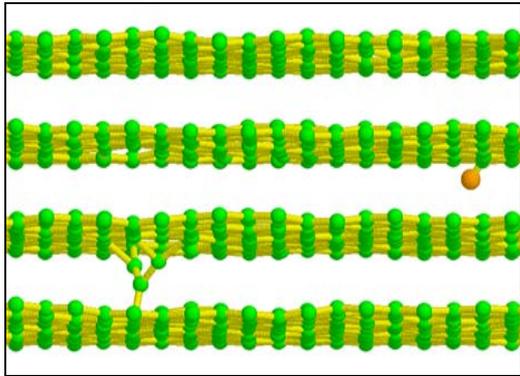
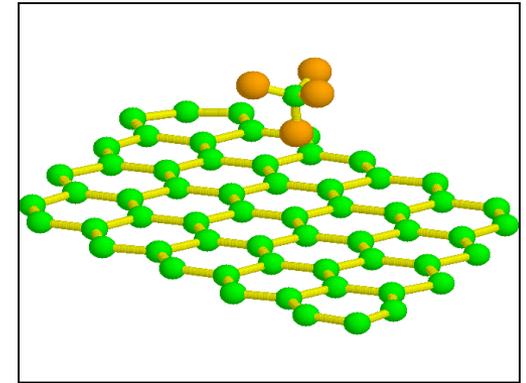


Atomistic simulations of ion-induced erosion and surface evolution



chemistry &
materials
science
MS

E.M. Bringa
ebringa@llnl.gov



with contributions from:

E. Salonen, Kai Nordlund (U. of Helsinki, Finland)

G.H. Gilmer, B. Torralva, M.J. Caturla, L.A. Zepeda-Ruiz (LLNL)

Collaborators:

B.D. Wirth (UC Berkeley), A.J. Caro (LLNL)

R.E. Johnson, L. Zhigilei (UVa)

APEX/ALPS meeting, Grand Canyon, April 2003

*This work was performed under the auspices of the U.S. Department of Energy and
Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48*

Outline



- **Motivation and applications**
- **Available atomistic simulation tools:
MD and BCA codes**
- **Example I: Chemical sputtering of a-C:H**
- **Example II: graphite erosion by H (50-200 eV)**
- **Example III: surface evolution induced by ion bombardment**
- **Other examples (if time permits ...)**
- **Summary and future work**

Motivation: simulating ion-surface interaction



WIDE ENERGY RANGE: 1 eV-10 GeV

Large number of applications:

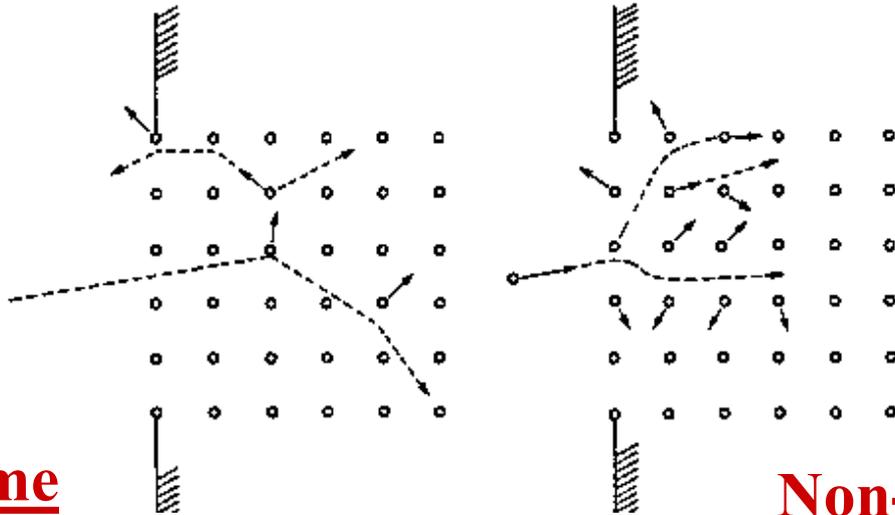
- Plasma wall interaction (fusion)
- Plasma processing of electronic materials
- Radiation damage (mainly bulk effects)
- Surface Physics: ion-beam modification and analysis of materials
- Astrophysical applications: supernova shocks, cosmic ray erosion

Same governing physics!

Collisional cross-sections, primary damage production, atomic diffusion, defect cluster physics, etc.

Laser-materials interaction also shares some of the same physics

Physical Sputtering Regimes



Linear Regime

- Collisions:
moving + static
- Sputtering occurs fast
(\sim ps)
- Events are independent
- Yield is linear in
energy deposition
- 1 keV D on graphite

Non-Linear Regime

- Collisions
moving + moving
- Sputtering persists for
“long” times (\gg ps)
- Events are correlated
- Yield is non-linear in
energy deposition
- 1 keV O on graphite

Tools for Ion-Solid Interaction Simulations



Binary Collision Approximation (BCA)

- Codes: VFTRIM, SRIM, MARLOWE, etc.
- Only works in “linear”, low energy density regime (electronic and chemical sputtering not included)

Hydrodynamics (HD)

- Codes: “HEIGHTS”, M. Jakas code (La Laguna), etc.
- Only Works in “non-linear”, high energy density regime

Molecular Dynamics (MD)

- MDCASK, PARCAS, REED, etc.
- Works in any regime

Same governing physics, different approximations!
Some hybrids tools (*i.e.* MD+BCA, MD+HD) do exist

Limitations of BCA



- BCA codes do not work well at low energies ($KE < 50 E_{cohesion}$) when collisions are no longer binary and many body collisions take effect. Need additional models for electronic or chemical sputtering.
- There are few datasets for collisions below few keV, and extrapolations are used, leading to results which may not be reliable.
- Surface effects are included poorly, since surface binding is a many body effect. Therefore sputtering calculations where low energy ejection occurs should be taken with care.
- Artificial corrections have to be applied to account for binding effects in alloys and molecular solids.
- Cluster bombardment gives only “linear” effects.
- Despite its limitations **BCA** is a very **powerful tool** to study Ion-Solid interactions in the **linear** regime because it is relatively fast (hours). Simulations with “large” samples, and layers of different compositions can be carried out.

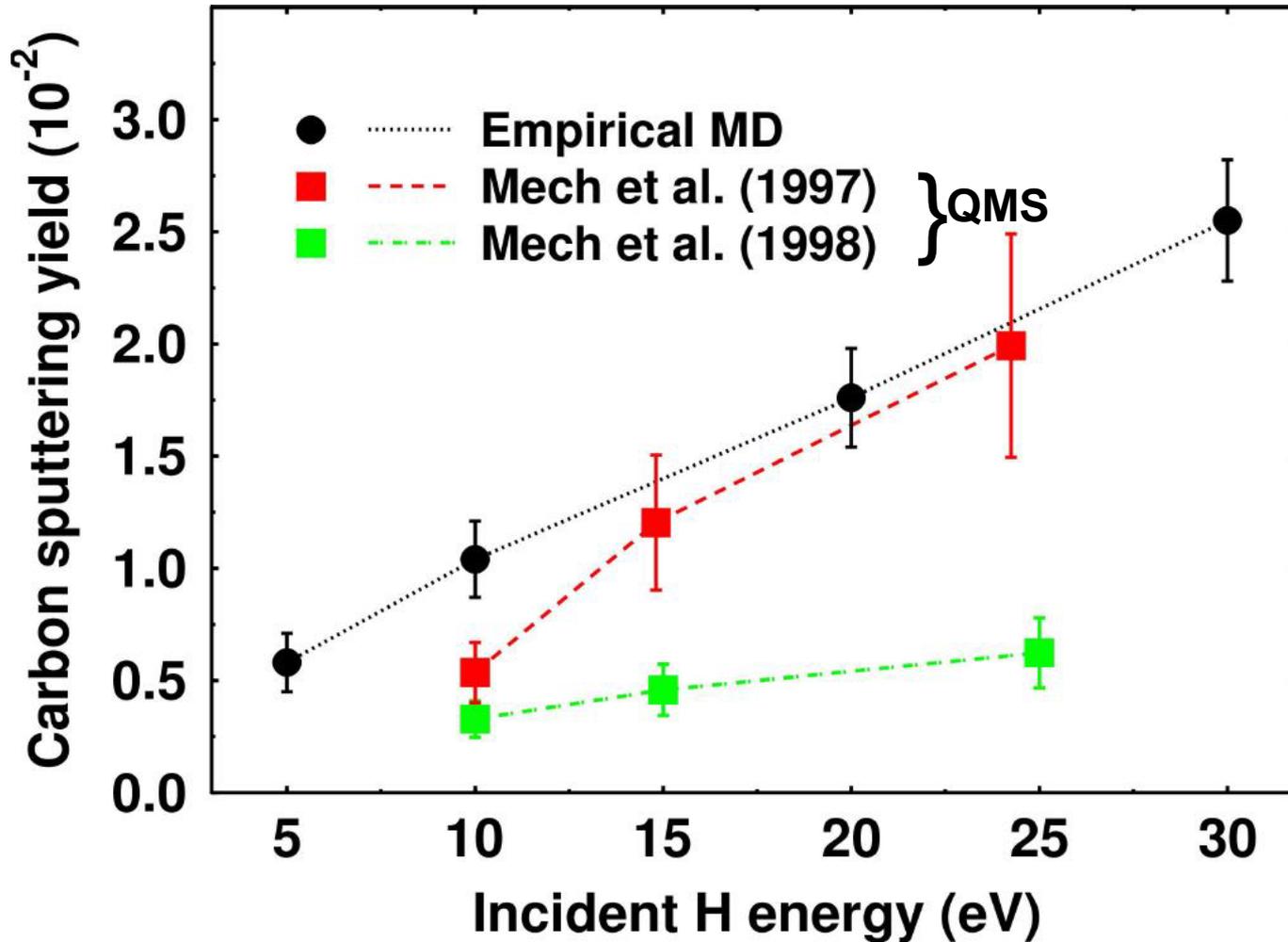
Limitations of MD



- Can simulate only small samples ($L < 1 \mu\text{m}$) – problem to study surface topography/roughness at large scales.
- Can simulate only short times ($t < 10 \text{ ns}$) – problem for large fluxes/re-deposition, long diffusion/thermal effects, etc.
- Computationally expensive for good statistics (weeks)
- Interaction potentials for alloys, molecular solids, and excited species not well known, but can be developed based on ab-initio calculations and experimental data.
- Simple models to account for electronic effects (charge exchange, ionization, e-phonon coupling, etc.) do exist, but are generally not accurate and include several parameters.

Despite its limitations, MD is the **most powerful tool** to study ion-solid interactions in both **linear** and **non-linear** sputtering regimes

Example I: a-C:H erosion



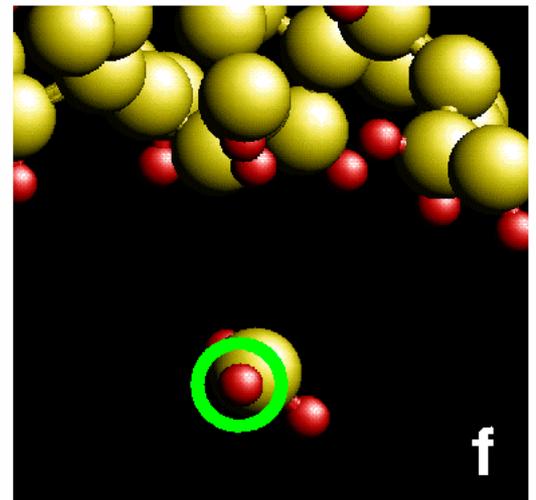
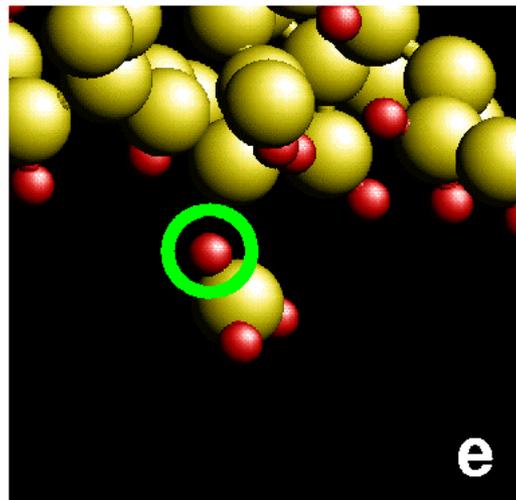
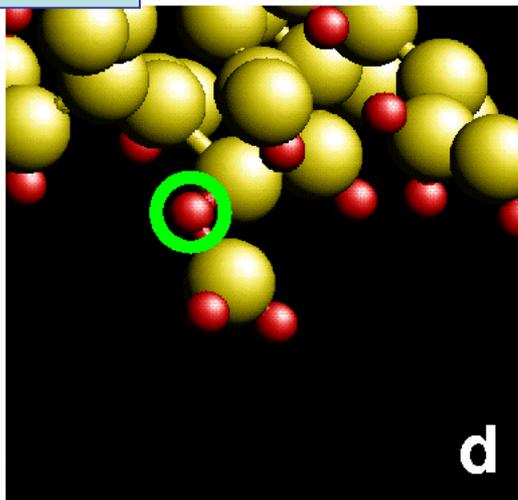
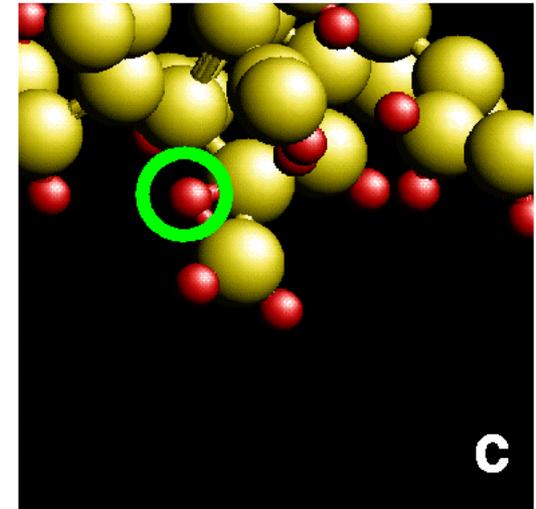
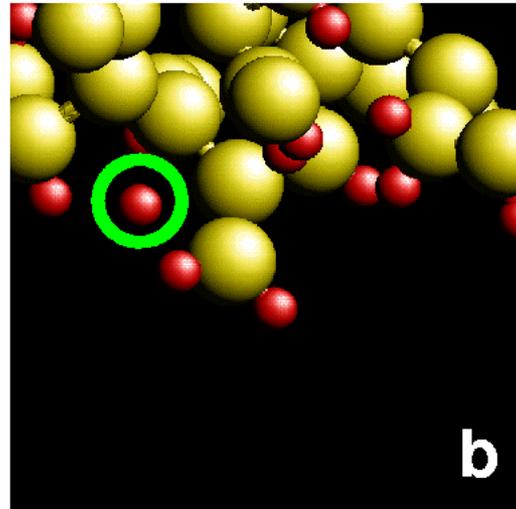
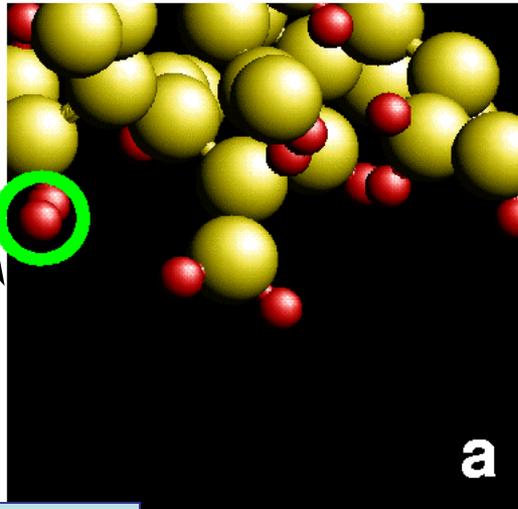
TRIM: $Y \sim 10^{-3}$
at ~ 50 eV

Experiments are difficult and can give very different results!

H_3 , D_2 beams instead of atomic beams, typically $Y_{QMS} < Y_{WEIGHT}$

CH₃ radical sputtering by a 10 eV H atom

E. Salonen et al, Phys. Rev. B **60** (1999) R14005 and **63** (2001) 195415



projectile

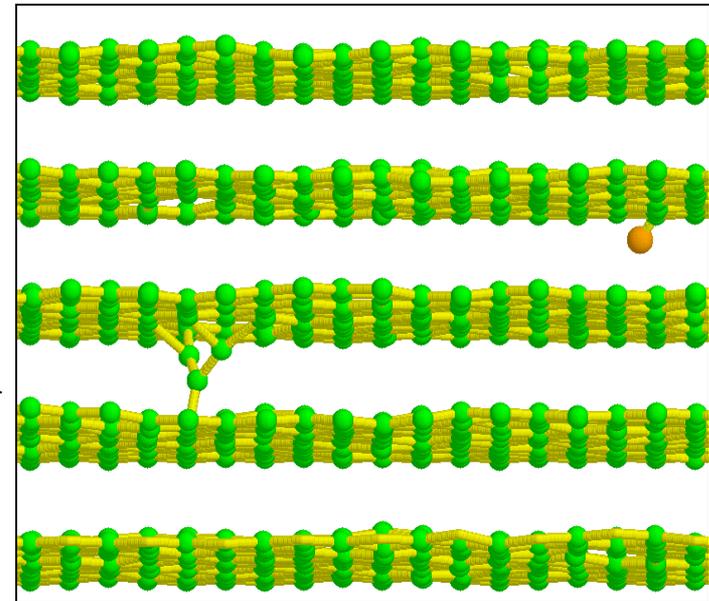
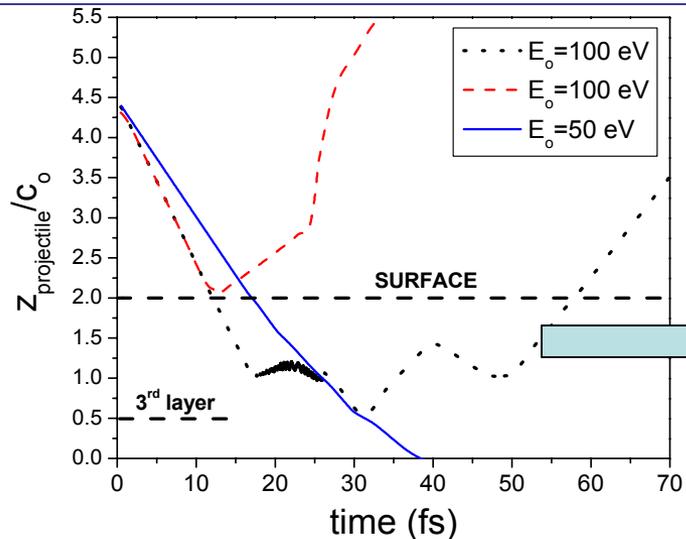
Bond breaking crucial in chemical sputtering. Event only lasts ~100 fs

Example II: H bombardment of graphite



Simulated with **MDCASK** (LLNL):
highly parallel, variable time step,
Potential: Brenner+long range+ZBL
Target: 7,000 C atoms, 300 K

H projectiles, 50-200 eV, normal incidence, hitting randomly an irreducible region on the surface.



Our code: ~1 ms/atom/step/CPU in ASCII Frost. Using 256 CPU's it would take ~4.5 days to run 10^3 D impacts at a target (10^4 atoms), during 1 ps each, i.e. to get a value of sputtering yield with good statistics.

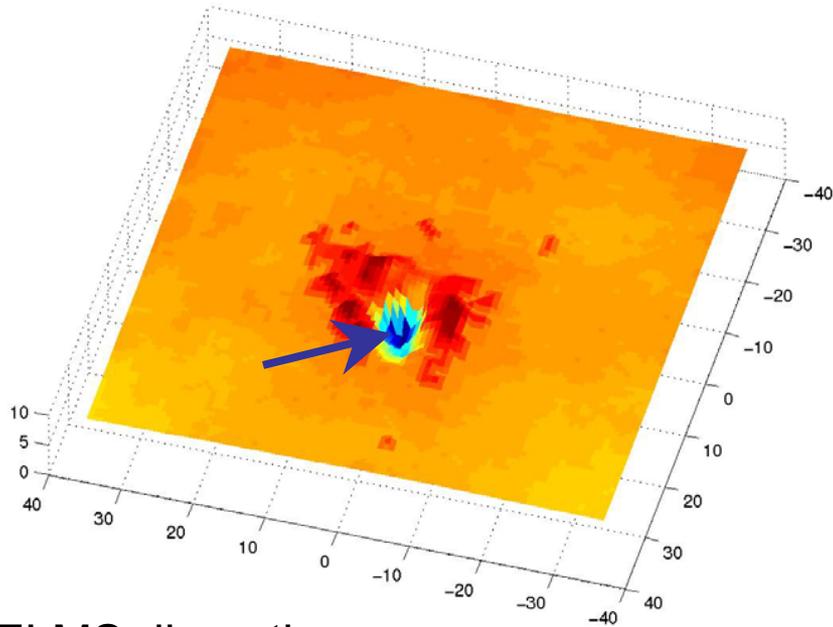
Snapshot at 40 fs for 100 eV H bombardment. Few C atoms displaced → damage few layers below the surface, as previously seen for simulations of keV impacts [Smith & Beardmore, Thin Sol. Films **272**, 255 (1996)]. Projectile has been temporarily “trapped” between two graphite layers, moving parallel to the surface.

Example III: surface evolution



MD: short time, large energy densities

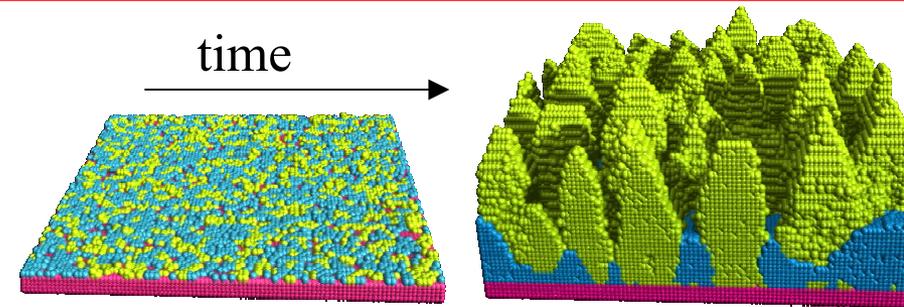
MeV ion-induced cratering in polymers,
E. Bringa *et al*, PRB **65** (2002) 094113



ELMS disruptions

Kinetic Monte Carlo (KMC): long time, diffusional processes

Al deposited at $1\mu\text{m}/\text{min}$, $T = 100\text{K}$ (G. Gilmer, LLNL). **Competitive growth** (low mobility): clusters with (001) orientation (yellow) catch more sputtered atoms, and shadow (111) clusters (blue).



Transition probabilities and migration paths obtained from ab-initio MD, classical MD, and experiments

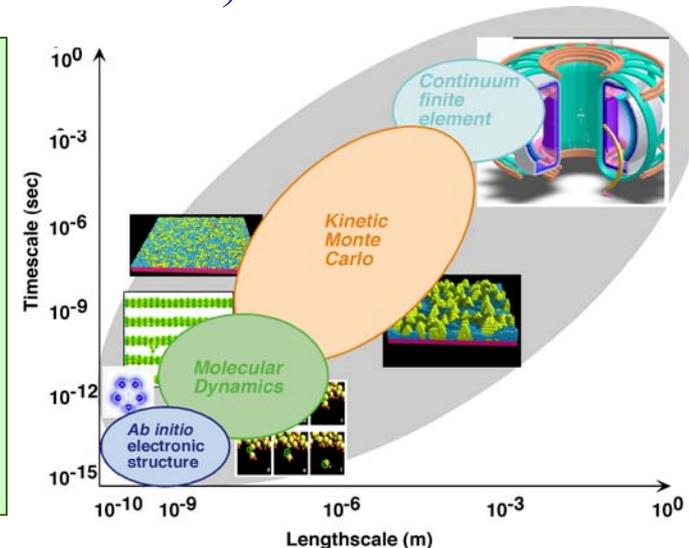
FUSION: Need to account for erosion/redeposition and complex plasma composition \rightarrow couple MD with KMC and plasma codes (WBC/UEDGE) to obtain more realistic surface evolution (topography/composition/etc.)

Conclusions



- Both BCA and MD are flexible and powerful tools to study ion-solid interaction, but they both have several limitations, should be combined into hybrid tools and calibrated with experimental data.
- MD can obtain “real” time evolution of the simulated system, temperature profiles that can be used in rate equations, desorption and sputtering as a function of projectile type, angular incidence, energy and angular spectra of ejecta, surface modification and damage.
- Carbon erosion due to “chemical” sputtering by H/D/T \Rightarrow MD could guide difficult experiments (it cannot be treated with BCA)

More realistic, multiscale simulations are needed for surface composition and topology, charging, erosion by other ions, etc. Use computational power and software available at LLNL to collaborate with the ALPS/APEX community in solving fusion problems

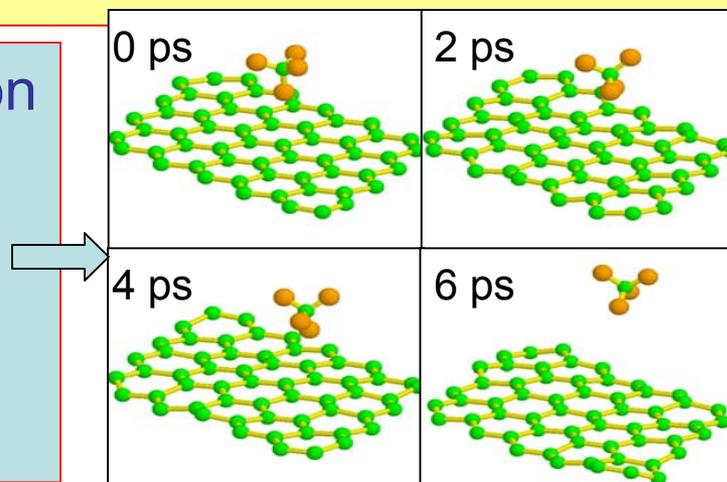


Future Work



- **COUPLE** Y(energy, angle, surface temperature, ...) with plasma codes
- **Surface evolution** → use multiscale modeling: MD and plasma codes as input to KMC evolution of surface topography
- **Tritium retention** evaluation

IN PROGRESS: Photo-induced desorption of organic molecules. 3 eV photo-desorption of methane from a graphite surface using non-adiabatic quantum molecular dynamics [B. Torralva et al., Phys. Rev. B **64**, 153105 (2001)]



More experimental results on energy loss, sputtering and ejecta distribution, together with coupling among different models, and better theories, are needed to understand the details of sputtering and surface modifications

Additional results and simulation movies available

