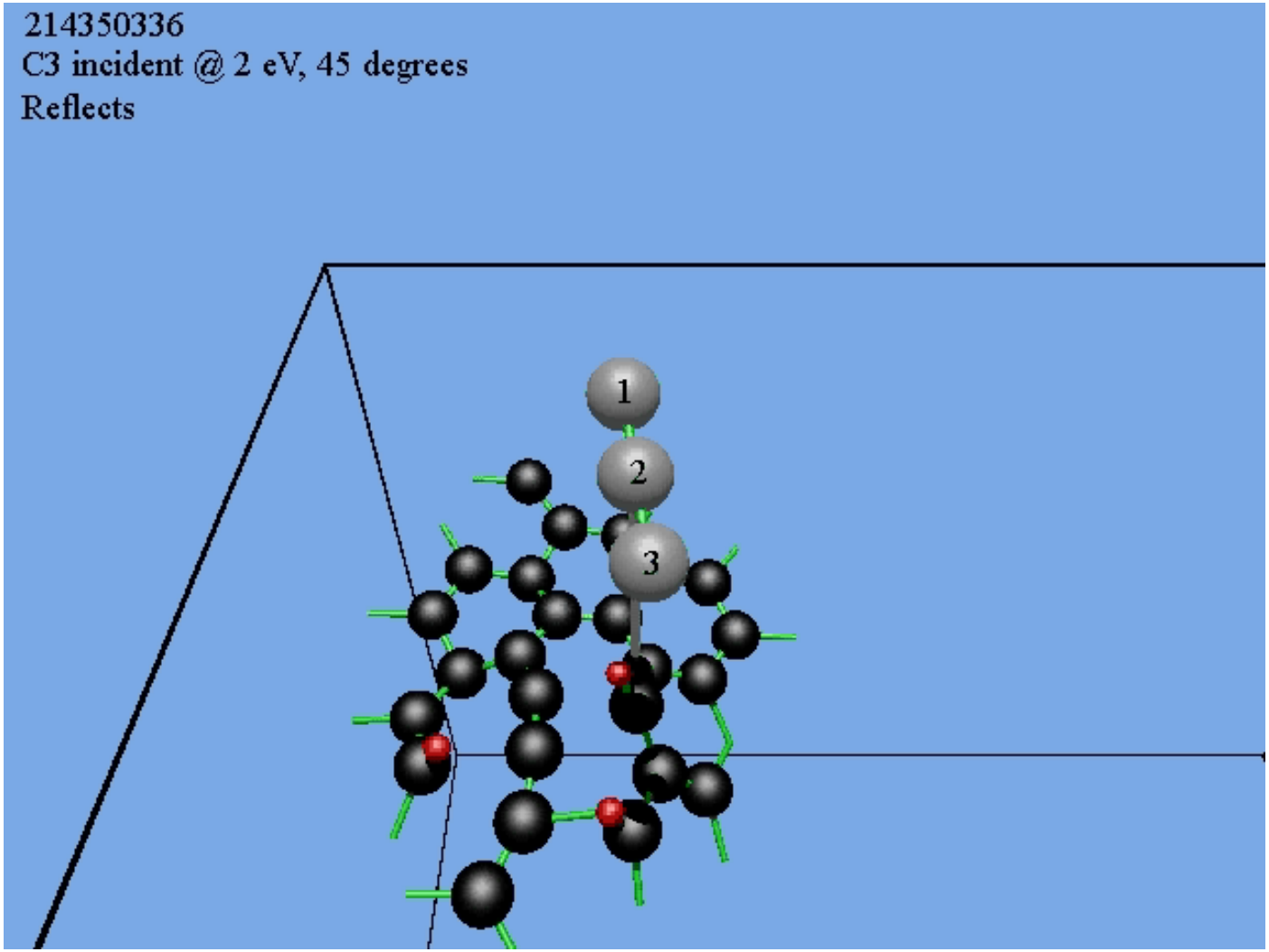
- Why does C_2 stick more readily than C_3?
  - One reason could be the fully bonded atom at the center of the trimer molecule
  - The end atom may attempt to stick, but when the molecule swings into position a repulsive force is created



Example MD simulation: C_2 sticking to H:C surface

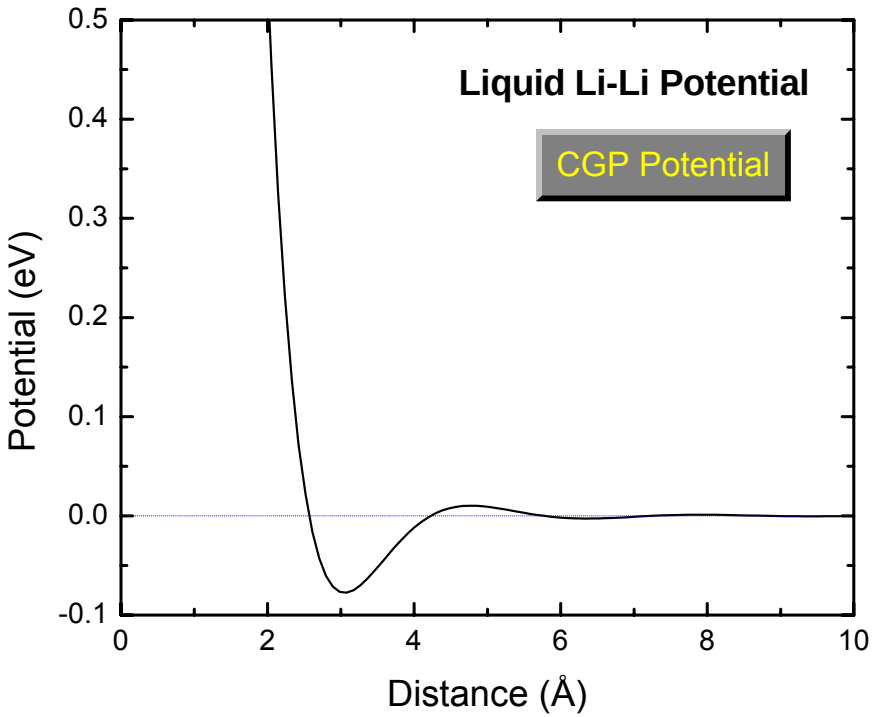


Example MD simulation: C_3 reflecting from H:C surface



Li-Li interaction potential used: CGP Potential

- Obtained from the Neutral Pseudoatom (NPA) method[1]
  - Involving structural and thermodynamic properties
  - Developed solely for liquid Lithium
  - Properties seen compare well to experimental results[1]



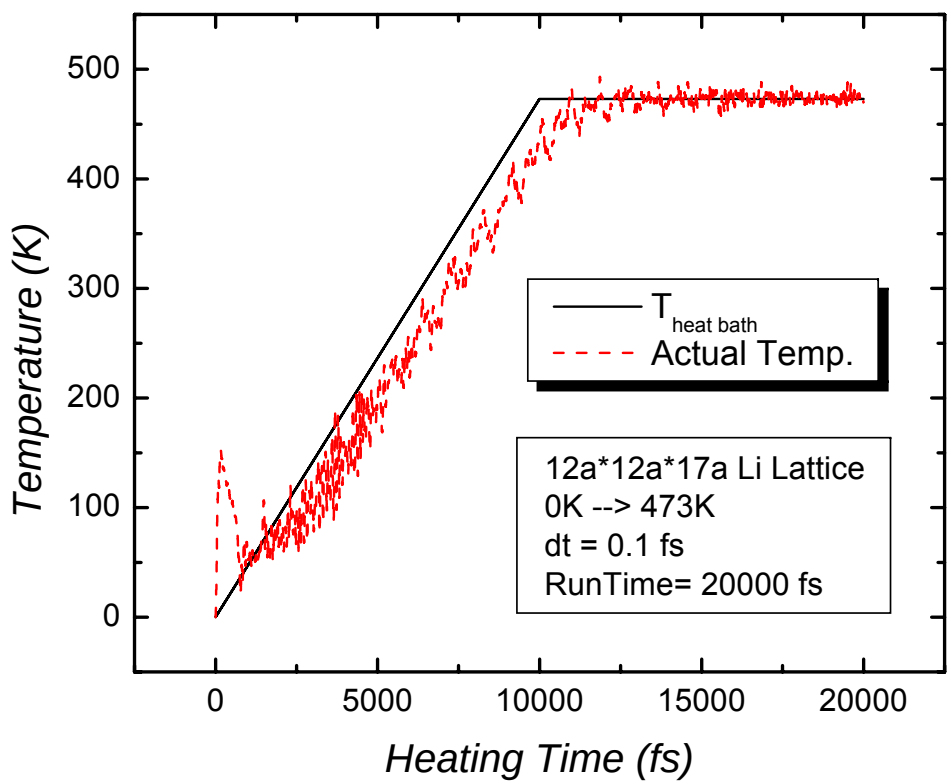
- Bond Energy: 0.082785 eV
- Interaction Cutoff: 5.75 Å @ 0.004623 eV, ~ 5.58% of the potential well
- Bond Min Distance: 2.60 Å
- Bond Max Distance: 4.125 Å
- Effective for high temperature ranging 470K ~ 843K
- The fluctuations around 5 Å show the ***long-range interaction***

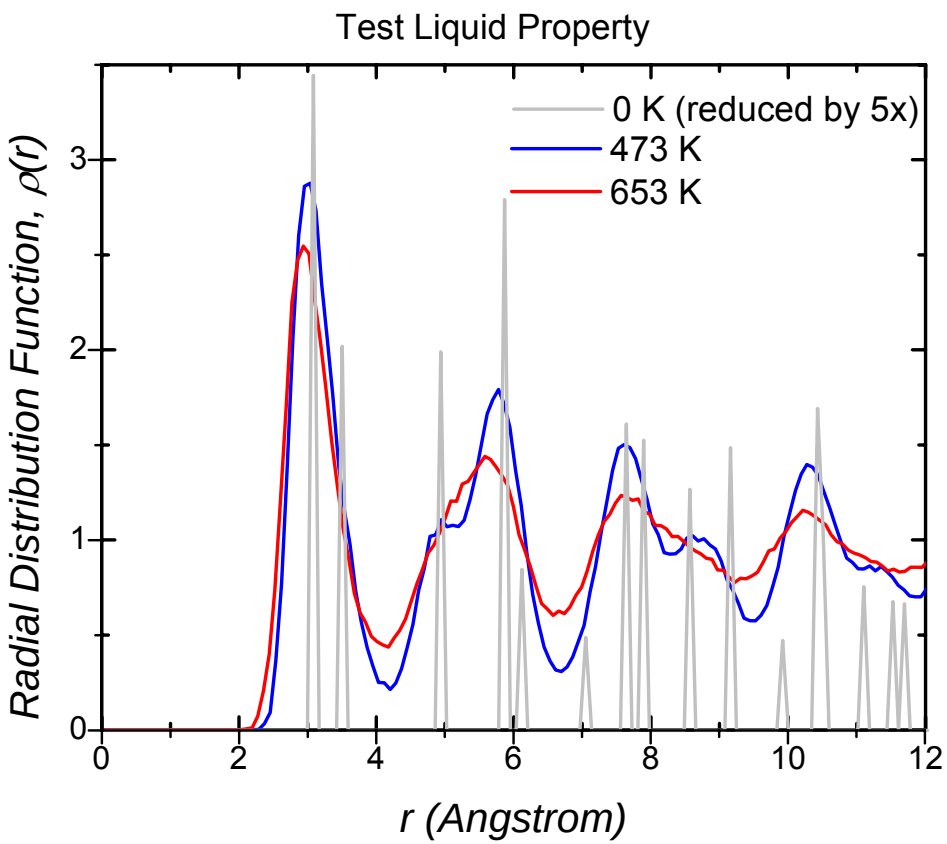
[1] Manel Canales, L. E. González, J. A. Padró, *Computer Simulation Study of Liquid Lithium at 470 and 843K, Physical Review E*, **50** (1994) 3656.

Liquid lithium MD simulations

- Creation of the Li lattice
  - Lattice size: 2727-5040 Li atoms
  - Dimensions: 42Å x 42Å x 39Å to 60Å (deep)
  - Periodical side boundary and fixed bottom
  - Initially a BCC structure
  - Slowly heated to required temperature

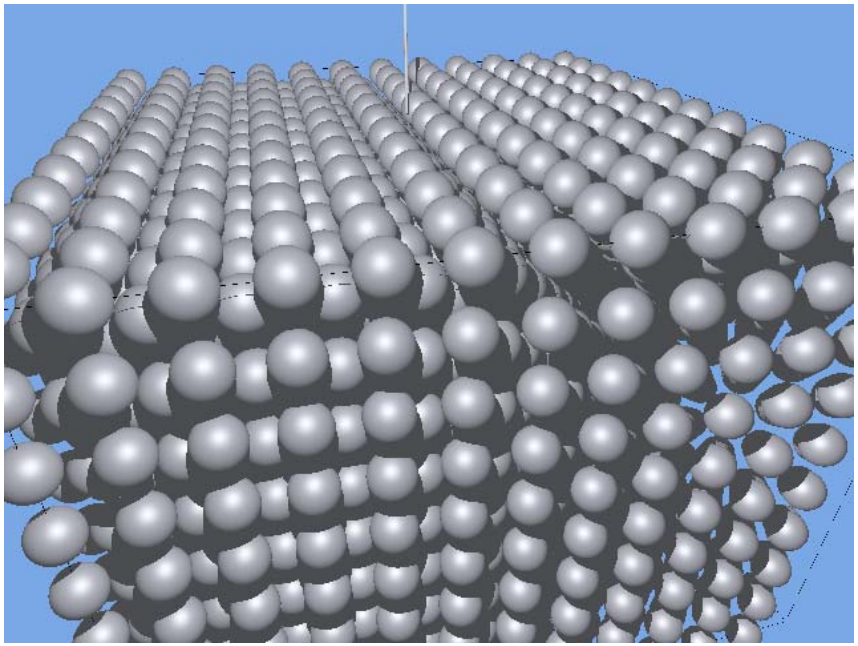
Creation of Liquid Li Lattice

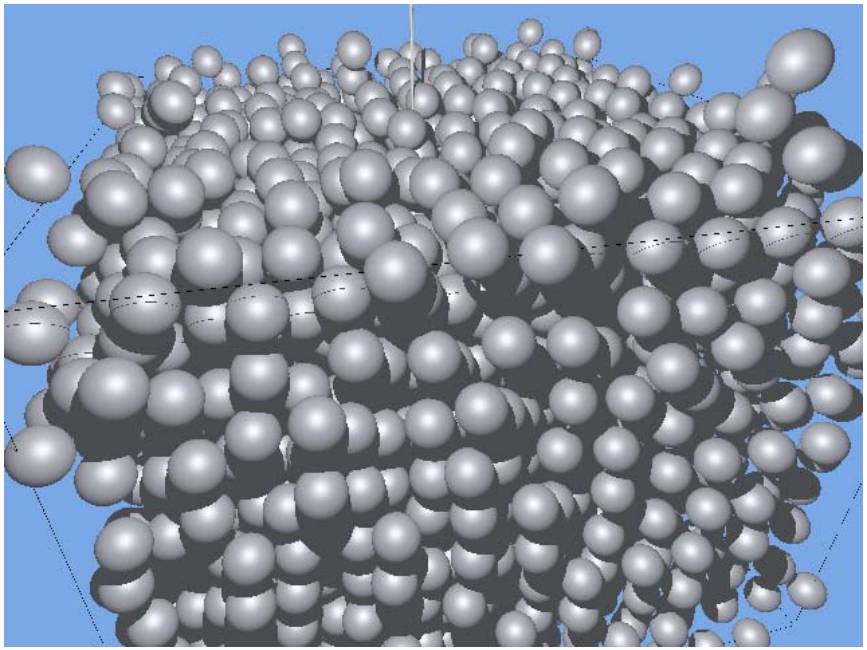




- Heat is added gradually over 10,000 fs
- The surface is allowed to equilibrate for another 10,000 fs
- The oscillating curves in the radial distribution function indicate the disordered, liquid state

Liquid lithium simulation setup





- Temperature control is achieved by using a simple velocity scaling technique at each time step[1-3] to maintain the desired temperature at the *edges* of the lattice.
- The resulting target surface is an amorphous liquid lithium surface

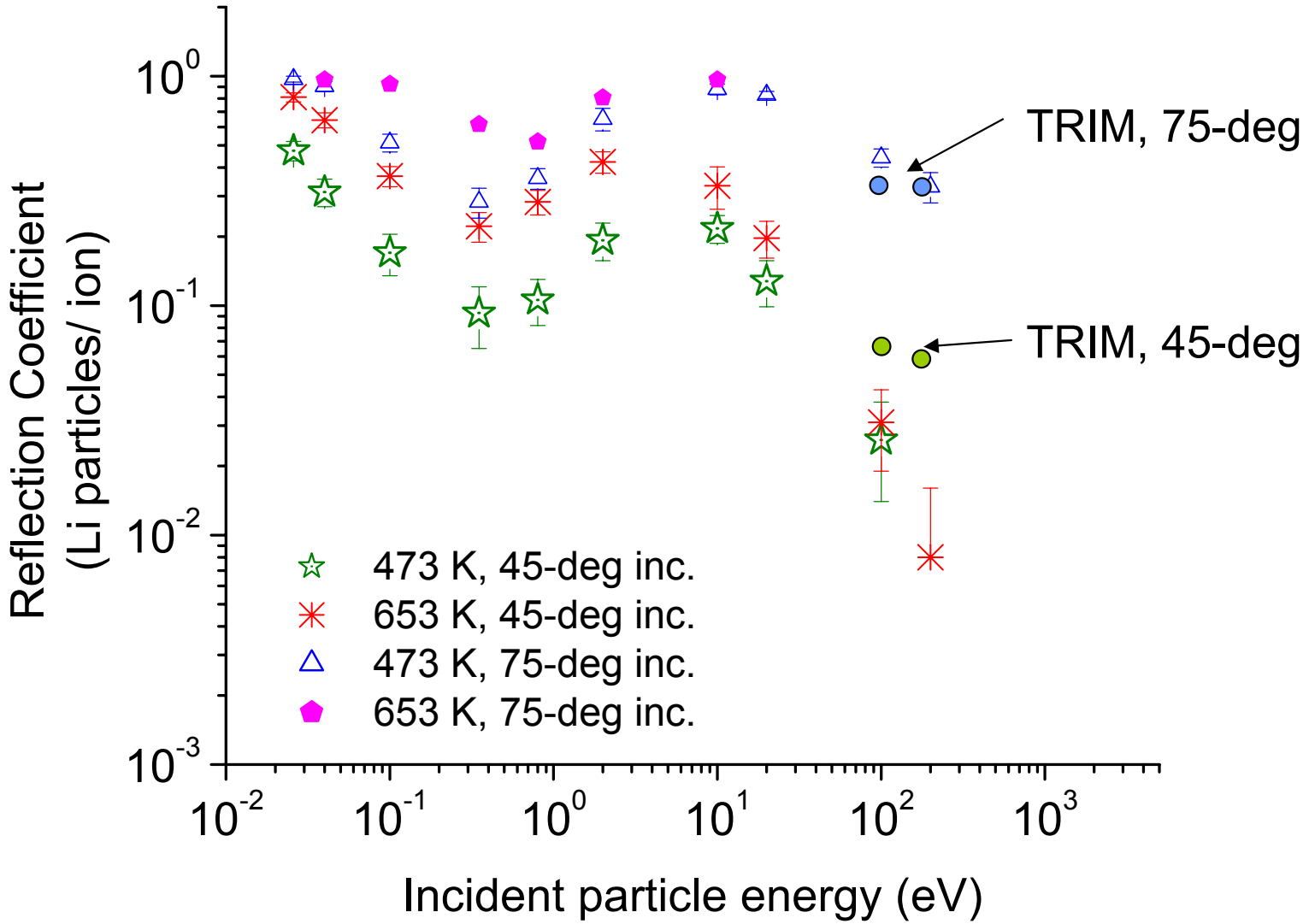
[1] L. V. Woodcock, Chem. Phys. Lett. **10**, 257 (1970).
[2] D. J. Evans, Mol. Phys. **37**, 1745 (1979).
[3] T. Schneider and E. Stoll, Phys. Rev. B **13**, 1216 (1976).

Surface potential of lithium lattice

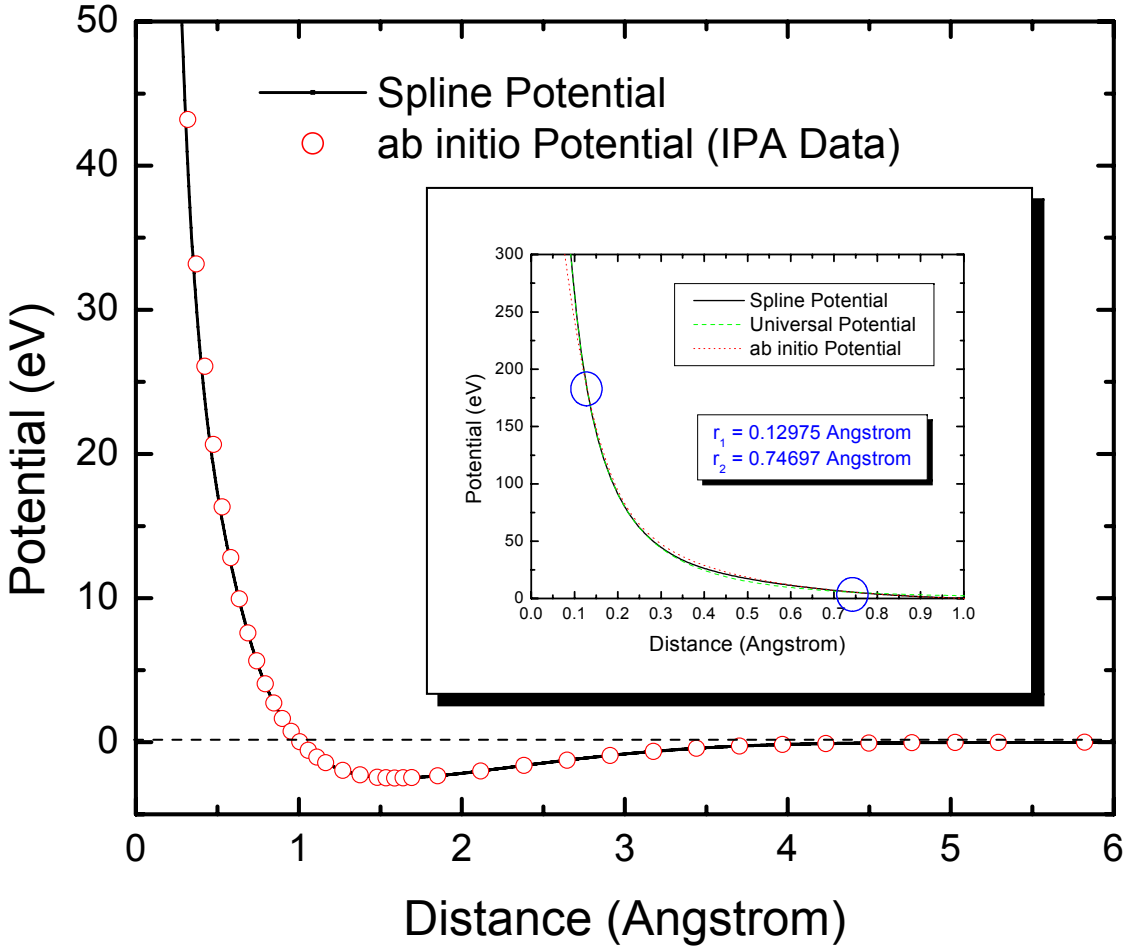
- Current
  - Surface Binding Energy (SBE) consistency
  - Weighted surface potential acts $\pm$ 2.5Å on the surface
  - CGP potential for all Li atoms
- Future
  - Cohesive Energy consistency
  - LBP[1] (many-body) potential for bulk Li atoms
  - LJ[2] + LBP potentials for Li in surface layers
  - To make a smoother liquid Li surface
  - Stratification along the depth

[1] Y. Li, E. Blaisten-Barojas and D. A. Papaconstantopoulos, *Phys. Rev.*, **B57** (1998-II) 519.
[2] A. Rahman, *Physical Review*, **136** (1964) 405.

Example results:
Lithium reflection at oblique incidence



H-Li Inter-Atomic Potential



- H-Li potential combines:
  - singlet *ab initio* potential [1,2]
  - universal potential at small distance to allow higher energy H-Li interactions
- Spline between 0.37 Å ~ 0.7565 Å to match the *ab initio* potential with the Universal
- Weight function [3] :

$$f_c(r) = \frac{1}{2} + \frac{1}{2}\cos\left[\frac{\pi(r - r_1)}{r_2 - r_1}\right]$$

[1] N. Geum, *et al.*, *Interaction potentials of LiH, NaH, KH, RbH and CsH, J. Chem. Phys.*, **115** (2001) 5984.
[2] R. Cote, *et al.*, *Enhanced Cooling of Hydrogen Atoms by Lithium Atoms, Physical Review Letters*, **84** (2000) 2806.
[3] R. Taylor, *et al.*, *Molecular Dynamics Simulations of Reactions between Molecules, Langmuir*, **11** (1995) 1220.

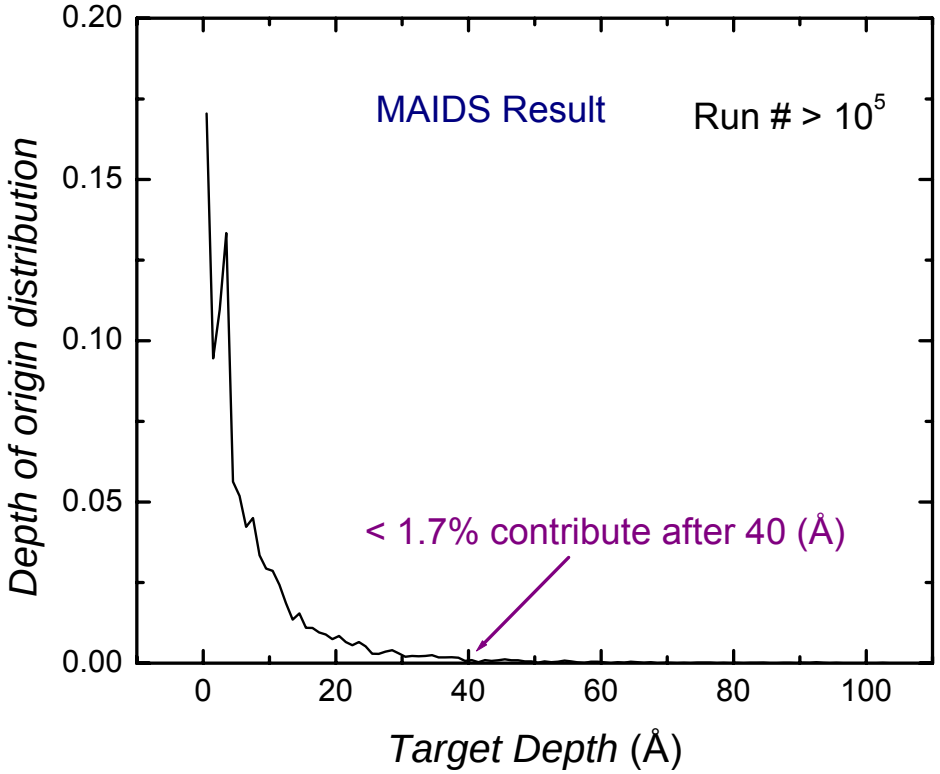
H-Li Inter-Atomic Potential (2)

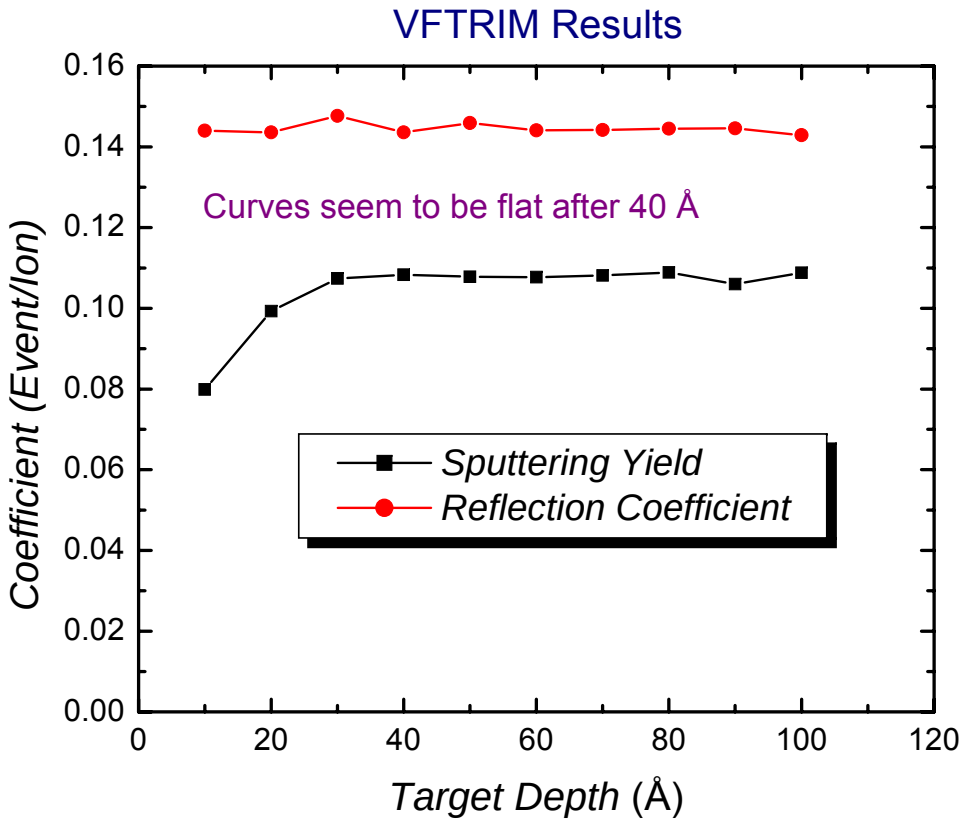
- Use singlet *ab initio* potential method [1,2]
  - Singlet: spin-paired electrons in the molecule
  - Semi-empirical character
  - Using a large scale configuration interaction method
  - Including dispersion forces and exchange interactions
- Bond parameters:
  - Interaction Cutoff: 5 Å
  - Bond Min Distance:1.05 Å
  - Bond Max Distance: 3.60 Å
  - Bond Energy: 2.526 eV

[1] N. Geum, *et al.*, *Interaction potentials of LiH, NaH, KH, RbH and CsH, J. Chem. Phys.*, **115** (2001) 5984.
[2] R. Cote, *et al.*, *Enhanced Cooling of Hydrogen Atoms by Lithium Atoms, Physical Review Letters*, **84** (2000) 2806.

Preparation of liquid Li lattice for 300 eV H bombardment at 45º incidence:

Determination of depth dimension



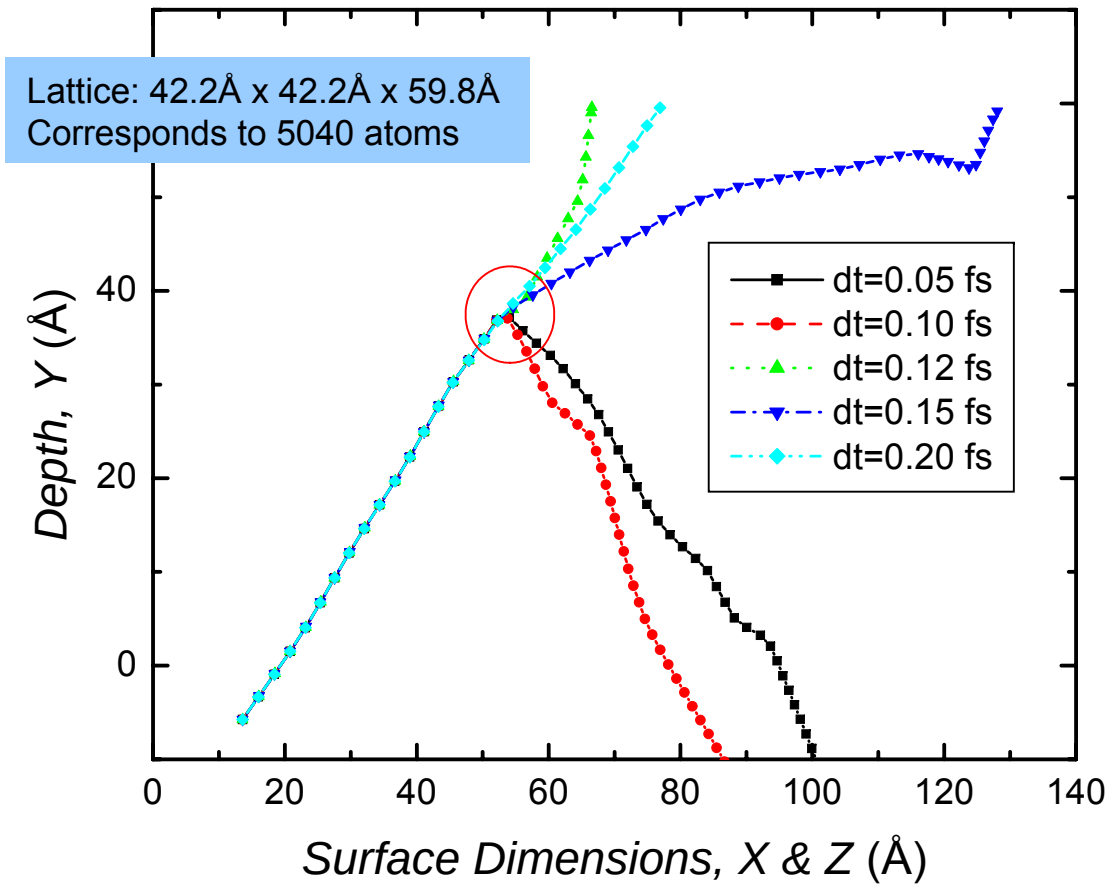


- MAIDS implies the deepest sputtered atom comes from 40Å
- VFTRIM shows no effect on sputtering yield by making the depth larger than ~40 Å
- Taking account for cascade effect, we choose 60Å as lattice depth

Preparation of liquid Li lattice for 300 eV H bombardment at 45º incidence:

Determination of time step

- Runs correspond to 473K lattice temperature
- dt > 0.1 fs gives an incorrect trajectory for a 300 eV H collision with a Li surface atom
- dt ⩽ 0.1 fs seems good enough to model the backscatter & sputtering
- As a tradeoff (accuracy vs. CPU time): dt = 0.1 fs



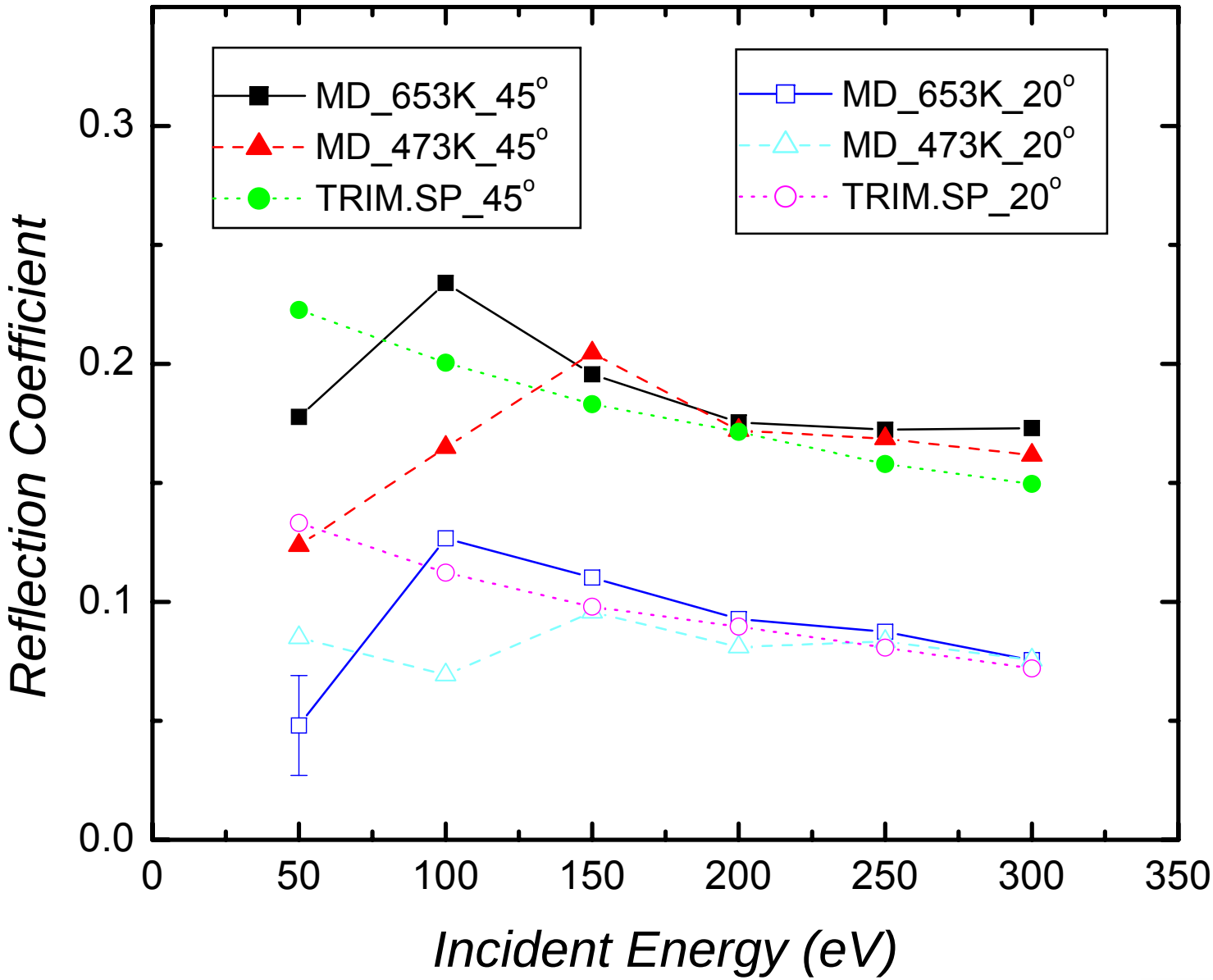
An aside on Monte Carlo atomic transport simulations: VFTRIM, TRIM.SP, TRIM

- While we primarily use VFTRIM[1] to model (E_{inc} > 100 eV) plasma-material interactions, TRIM.SP[2] and TRIM[2] are also widely used throughout the community, and results from them will be shown.
- TRIM.SP adds "soft collisions" to help account for multi-body effects not present in BCA; it is specifically designed and benchmarked for sputtering yields
- VFTRIM adds the option of simulating surface roughness using a fractal algorithm to help simulate a realistic surface
- Lower energy simulations are somewhat unreliable using MC methods such as these and variations in temperature are not possible, outside of varying the surface binding energy (sbe)

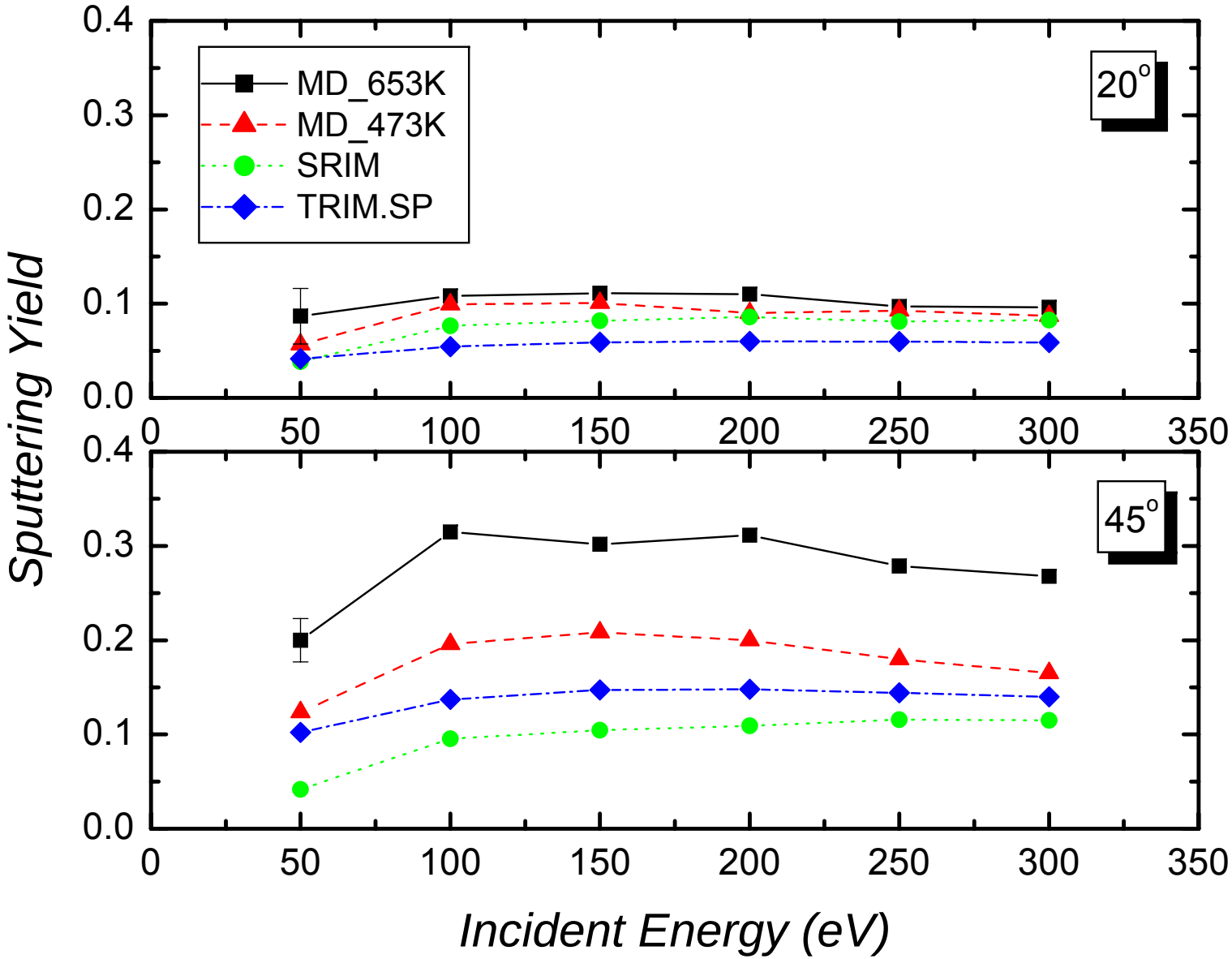
[1] Ruzic, D.N., *The Effects of Surface Roughness Characterized by Fractal Geometry.* Nuclear Instruments and Methods in Physics Research, 1990. **B47**: p. 118-125.

[2] Eckstein, W., *Computer Simulation of Ion-Solid Interactions*. Springer Series in Materials Science, ed. U. Gonser, et al. Vol. 10. 1991, Berlin: Springer-Verlag.

H → pure Li results: Reflection coefficient



H → pure Li results: Sputtering yields



Conclusions for H on pure Li

- Sputtering yield increases with surface temperature and incident angle, as observed in Li+ on liquid Li experiments
- Reflection coefficient is slightly affected by surface temperature only at low incident energies and agrees with VF-TRIM & TRIM.SP at higher energies

…how about buildup of H in a real Li lattice???

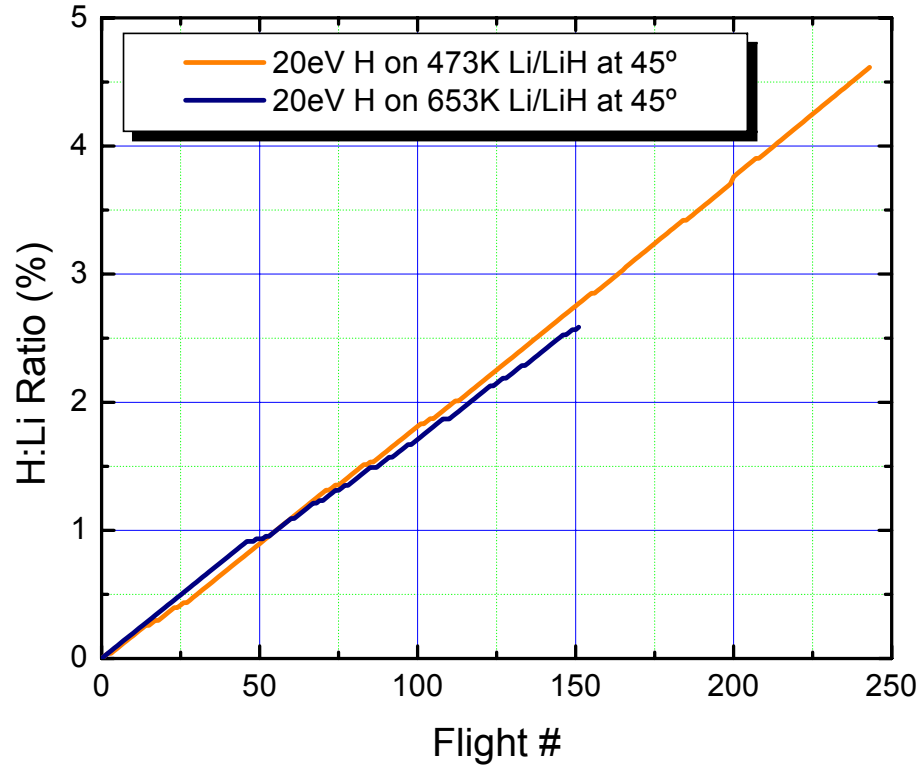
Preparation of the Hydrogenated Lithium Surface

- Initially, a pure lithium lattice at a given temperature using MolDyn is relaxed
- Code modified to include I/O of the lattice atoms – This allows the development of a more realistic hydrogenated surface with 1000's of individual H/D impacts
- The generated H:Li surface is used as the input in all subsequent simulations

Hydrogenated Liquid Li Surface

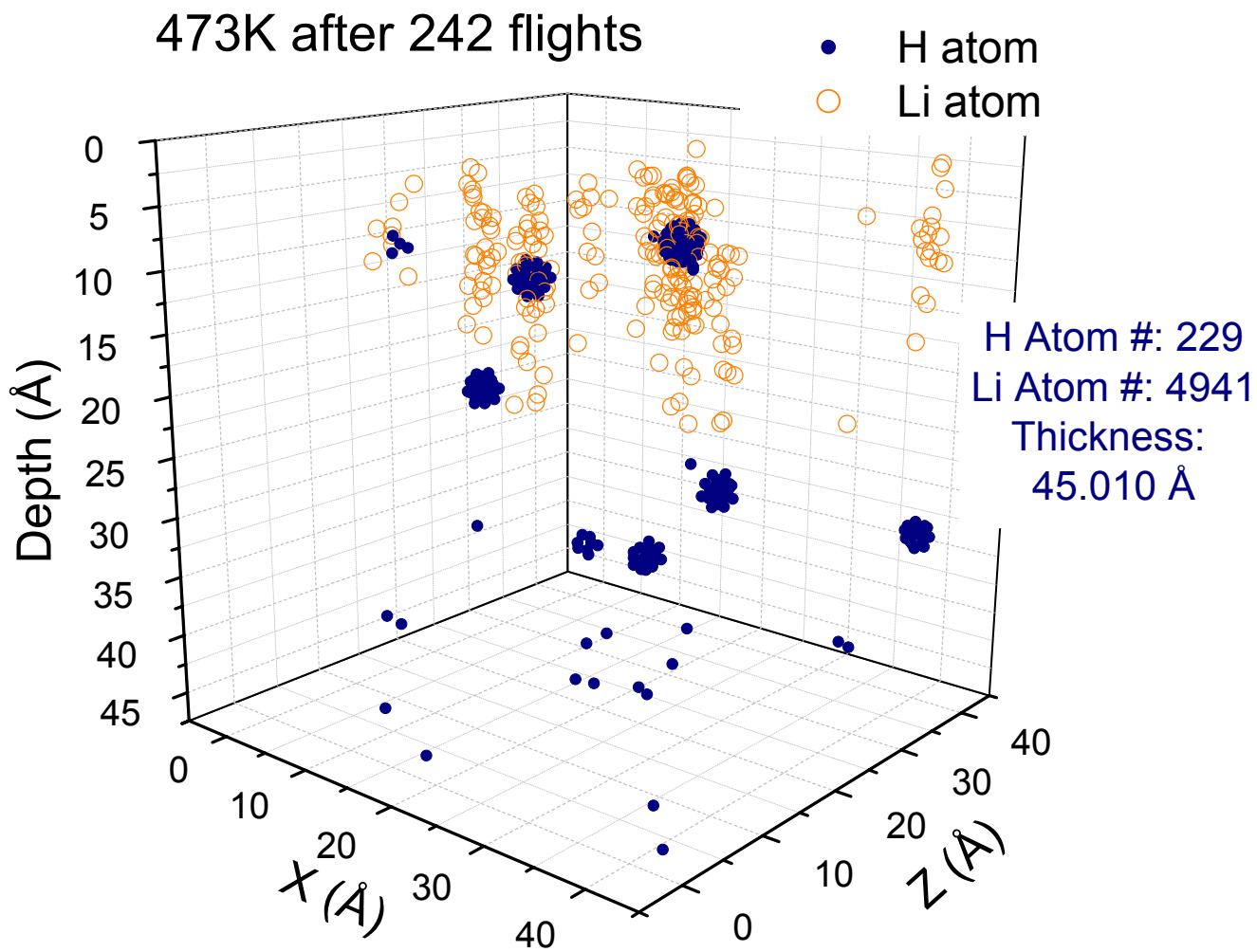
- Continuously incident H on the hydrogenated liquid Li surfaces reusing lattice
- Results
  - Sputtering and Reflection
  - Hydride concentration H:Li
  - Implantation/characterization of H

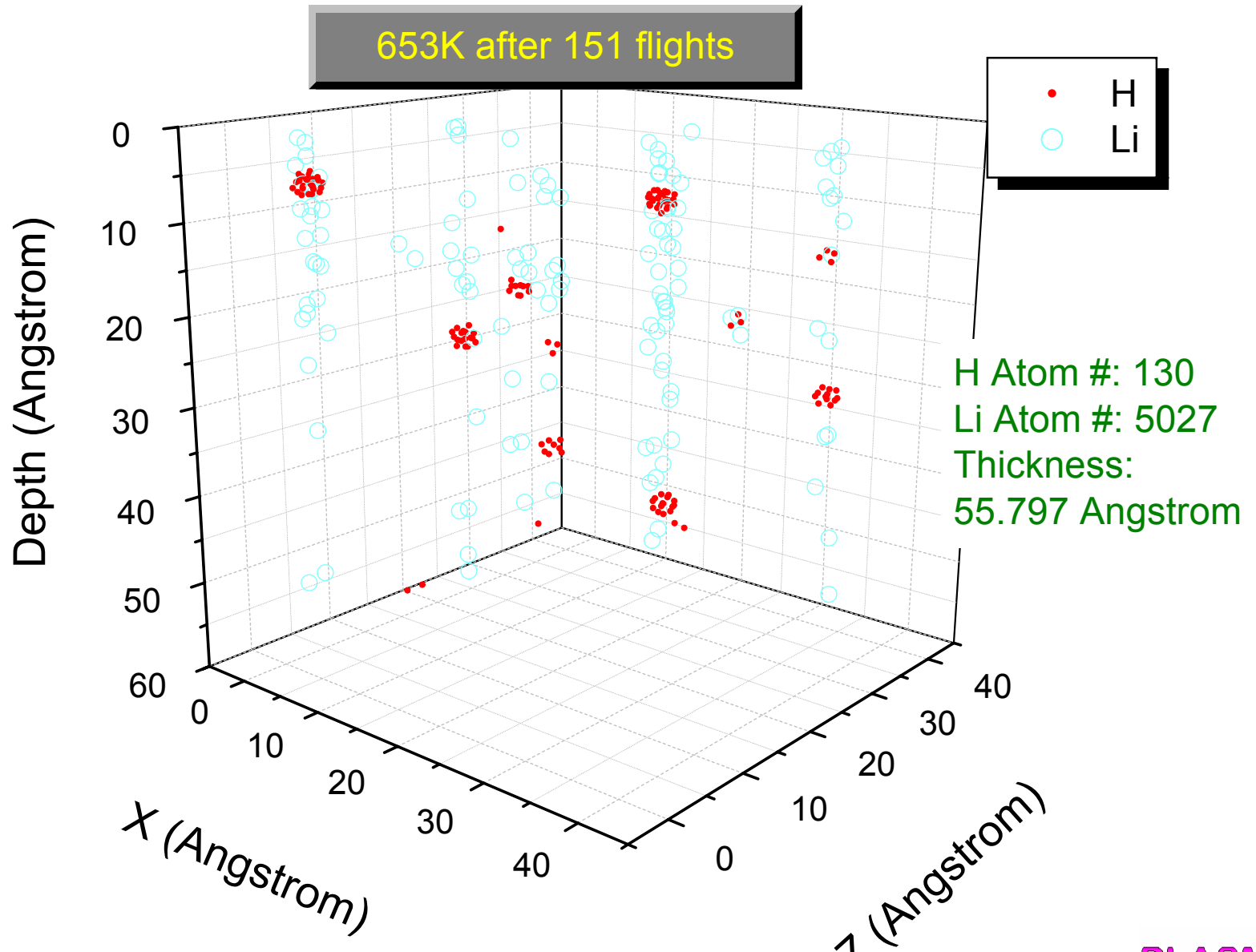
- Saturation is not obtained due to the insufficient time scale (~2 *ps*)
- The actual concentration can not stay at such a high level at thermodynamic equilibrium according to the phase diagram



H atoms seem to form clusters within Li/LiH lattice within 2 ps timeframe

- H atoms preferably diffuse towards the other H atoms within around layers in similar depth, H-H bond energy > Li-H bond energy.
- H atoms are more likely to bond together with H atoms
- Thickness decreases due to removal of sputtered Li atoms




653K after 151 flights
H
Li
Depth (Angstrom)
0
10
20
30
40
50
60
X (Angstrom)
0
10
20
30
40
Z (Angstrom)
0
10
20
30
40
H Atom #: 130
Li Atom #: 5027
Thickness:
55.797 Angstrom

Conclusions for H bombardment of Li/LiH

- The implanted H atoms diffuse towards the other H atoms around the similar depth in the hydrogenated Li surfaces
- H atoms are more likely to bond together with the around H atoms, resulting in the forming of H clusters, especially near the surface
- Hydrogenated Li surfaces with continuous H incident on Li/LiH surfaces might have a higher H:Li concentration than the phase diagram due to the short computation time scale, 2ps
- The thickness of the hydrogenated liquid Li surface decreases due to the continuously removal of the sputtered Li atoms

Initial results of 300 eV H → Li / LiH at 45°

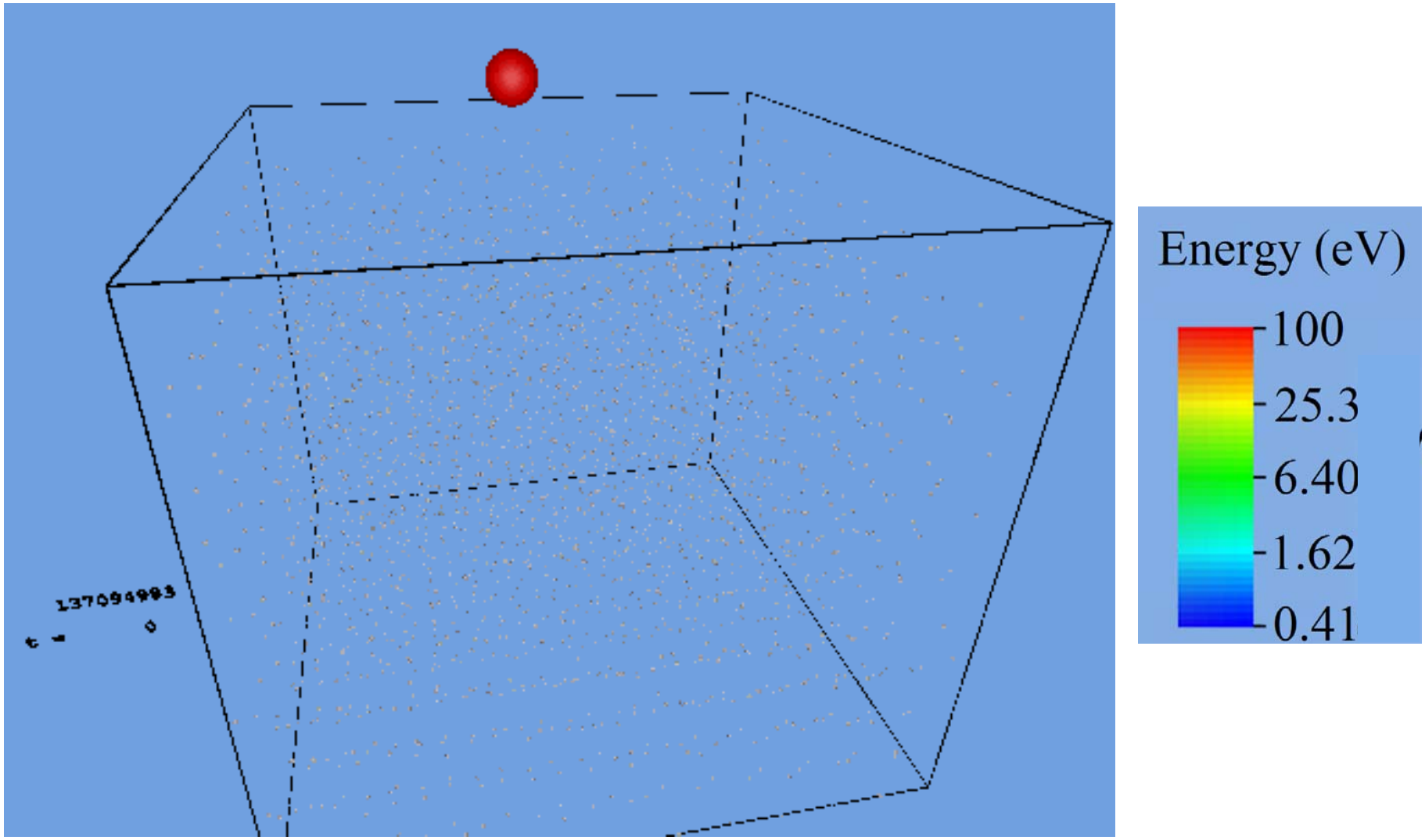
Lattice Temp	Sputter Yield	Mean Sputtered Energy	Mean Sputtered Theta	Reflection Coeff.	Mean Reflected Energy	Mean Reflected Theta
473K	0.1695	7.9 eV	49.11°	0.0636	137. eV	47.02°
653K	0.0861	7.0 eV	52.69 °	0.1391	64. eV	48.53°

TRIM	0.061	7.7 eV	46. °	0.189	110.0 eV	45. °

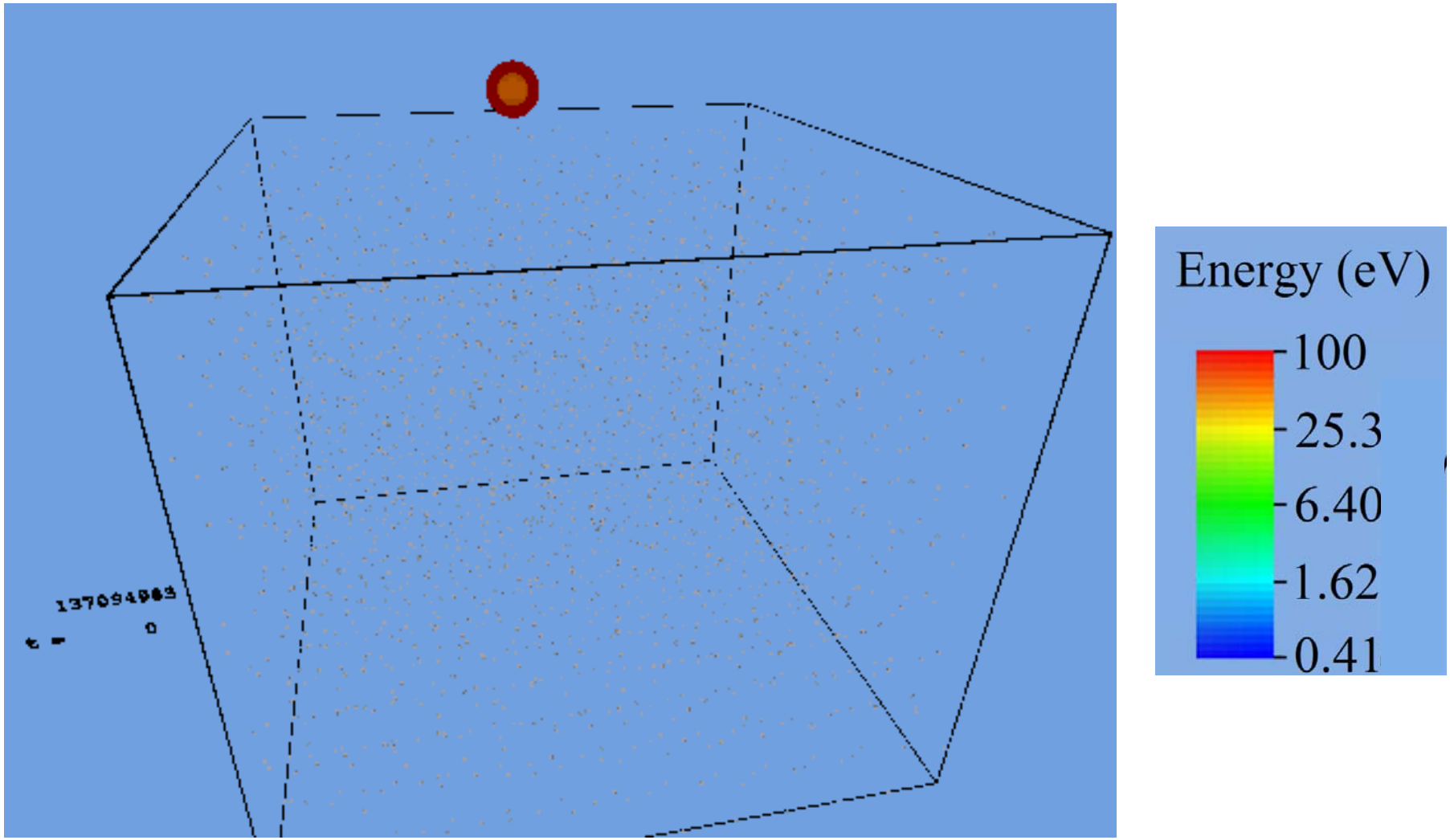
ILLINOIS

PLASMA MATERIAL INTERACTION GROUP
University of Illinois at Urbana-Champaign

100 eV Li → liquid Li at 473K at 45º



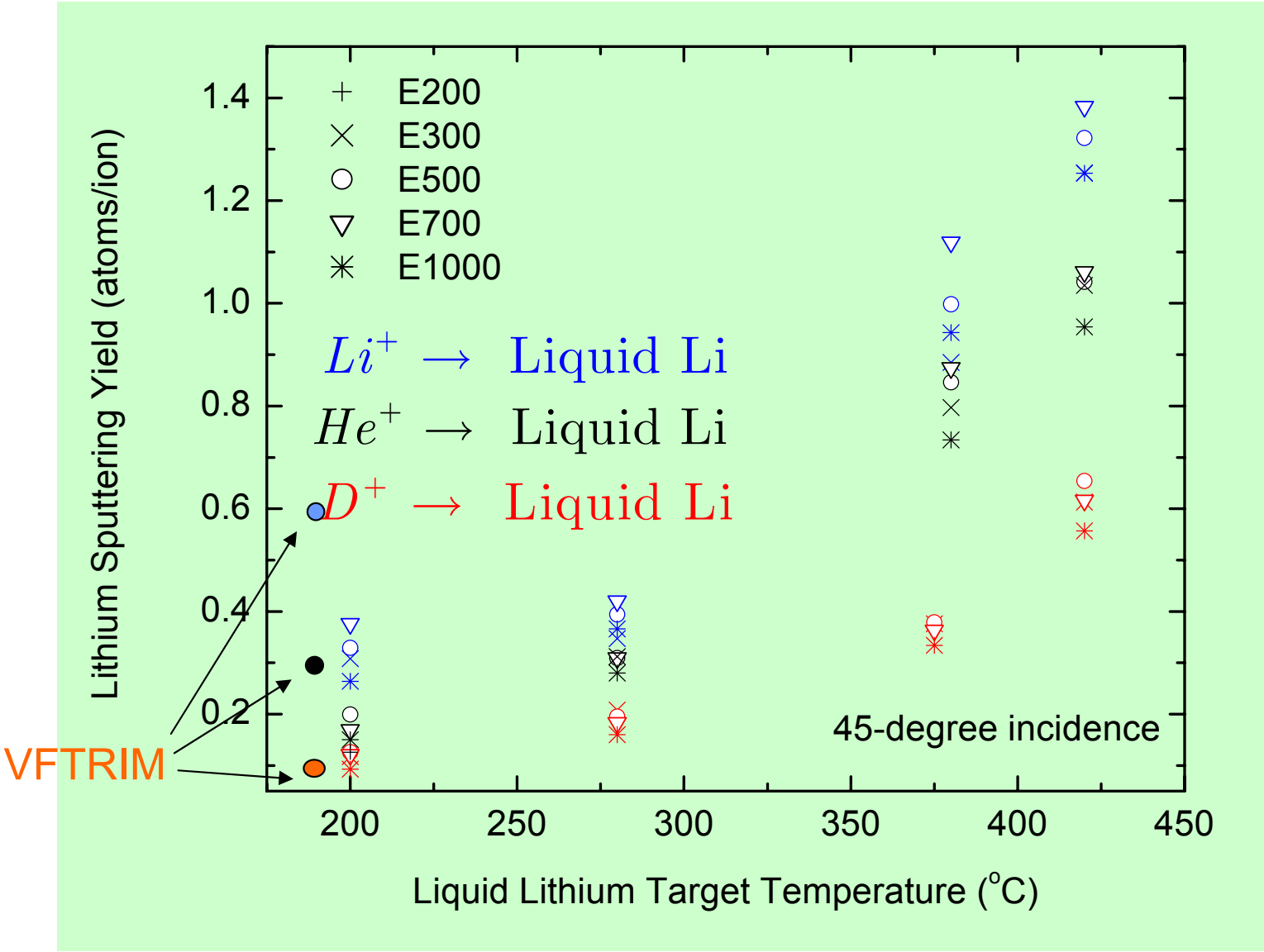
100eV Li → liquid Li at 653K at 45º



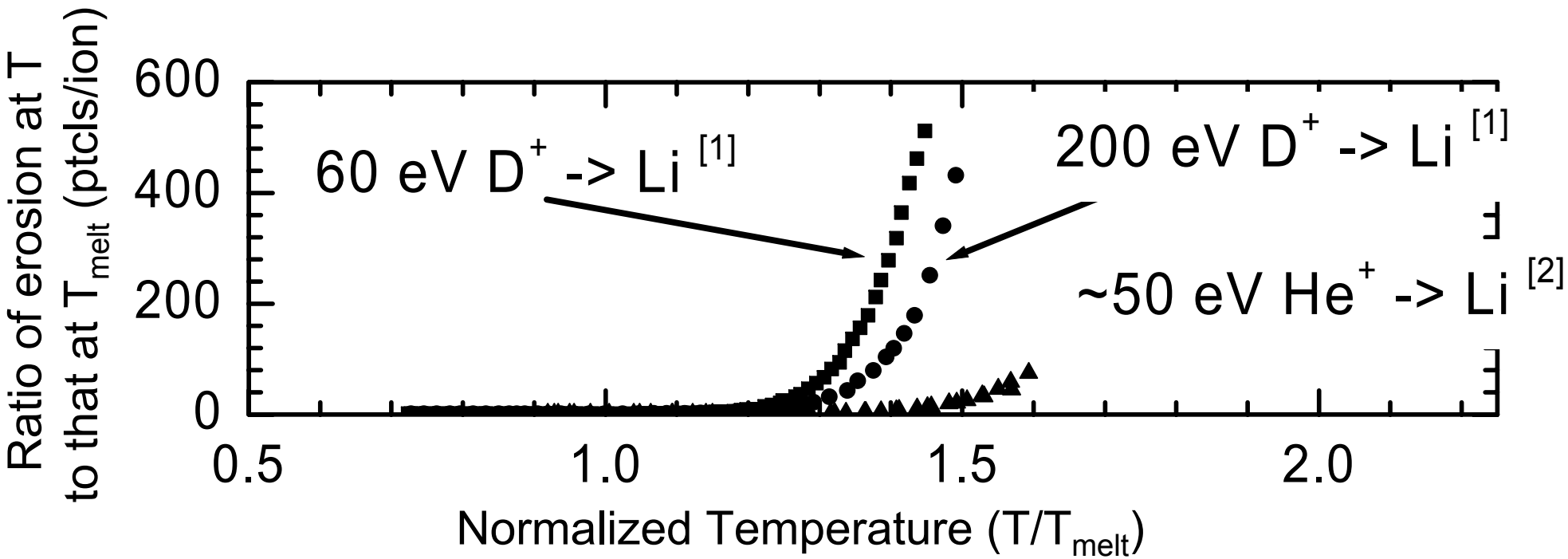
Temperature-enhanced sputtering

- Increases in surface temperature has been shown to increase the sputtering yield of materials in several labs
  - While our focus is on lithium, it has also been noted for materials
    - Liquid Sn-Li, Sn, and Ga
    - Solid Be and graphite
- Pure MD model shows temperature effects too, (though perhaps not the right direction)

Example data: IIAX temperature-dependent Li sputtering yields for various incident particle energies



Example data (2): Further experimental evidence from PISCES-B



[1] R.P. Doerner et al., J. Nucl. Mater. **313-316** (2003) 383-387.
[2] M.J. Baldwin et al., Fus. Eng. Des. **61-62** (2002) 231-236.

Conjectures on possible mechanisms responsible for erosion enhancement

- Bubble formation of implanted He or D could precipitate into nano-size bubbles reaching the surface and emitting a non-linear amount of material
  - To date no experimental data with liquid Li has shown signs of bubble formation (from surf. analysis of QCO)
- Localized rise in temperature (in the form of thermal or elastic spikes) could lead to a larger Li yield due to its low vapor pressure (a rise of 200 °C could easily do this)
- Other models for liquid metals?... fluid dynamics model of sputtering?
  - **M.M. Jakas, E.M. Bringa, R.E. Johnson, Phys. Rev. B 65 (2002) 165425-1**

Mechanisms believed to explain enhancement

- Near-surface energy deposition to “weakly-bonded”, mobile lithium atoms lead to non-linear erosion even for low-incident particle energies (true for materials with low sublimation heat such as: alkali metals or the alkaline earths, others: Ga, In, Sn, Sn-Li)… This occurs *even* for solid-phase materials becoming more conspicuous in the liquid state
- In addition, a decrease in surface binding energy under non-equilibrium conditions of ion-bombardment and a rise in the thermodynamic state of the liquid-metal by means of an increase in surface temperature
- Surface stratification (characteristic of liquid-metals) could in fact play a role in the enhancement of erosion

What can MD tell us about any possible <u>mechanisms</u> behind temperature-enhanced sputtering?

That is, in addition to using MD output to directly estimate the response (sputtering, reflection, …), what can we learn from our MD simulations?

- *Qualitative:* Examination of movies using position or energy dependent coloring showed that higher temperatures lead to a larger amount of exchange between surface and bulk atoms and an unexpected tendency for the incident atom to deposit an increased amount of energy in the plane parallel to the surface
- *Quantitative:* Analyzing the kinematic information output from MolDyn, one can find several trends with increasing temperature, each of which can lead to enhanced sputtering

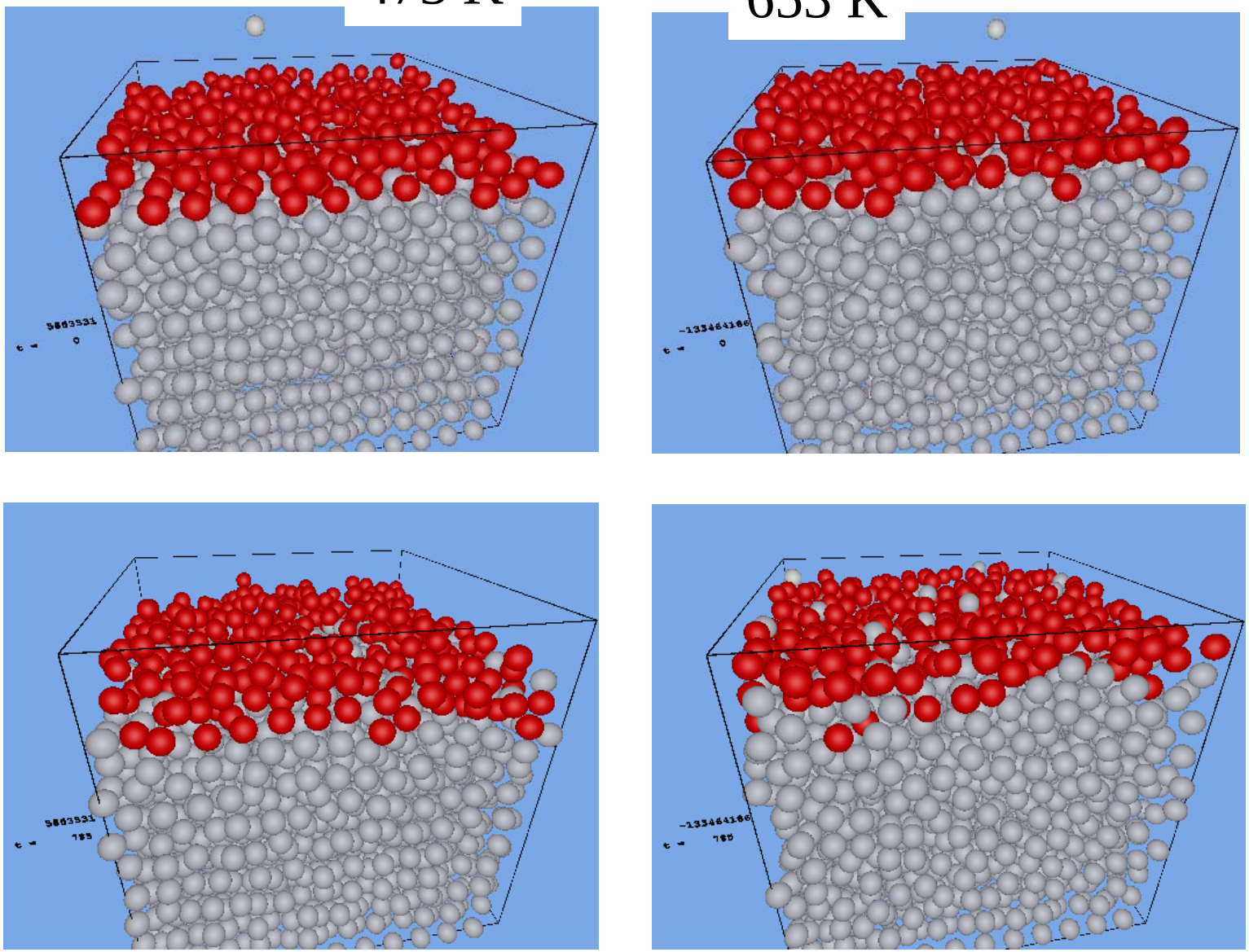
…MD did show some temperature-enhanced mixing of bulk atoms with those on the surface…

473 K

653 K

Initial

After 50 ps



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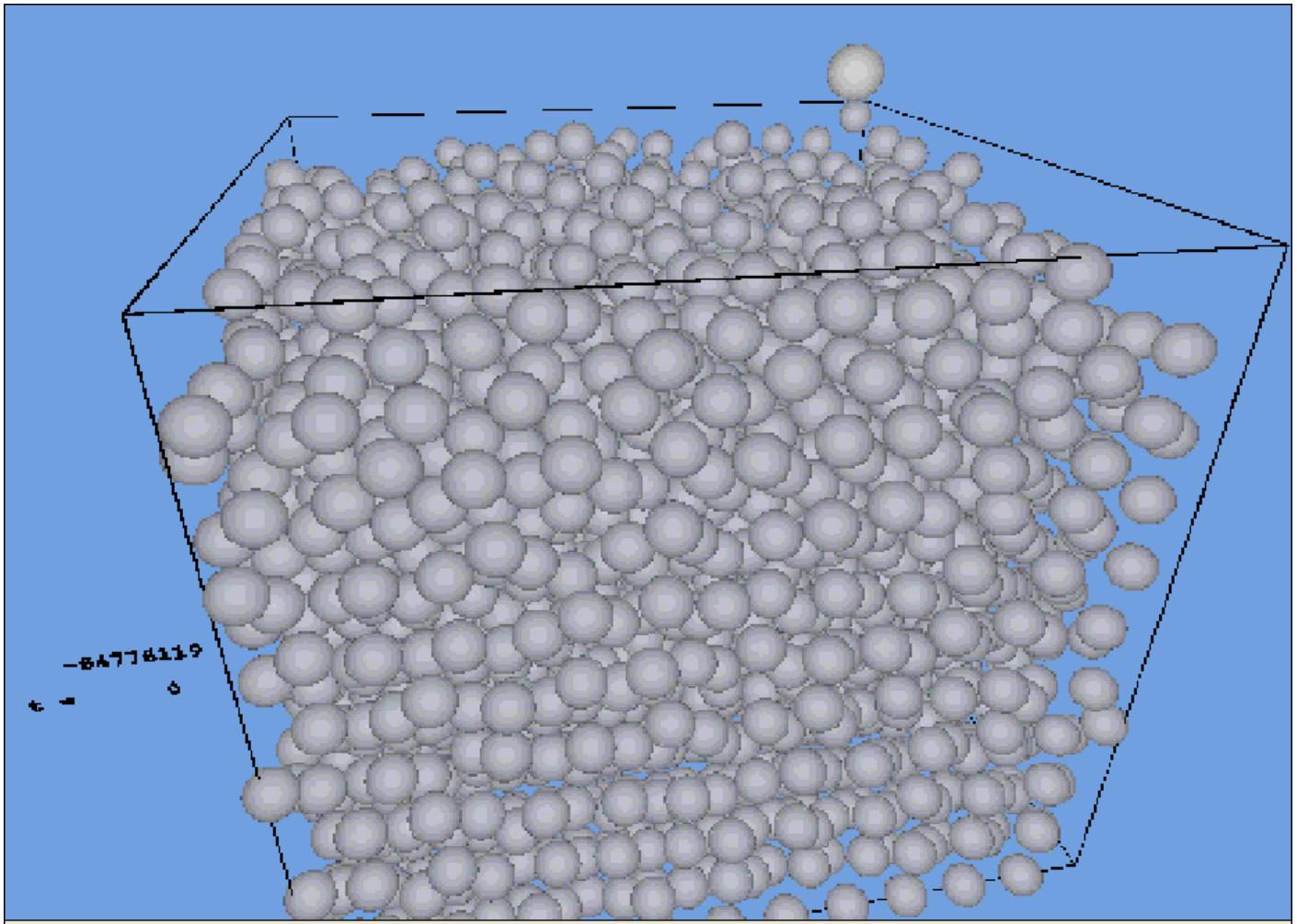
University of Illinois at Urbana-Champaign

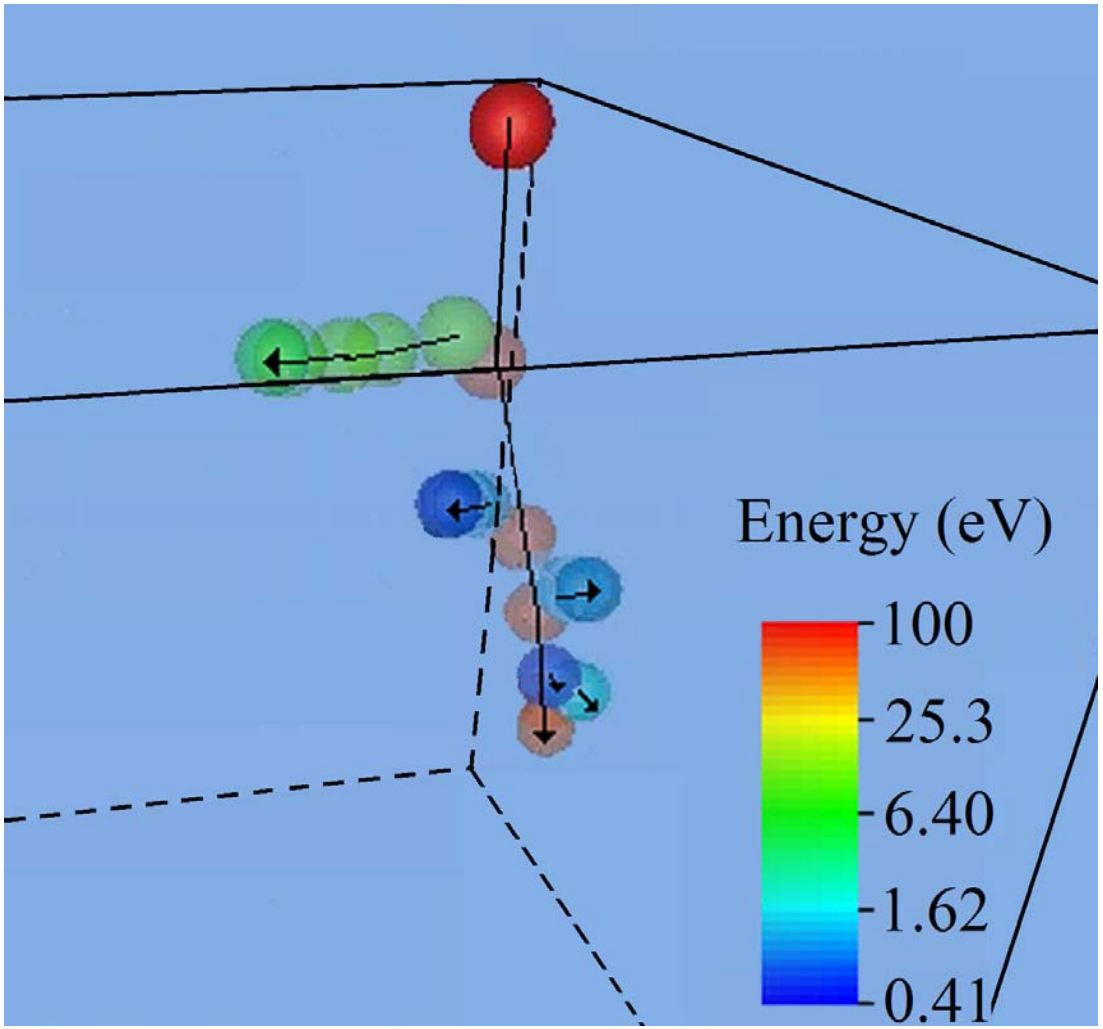
Example movie of the development of the collision cascade along the surface plane



Li_100.0eV_20deg_473k_-64776119_new LATERAL energy dep.avi

Interesting energy deposition behavior

- A composite image of six frames taken from a movie showing a lithium atom incident on a liquid lithium surface.
- The incident atom is shown in red (the highest energy in the system), and various primary knock-on atoms with lower kinetic energy are generated during the transit of the incident atom.
- A trend of increasing lateral energy deposition with increasing temperature was noted

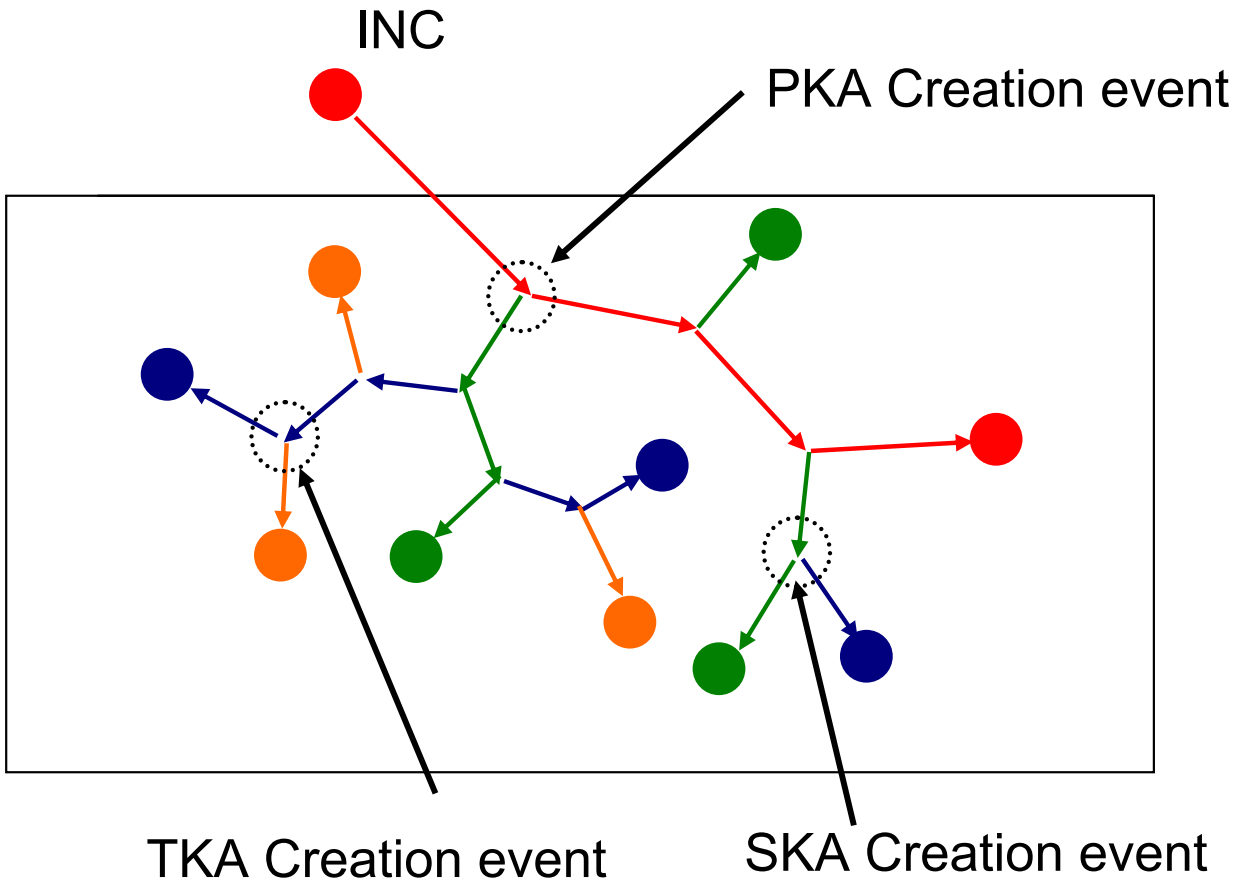


MD also gives us direct kinematic information (#'s) for comparison to each other over varying temperatures and to other simulation methods (e.g., VFTRIM)

- In addition to estimated physical parameters (sputtering and reflection coefficients), we have found that some of the fundamental *kinematic relations change* with changing lattice temperatures and differ greatly than that for BCA models commonly used

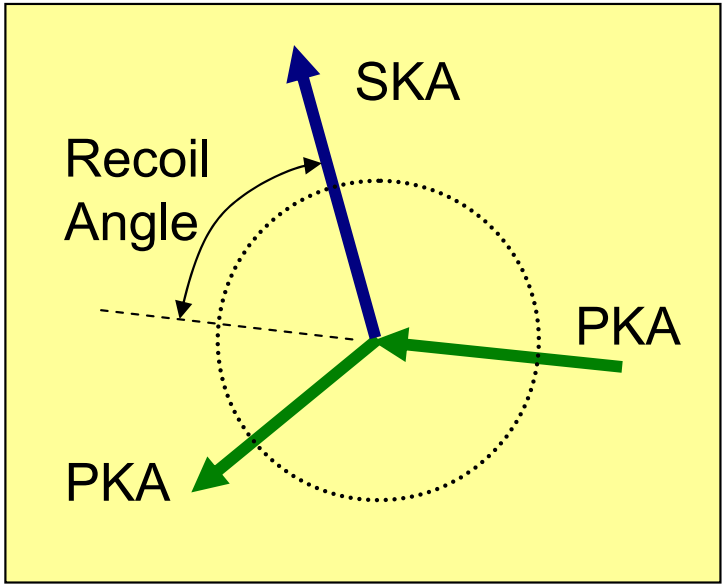
What information are we talking about…?

Data extracted from MolDyn simulations



Parent-daughter knock-on relations:

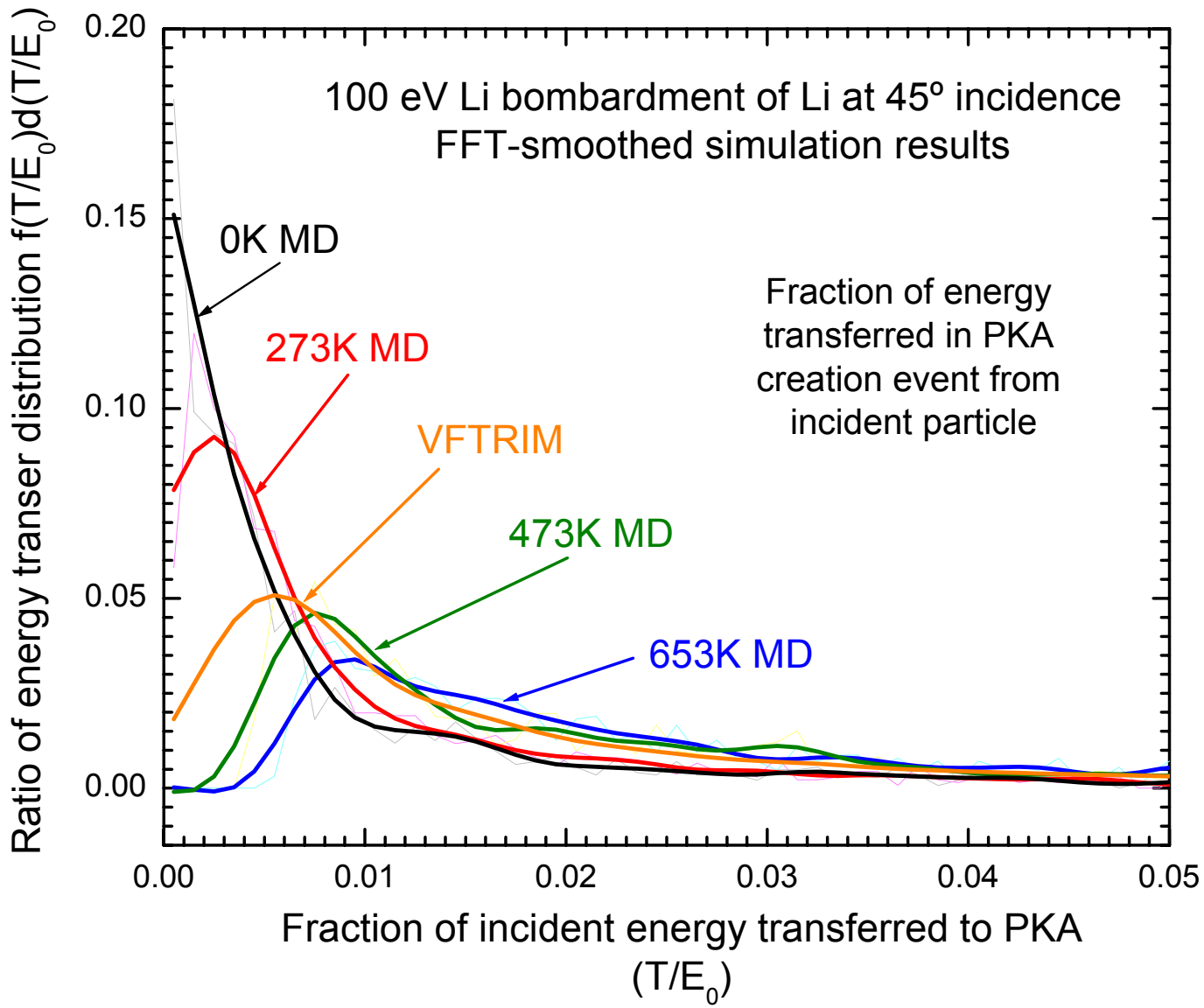
- Fraction of incident energy transferred
- Recoil angle



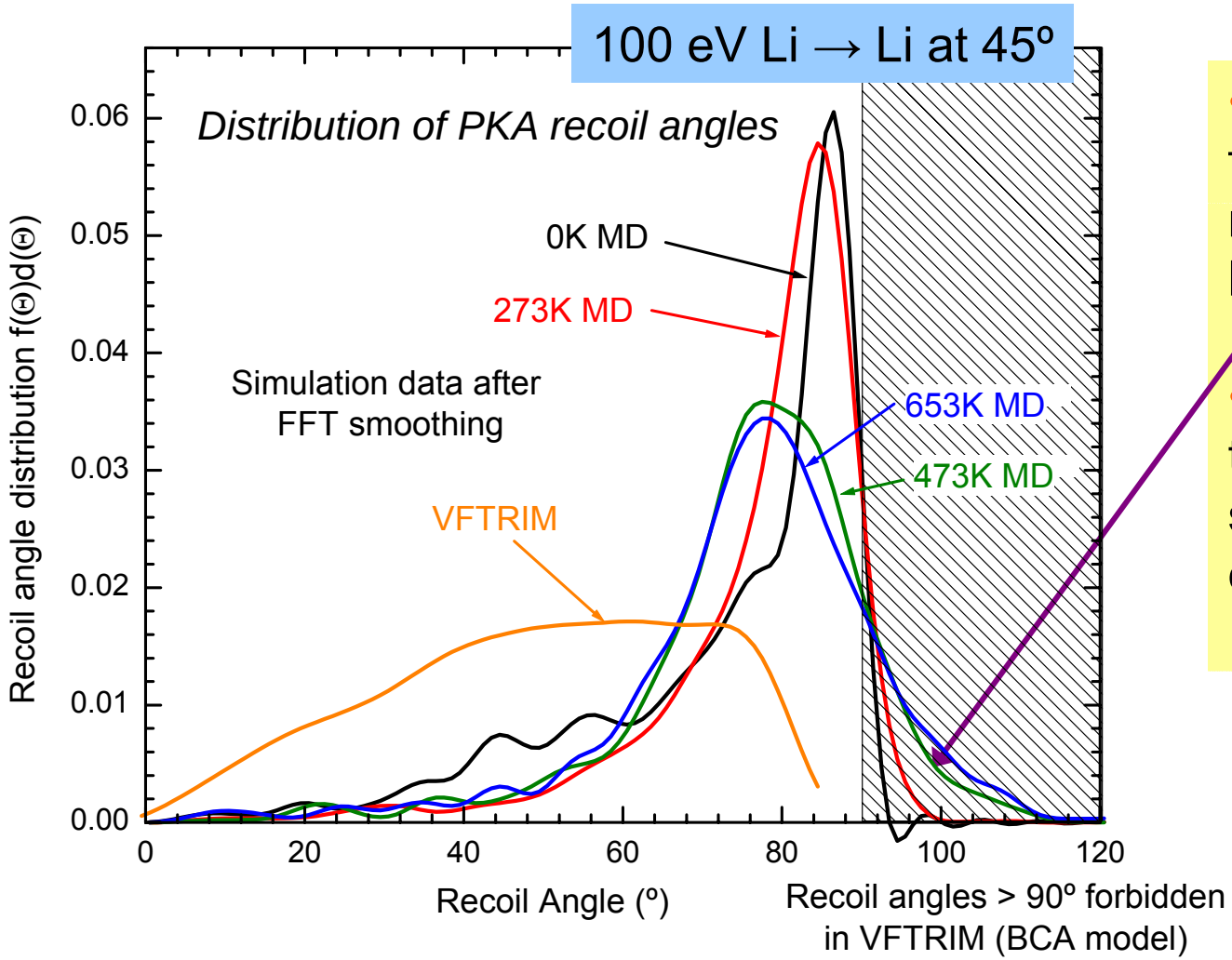
Primary, secondary, and tertiary knock-on creation events are tracked during MD simulations

Changes in energy transferred to PKA's due to temperature changes

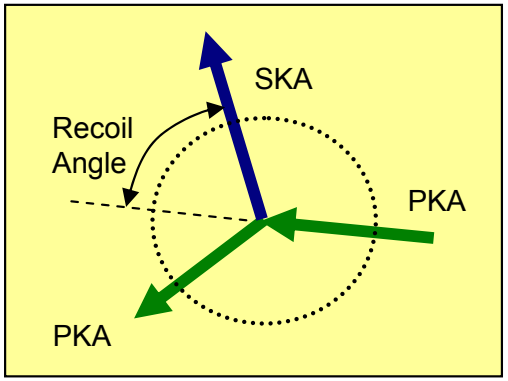
- VFTRIM distribution lies within those of MD
- Increasing lattice temperature increased the mean and peak fraction of energy transferred to PKA
- This results in higher energy density levels in the near-surface region



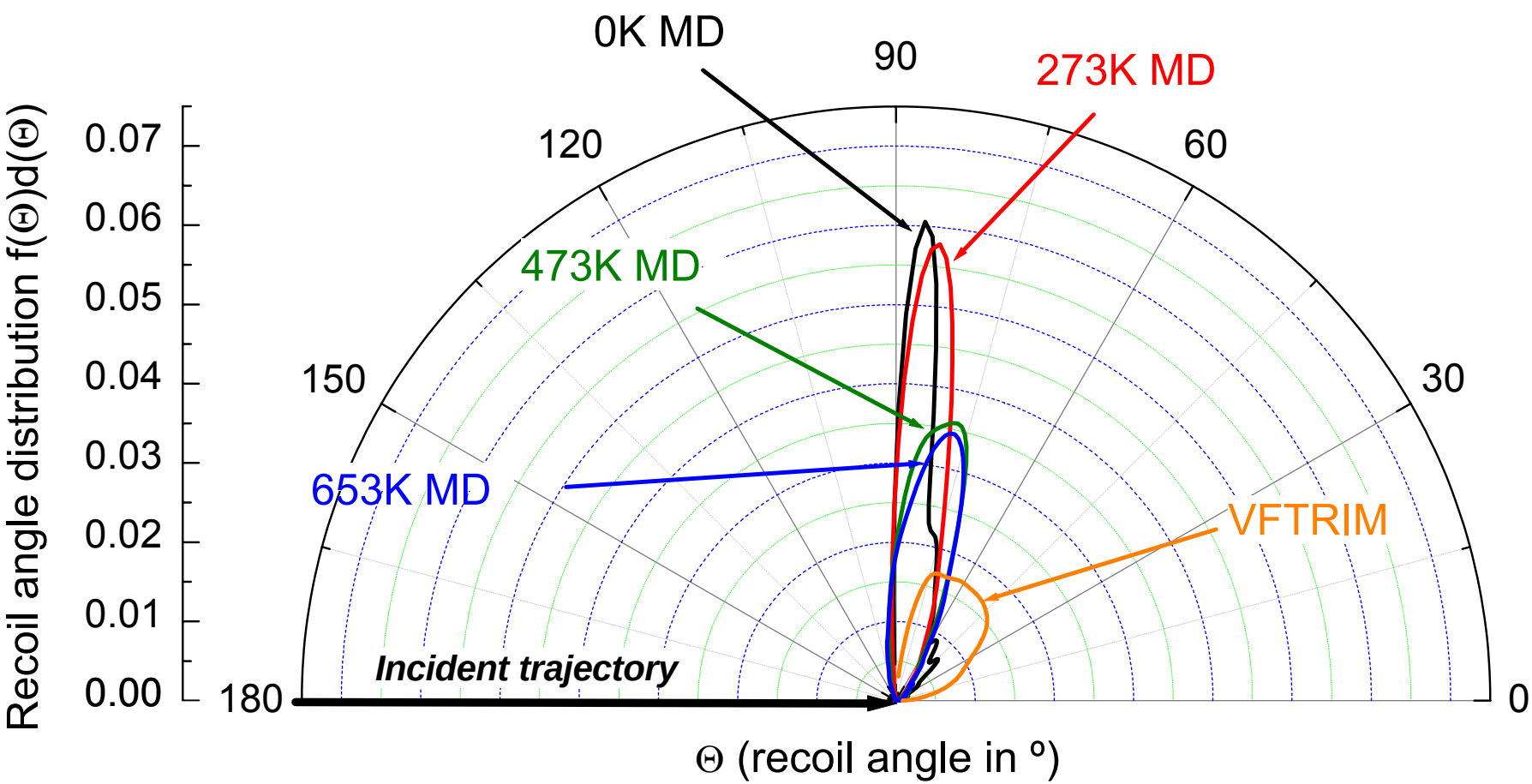
PKA recoil angle distribution depends on *lattice temperature* and simulation method used



- Penetration into BCA-forbidden area of *net* recoil angle due to multi-body effects
- MD shows increasing temperature leads to stronger multi-body effects in recoil creation



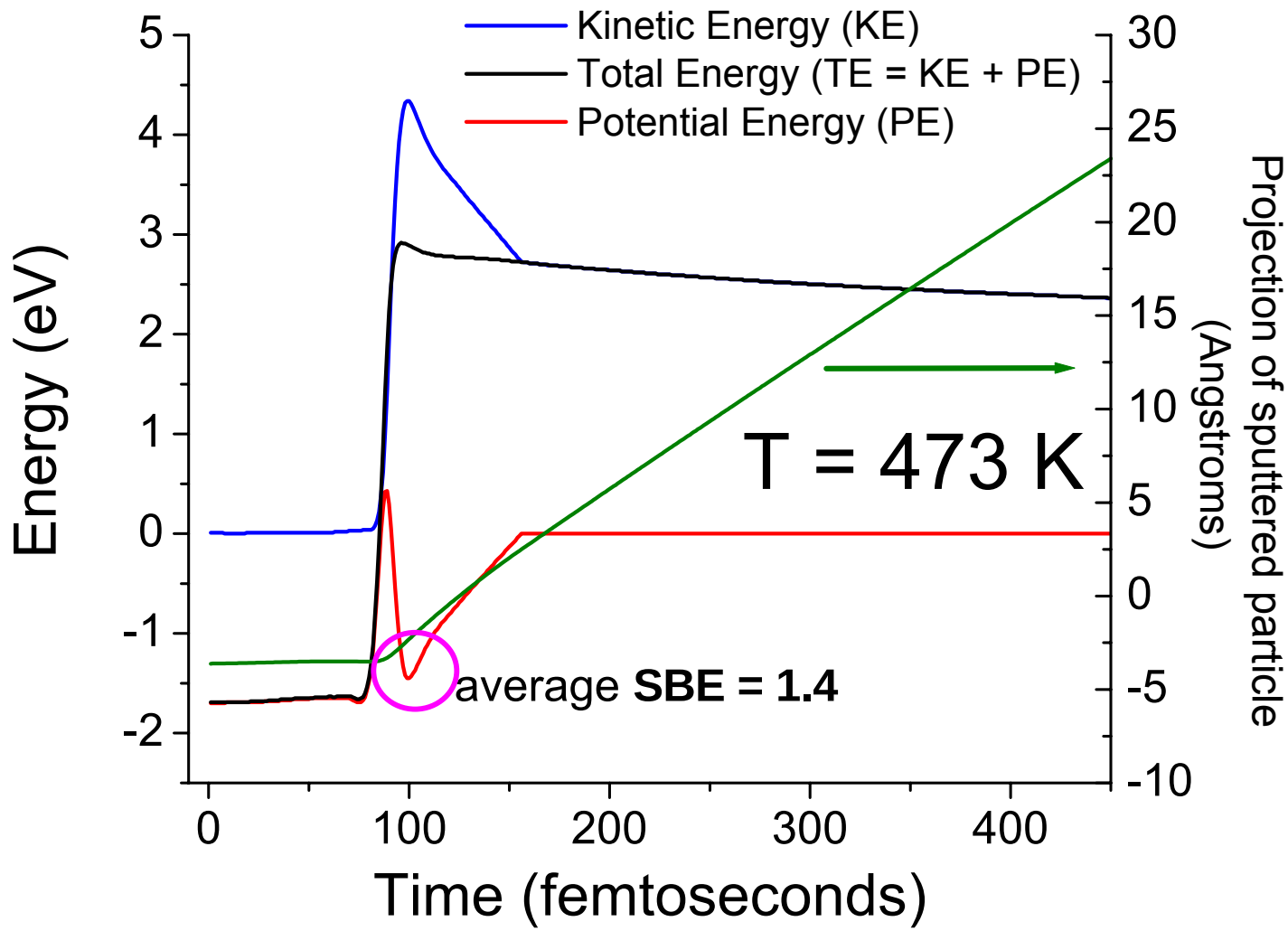
Or alternatively, a polar plot…



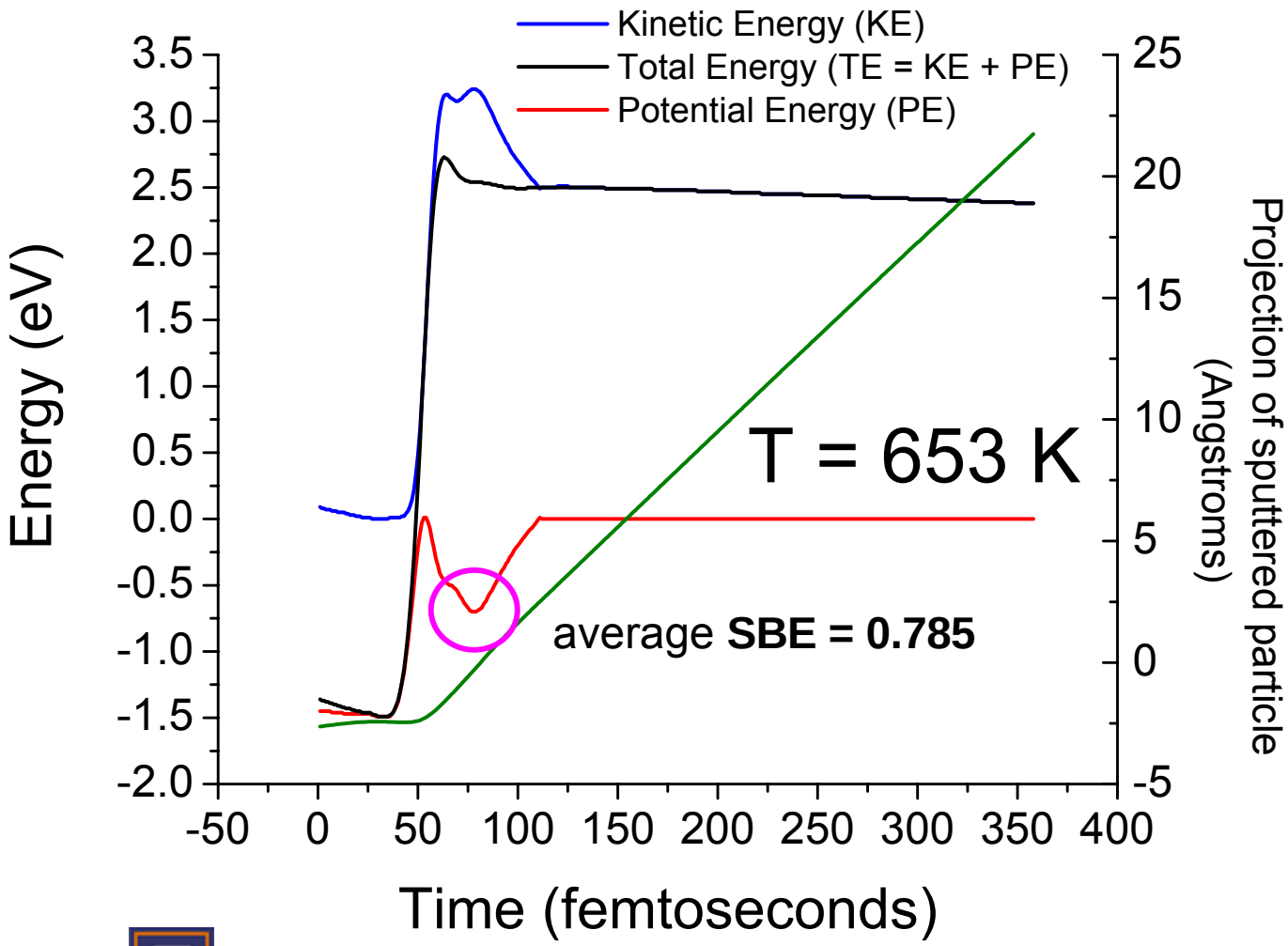
Recoil angles of PKA's for 100 eV Li^+ on Li at 45º
(FFT-smoothed simulation results)

How surface binding energy is obtained from MD simulations (473 K)

- The sbe is obtained from the potential well of the sputtered atom's PE curve
- An average sbe is calculated from the sputtered atoms obtained from 100 MD flights



Surface binding energy from MD simulations at 653K is considerably lower than at 473K!



- Reduction in sbe corresponds to higher sputtering yields in both VFTRIM simulations and in Sigmund's linear cascade theory
- The change in sbe alone, however, is insufficient to explain the complete behavior

Summary of observed temperature-dependent kinematic parameters

- Energy transfer fraction (from incident atom to PKA) distribution
  - *Increases* with increasing temperature (both mean and peak location)
  - Peak widens with increasing temperature
- Recoil angle distribution
  - Mean angle *decreases* with increasing temperature, but profile flattens
  - Greater fraction of recoil angles exceeding 90º with increasing temperature
- Effective surface binding energy
  - Energy lost by bulk atom being removed from lattice (sputtered) decreases with increasing temperature

How to put this knowledge to use?

- MD simulations are not accurate at high energies
  - Time step needs to very small
  - Potential at short distances uncertain

- Use MD knowledge in a BCA framework

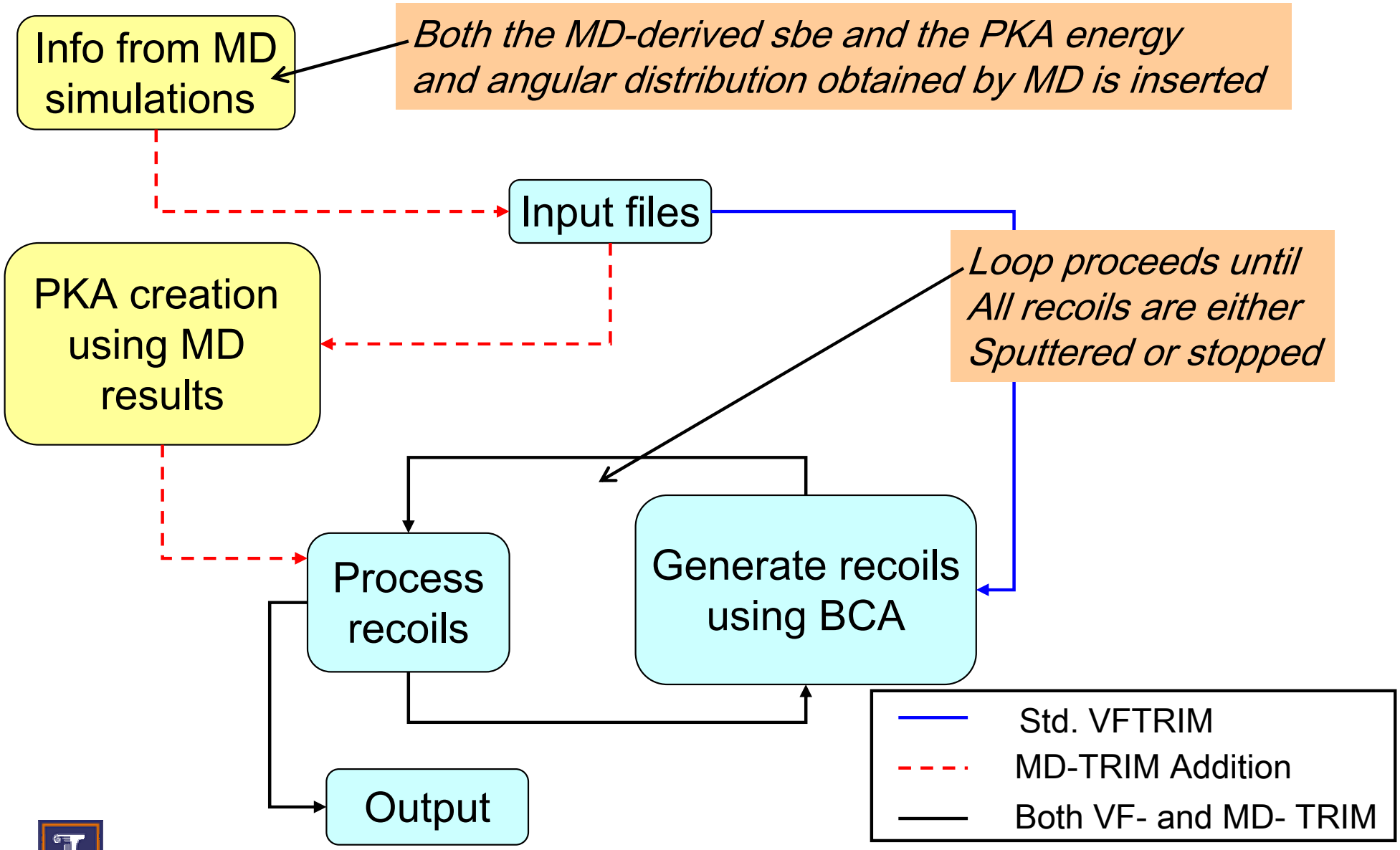
Concept of hybrid-MD code

- Import recoil energy transfer efficiencies and recoil angles from MD results into a Monte Carlo BCA-type code to see the effects of the varying of kinetic parameters with target temperature
- Initial work focuses on using PKA-creation event information for the incident atom and use the BCA model for subsequent generations from the ease of information processing
- Combine the realism of molecular dynamic simulation with the convenience and speed of Monte Carlo

Why hybrid?

- With the vast amount of computing resources available, some have asked why MD cannot be used exclusively to determine PSI results
  - *Energy limitations of MD:*
    - *The lattice necessary to contain the cascade is very large resulting in drastically-increased computing time*
    - *Time step also decreases due to higher atom velocities involved*
  - *Convenience of MC's inherent speed*
  - *Elucidation of physical mechanisms*
  - *Possibly use with multi-scale modeling efforts to include diffusion, segregation, fluid flow*

Flow of MD-TRIM



MD-TRIM Results

Estimated Experimental Results : 0.8 1.2

PKA Dist. Type / sbe	VFTRIM (Std. Ver.)	MD-TRIM Results		
		VFTRIM (BCA Dist.)	473°K (MD Dist.)	653°K (MD Dist.)
0.79 eV	0.8415	1.1130	1.1827	1.2938
1.40 eV	0.4915	0.6430	0.7549	0.8084
1.68 eV	0.4152	0.5180	0.6378	0.7048

Future work in MD and hybrid-MD simulations of liquid lithium systems

- Continue H/D bombardment of liquid Li/LiH (large lattice and small time step require a large amount of processing time)
- Develop a better, more physical surface potential that is consistent with the cohesive energy for the Li atoms within the surface layers and exhibits surface stratification
- Look at the effects of higher temperature on the sputtering and reflection which is leading to predict sputtering yield as a function of surface temperature
- Extend import of MD-derived kinematic information: Use for higher-generation recoils instead of relying on VFTRIM's BCA methods

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