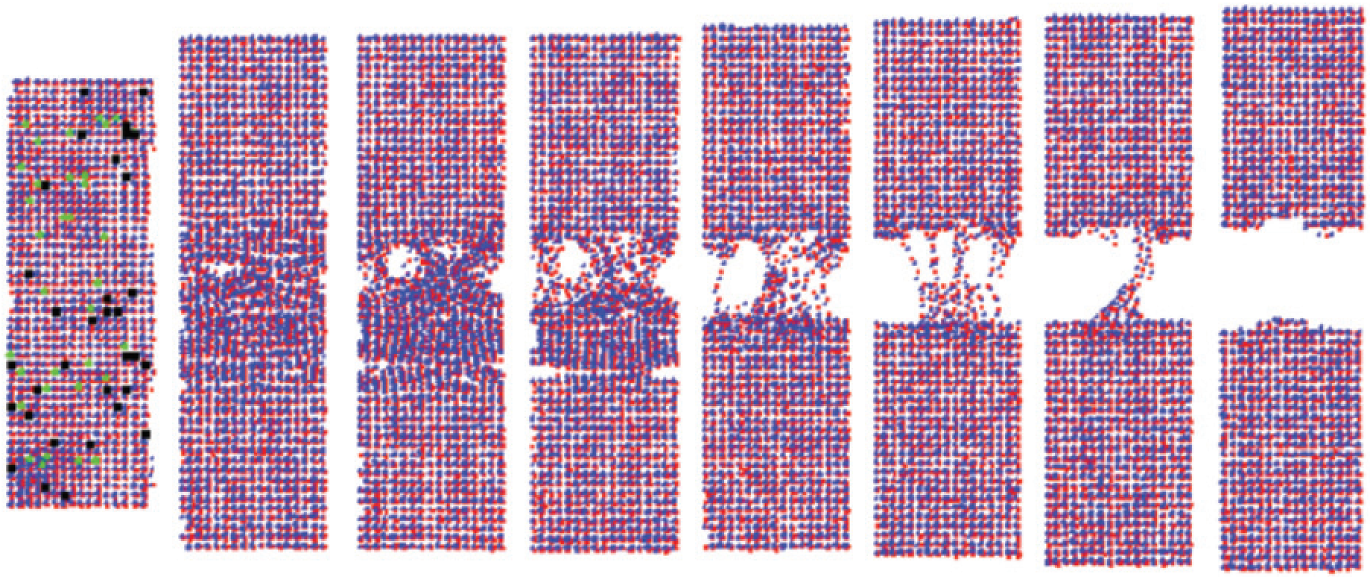
Modeling of Laser Ablation of LiF - Influence of Defects

H. M Urbassek, Y. Cherednikov
Physics Dept., University of Kaiserslautern, Germany

N. A. Inogamov
Landau Institute, Russian Academy of Science



Theoretical tools

- **Molecular dynamics**

Solve Newton ‘s equations.

Advantages:

- Input: only interatomic forces, nowadays available for many materials
- as realistic as possible
  - for many-body simulations
  - for thermal nonequilibrium situations
- easy visualization / animation: appeals to imagination

Disadvantages:

- computationally slow
- cannot handle time scales & 1 ns
- cannot handle space scales & 100 nm [1 μm]

Isaac Newton (1643 – 1727)
1687: Philosophiae Naturalis Principia Mathematica

Outline

- **Two-temperature model / MD for metals**
- Two-temperature model / MD for LiF
- Melting and spallation of thin LiF films
- defects
- swift-ion tracks in LiF

Two-temperature model + MD for metals:

assumes electronic and atomic system to be internally thermalized with temperatures T_e, T_a

heat conduction equation for electrons
Newton ‘s equations for atoms

electron-ion coupling terms

$$C_e(T_e)\frac{\partial T_e}{\partial t} \;=\; \frac{\partial}{\partial z}\left(\kappa\frac{\partial}{\partial z}T_e\right) - g\cdot(T_e - T_l) + S(z,t)$$

$$M\frac{d^2\mathbf{r}_i}{dt^2} = -\nabla_{\mathbf{r}_i}V(\{\mathbf{r}_j\}) - \frac{g}{C_a}\frac{T_a - T_e}{T_a}M\frac{d\mathbf{r}_i}{dt},$$

Schäfer, Urbassek, Zhigilei 2002

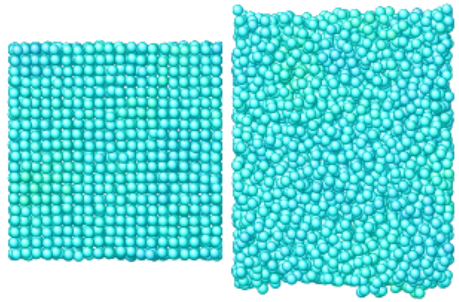
typical results for thin metal films:

here: Al

with increasing energy input

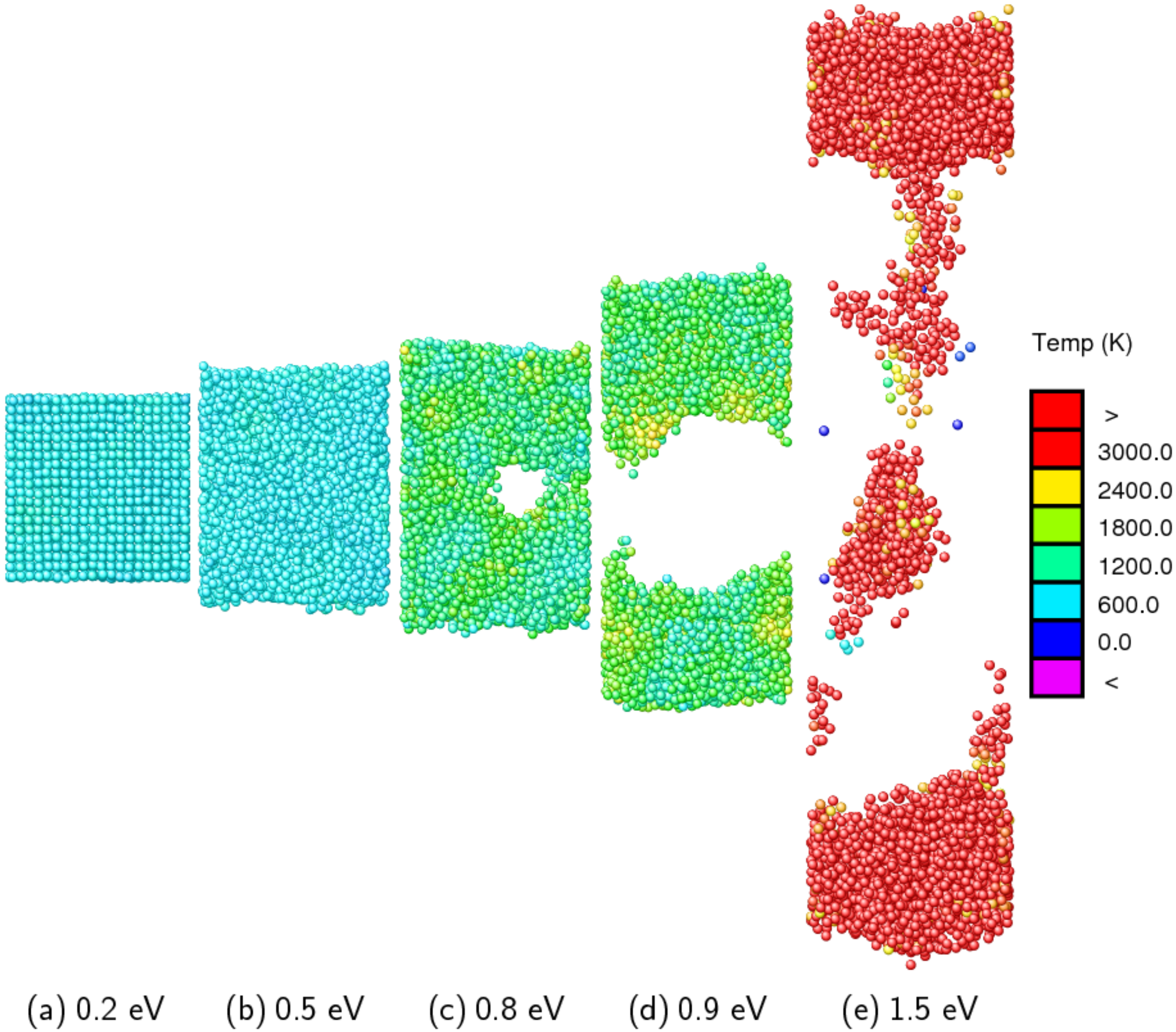
E_0 = absorbed energy / atom

melting ...



(a) 0.2 eV (b) 0.5 eV

metal
(Al)
target



melting

spallation

multi-fragmentation

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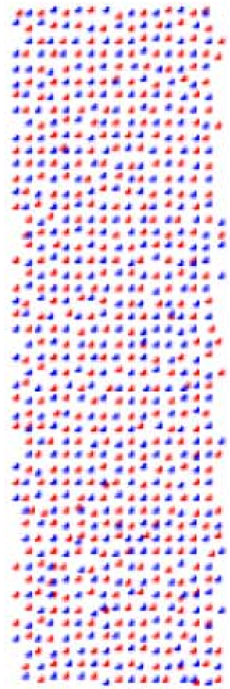
System:

LiF thin Film (10 nm)
(100) surface
lateral size

X-ray pulse: 7 ps
photon energy 90 eV



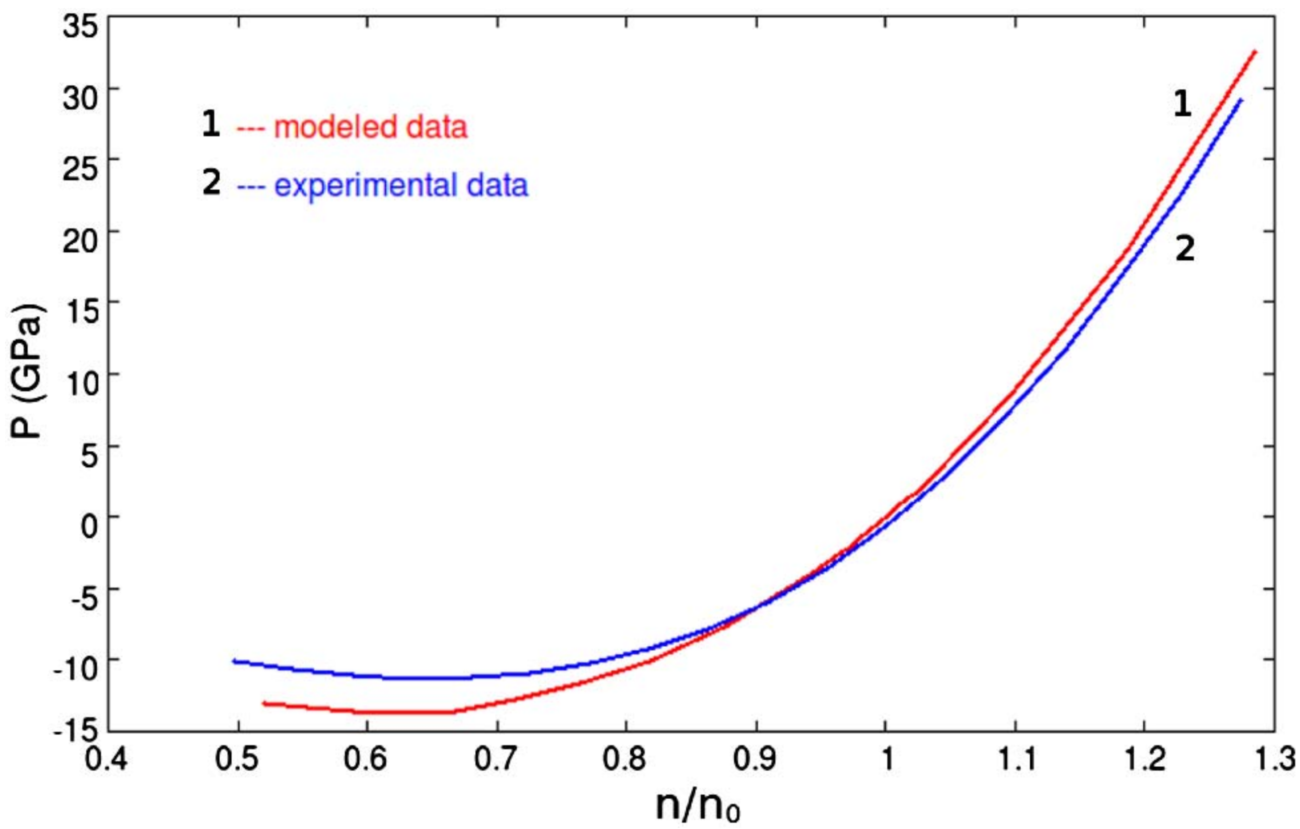
MD:

Buckingham potential + dispersion forces

$$V_{ij}(r) = \frac{q_i q_j}{4\pi\epsilon_0 r} + A_{ij}\exp(-r/\lambda_{ij}) - \frac{C_{ij}}{r^6}$$

describes well:

elastic constants
yield strength
melting temperature (T_m = 1118 K)
...

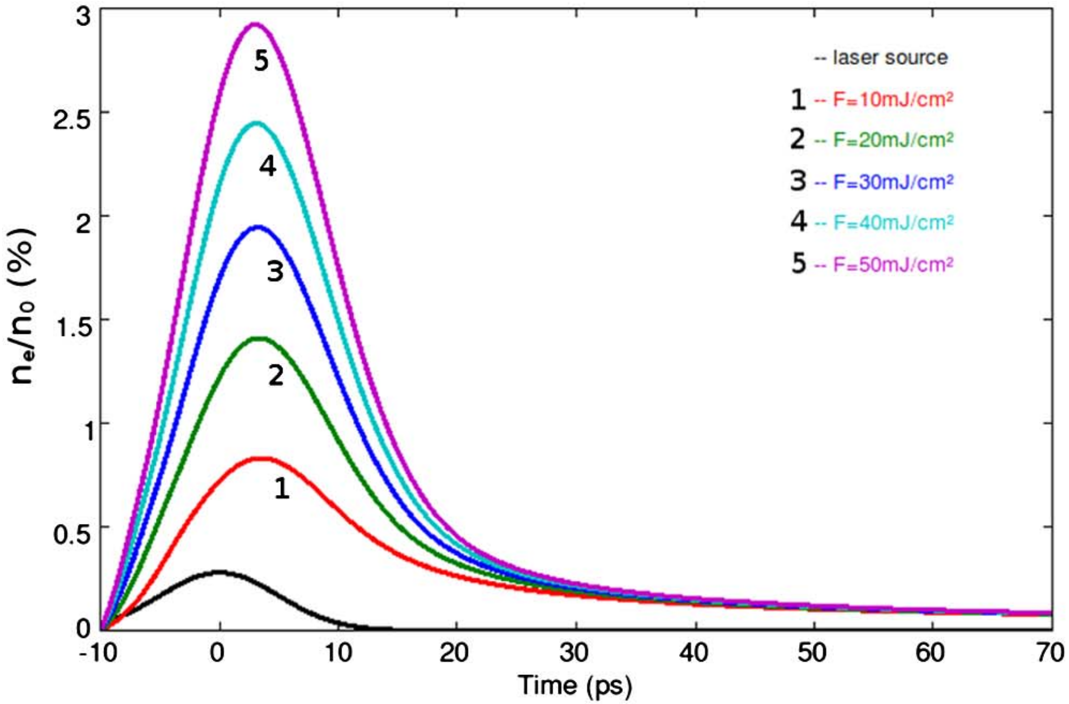


cold curve (T=0)

LiF
electron kinetics

n_e: electron density in conduction band

$$\frac{dn_e}{dt} = \frac{Q}{E_{\mathrm{gap}}} + \nu_{\mathrm{imp}} n_e - \kappa_{\mathrm{rec}} n_e^3$$



Q(t): laser source
ν_{imp}: impact ionization
κ_{rec}: recombination
E_{gap}: gap energy

Result:

Electron concentration
< 3 %

Inogamov et al 2009

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Q(t): laser source
ν_{imp}: impact ionization
κ_{rec}: recombination
E_{gap}: gap energy

E_e: electron energy in conduction band

$$\frac{dE_e}{dt} = Q - \left(\frac{dE}{dt}\right)_{e \to a} = Q - AE_e$$

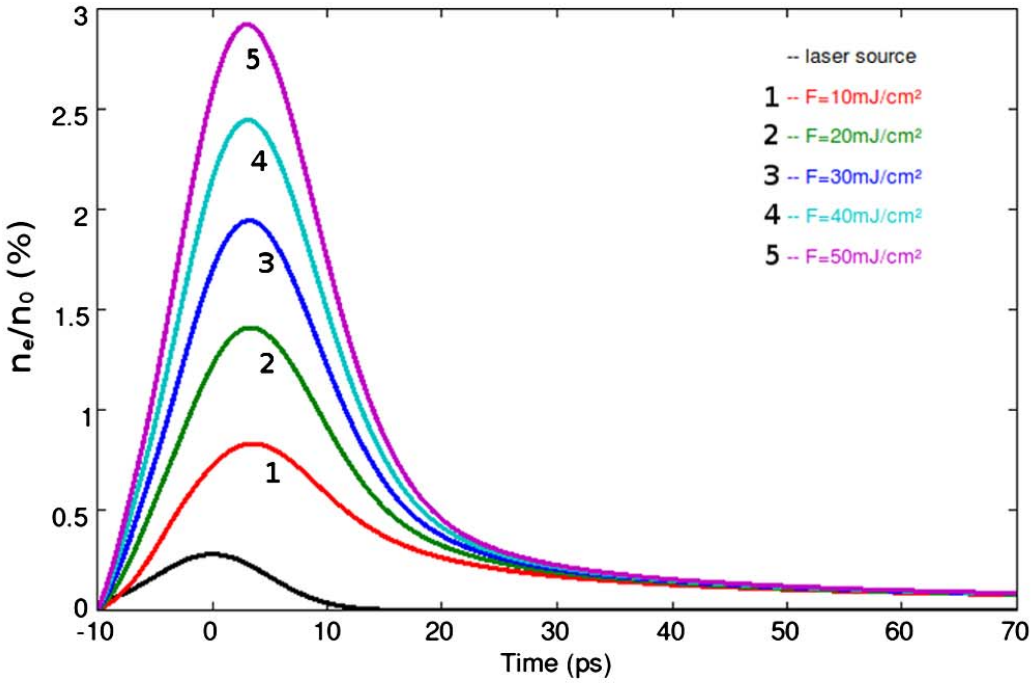
A: energy transfer in electron-atom collisions

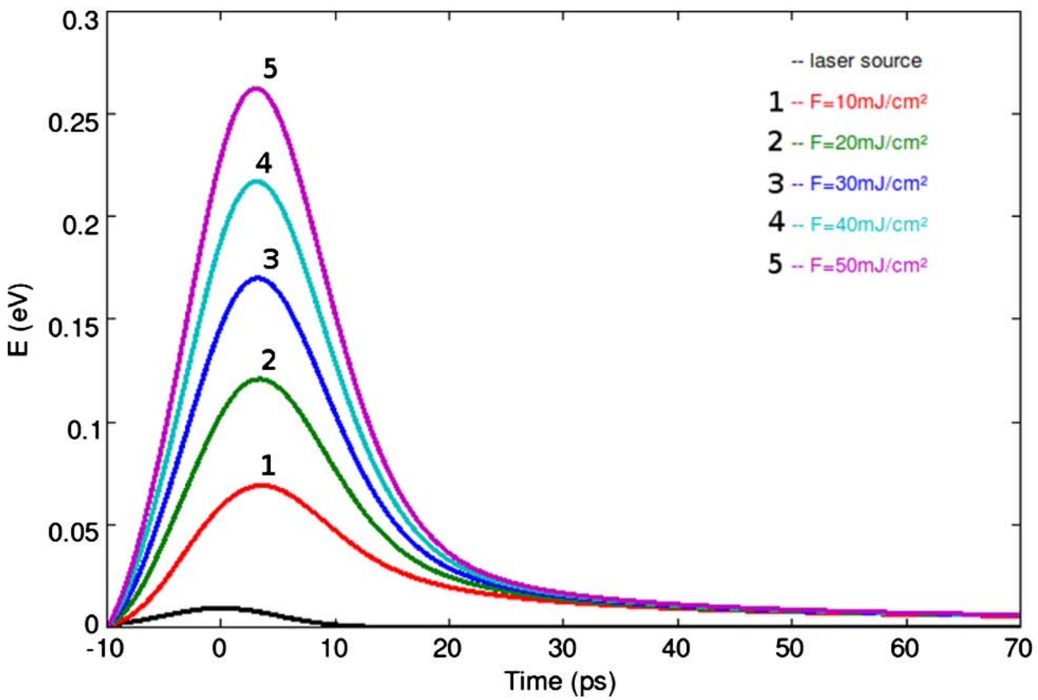
T_e: electron temperature

A = 1 / ps

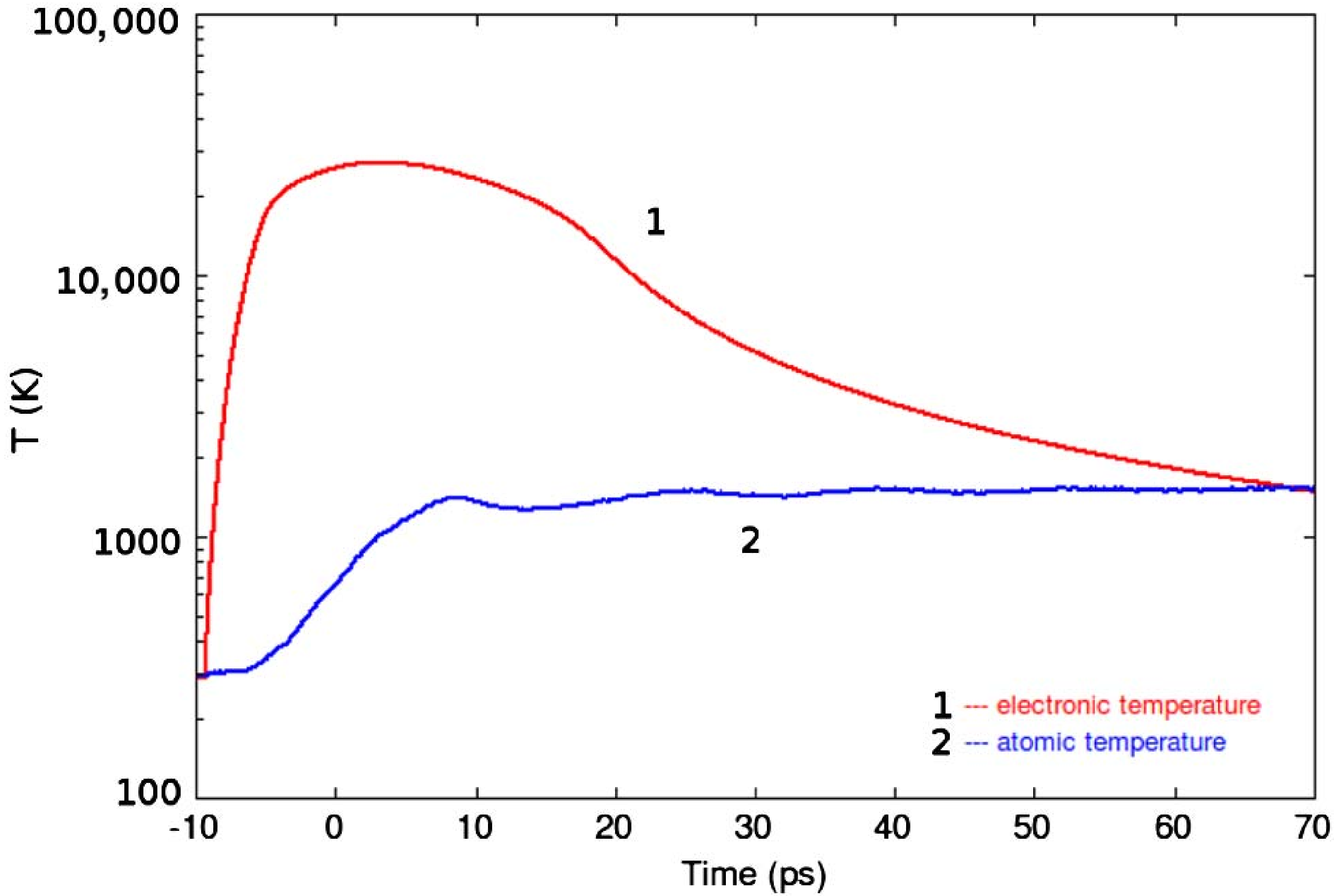
$$E_e = n_e E_{gap} + E_{e,kin} = n_e E_{gap} + \frac{3}{2} n_e k T_e$$

Inogamov et al 2009





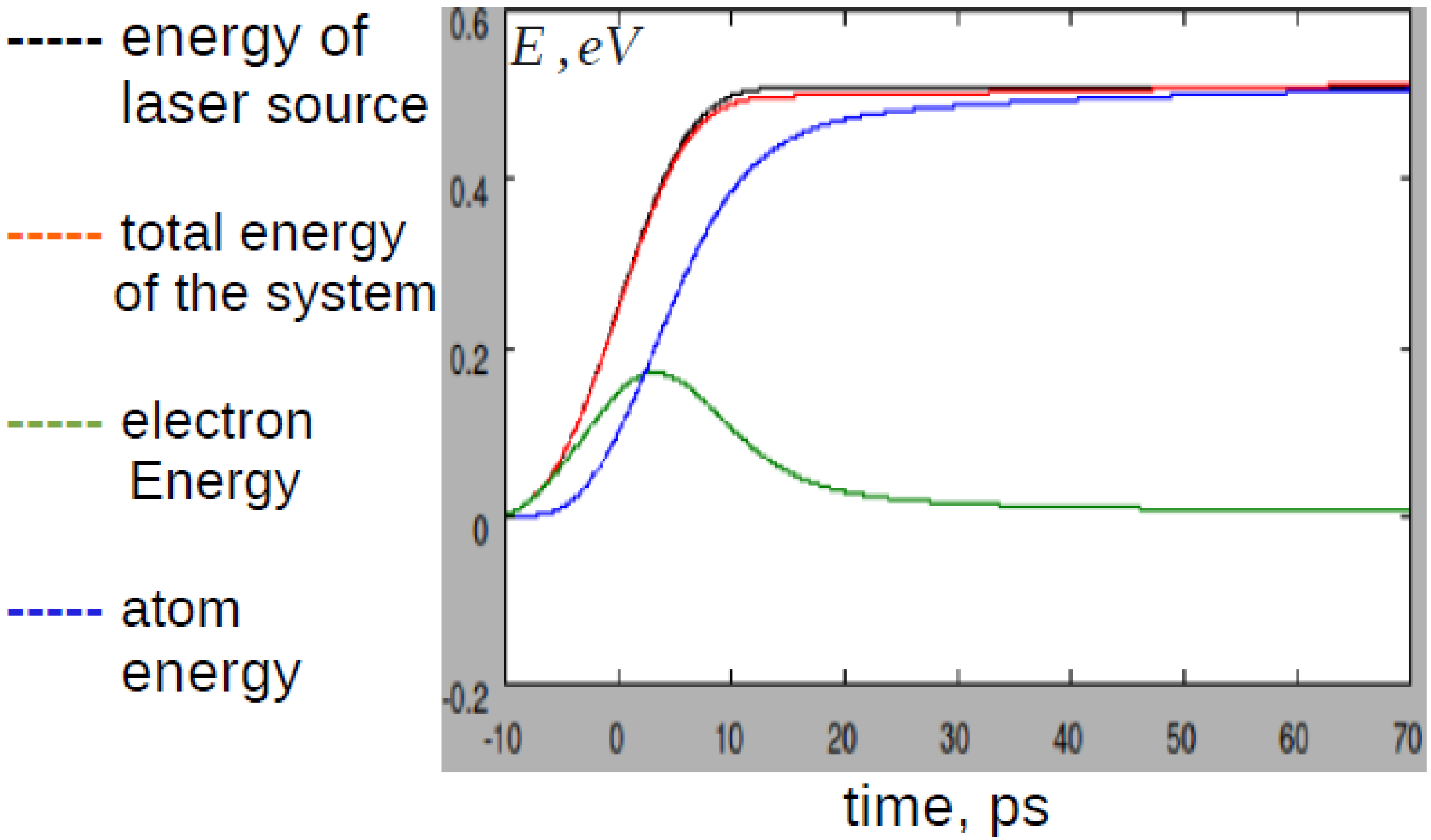
electron kinetics



Electron and atom temperatures
after F= 30 mJ/cm^2

LiF
coupling of electron kinetics and molecular dynamics

Energy conservation (per atom)



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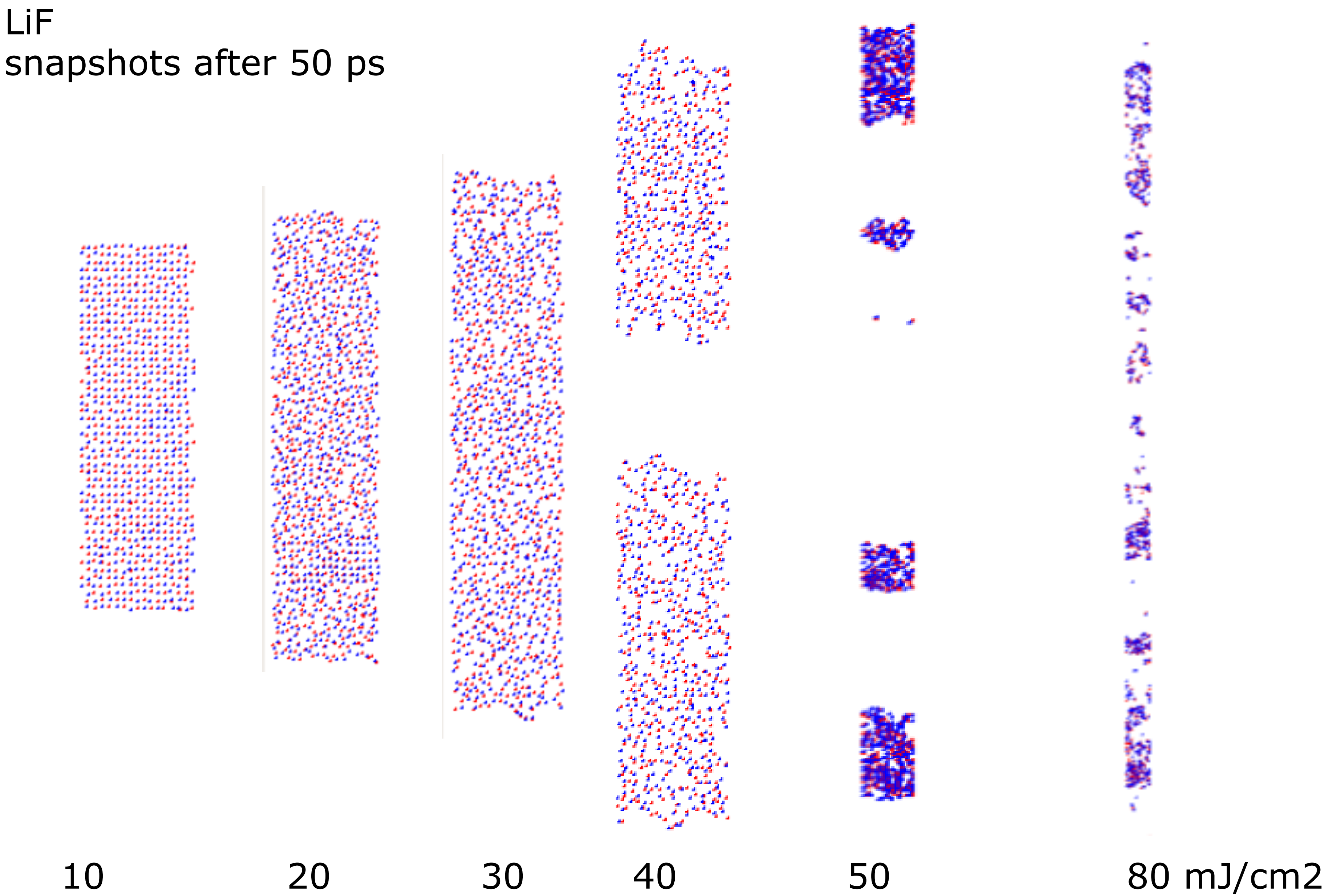
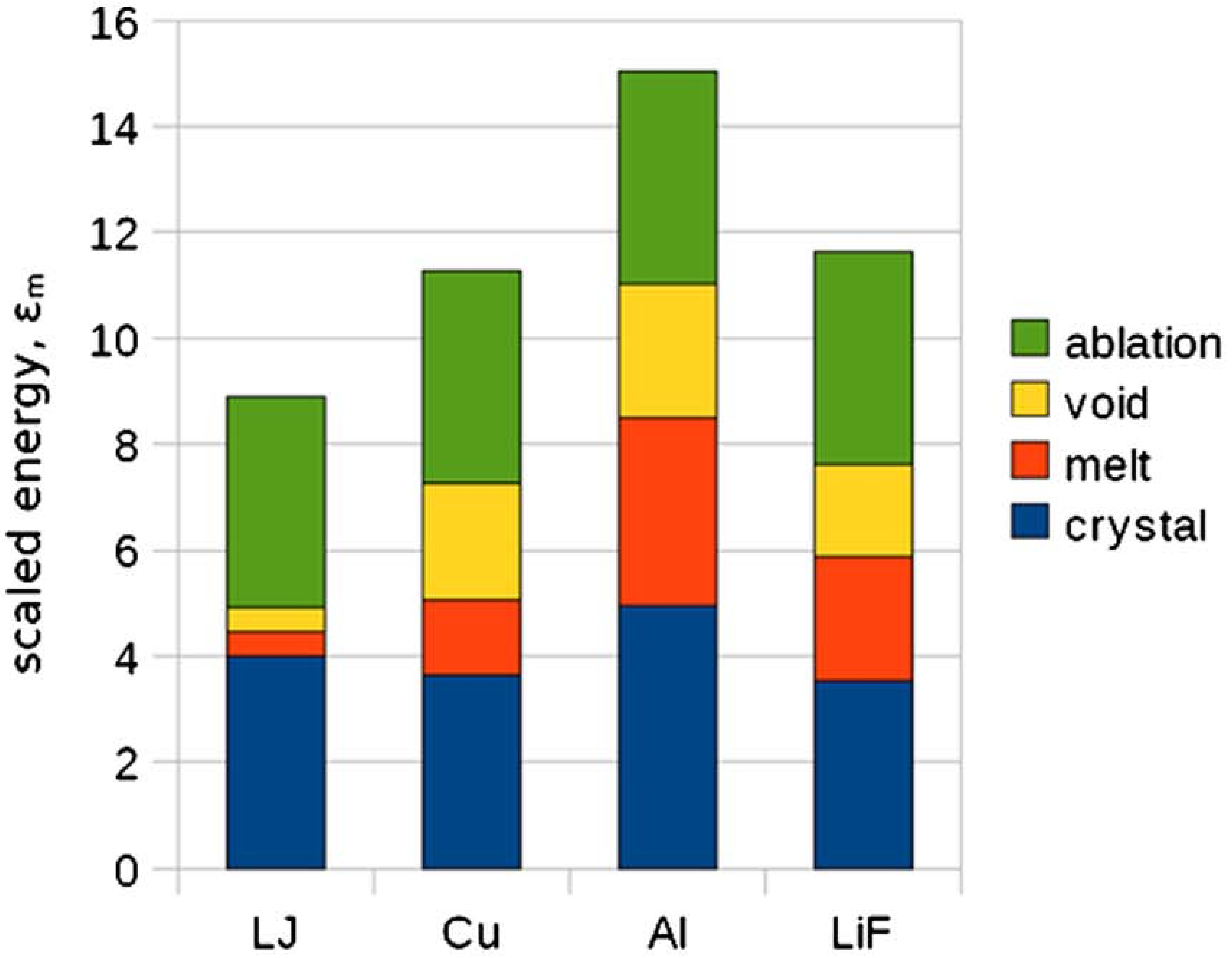
Cherednikov et al., J. Opt. Soc. Am. B 28, 1817 (2011)


LiF
snapshots after 50 ps
10
20
30
40
50
80 mJ/cm2



Synopsis:threshold energies for various material classes

Absorbed energy / atom is scaled to melting temperature: E_0/kT_m

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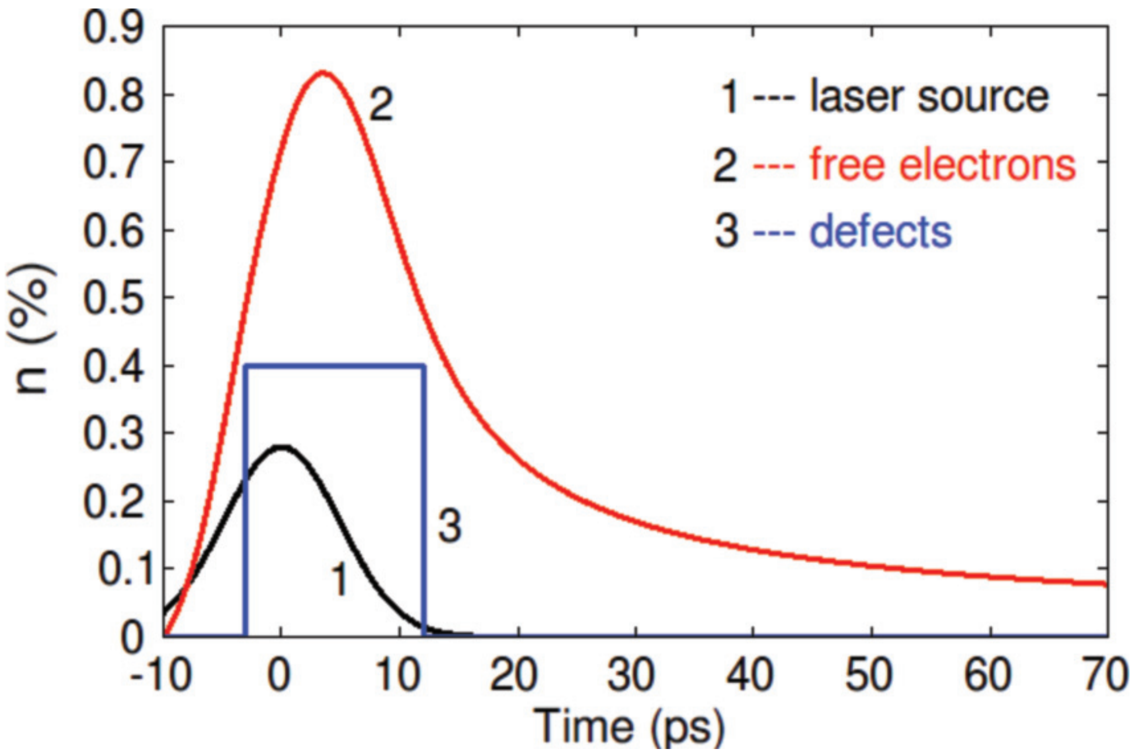
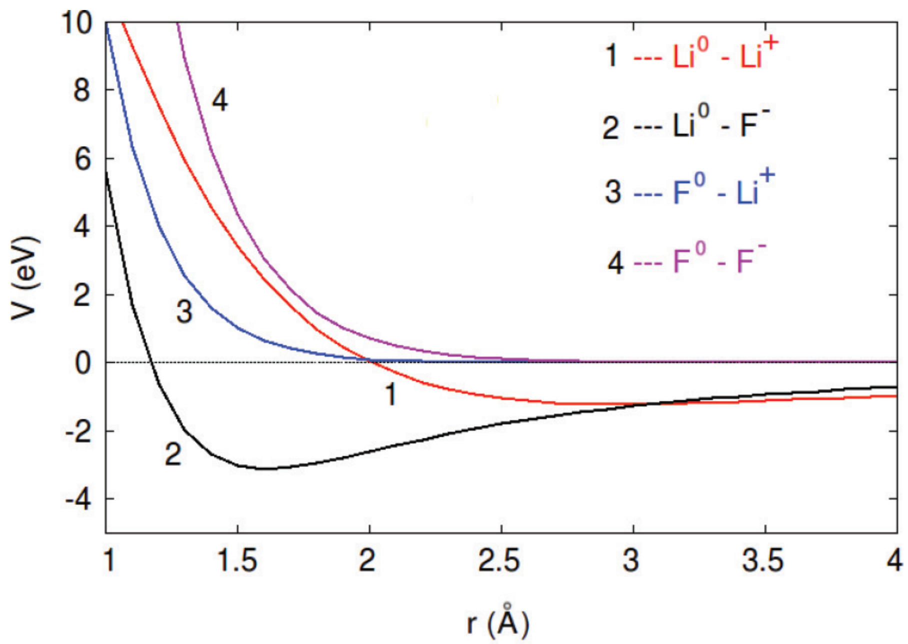
Cherednikov et al., Phys Rev B 88, 134109 (2013)

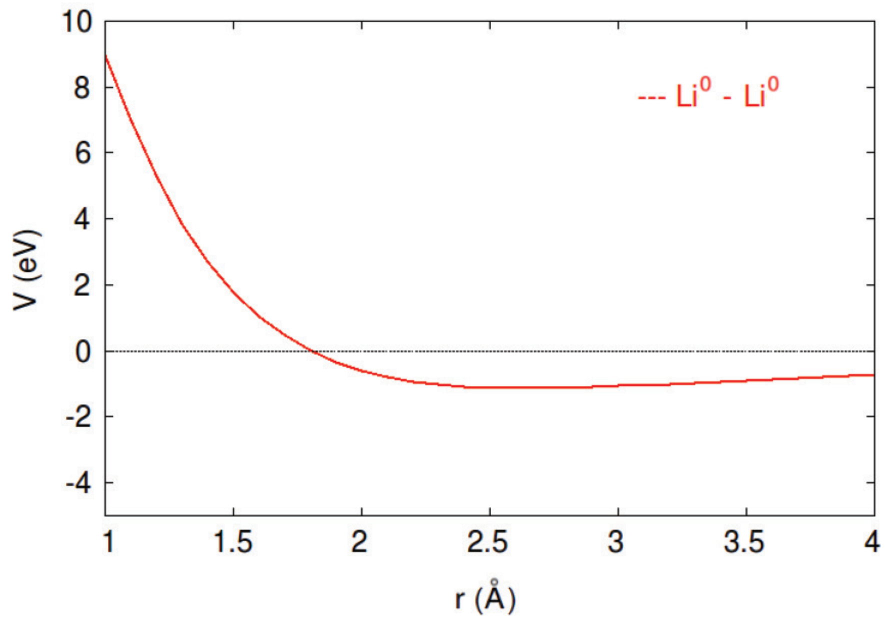
Defects : neutral Li and F atoms

$F^- \rightarrow F^0 + e^-$
$Li^+ + e^- \rightarrow Li^0$

Defects are introduced *ad hoc*

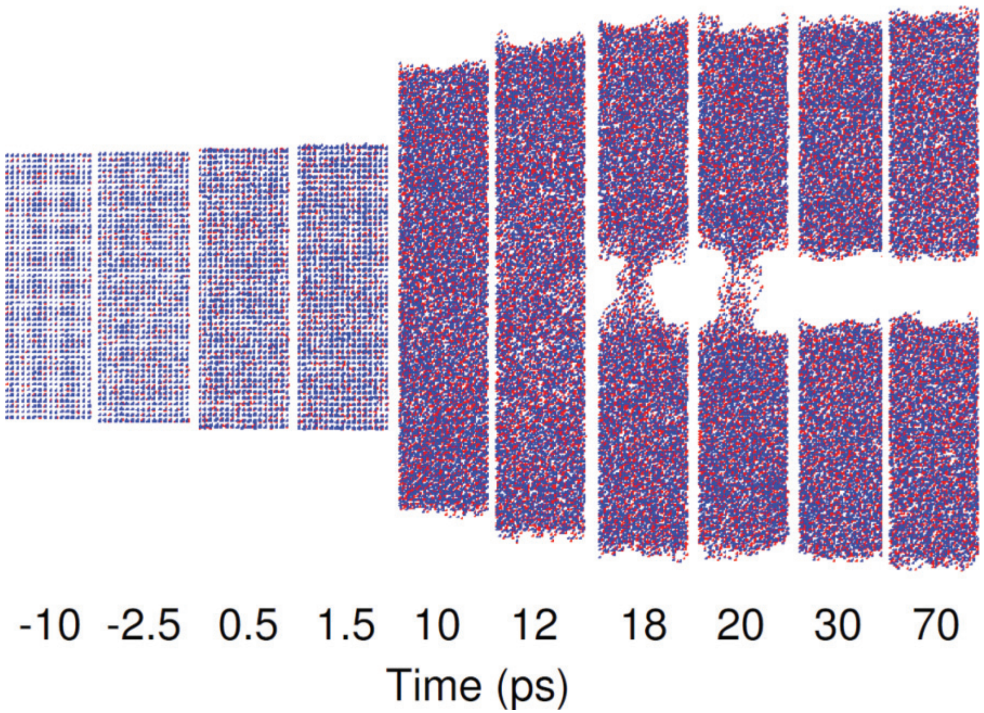


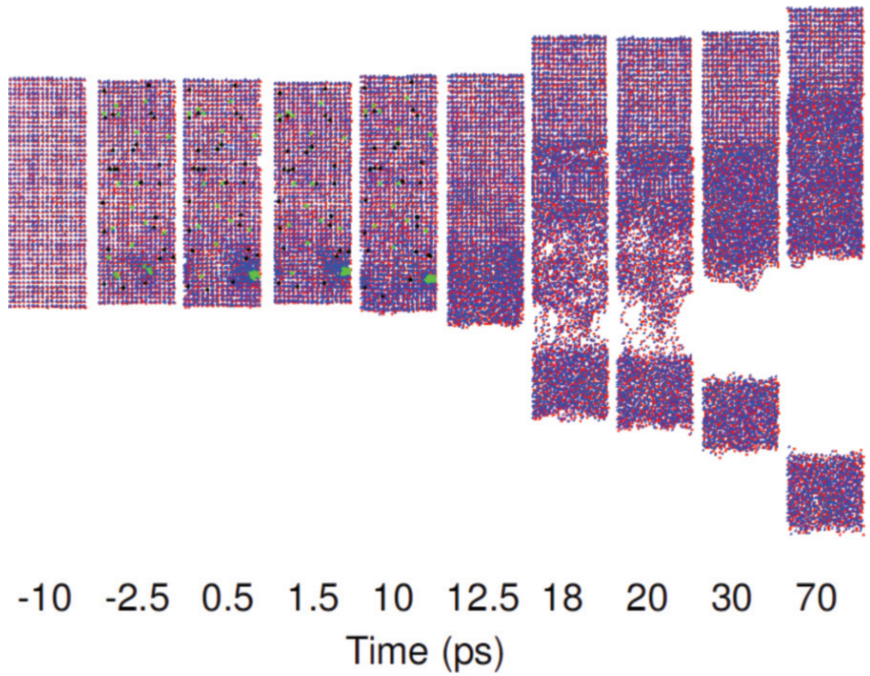




Potentials taken from quantum chemical calculations:
Wang et al, Phys Rev B 68 (2003) 115409

Note: Li^0 is small -> may easily diffuse
high polarizability -> attractive binding





Thermal ablation (no defects)

$F = 38$ mJ / cm^2

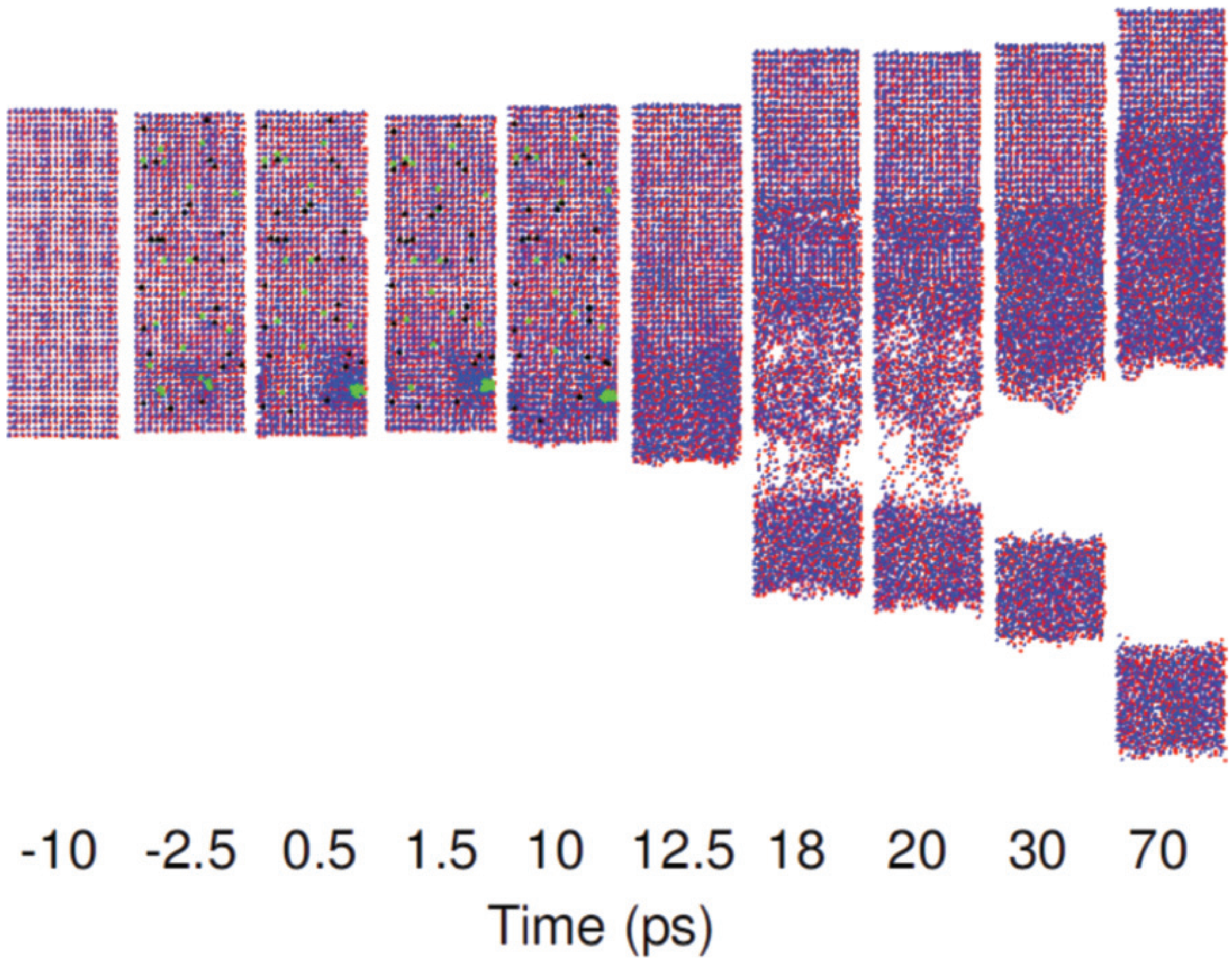
defect-supported ablation (0.45% defects)

$F = 10$ mJ / cm^2

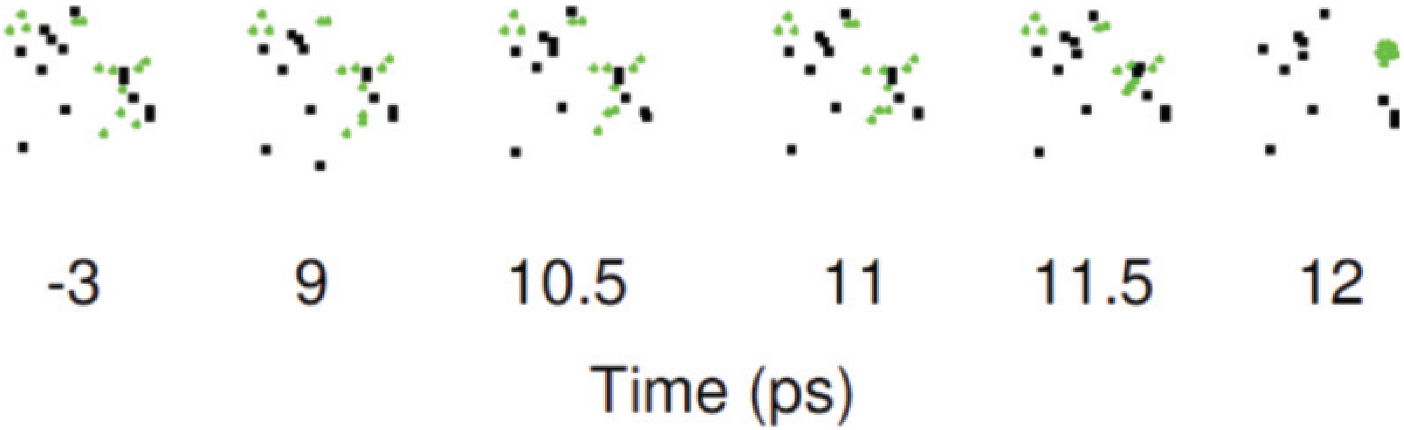
Result: defects lower ablation threshold

- bond weakening
- tensile pressure due to smaller atom radii

agrees with experiment



Green: Li cluster (metallic colloid)
destabilizes lattice



Top view of defects:
black: F^0, green: Li^0

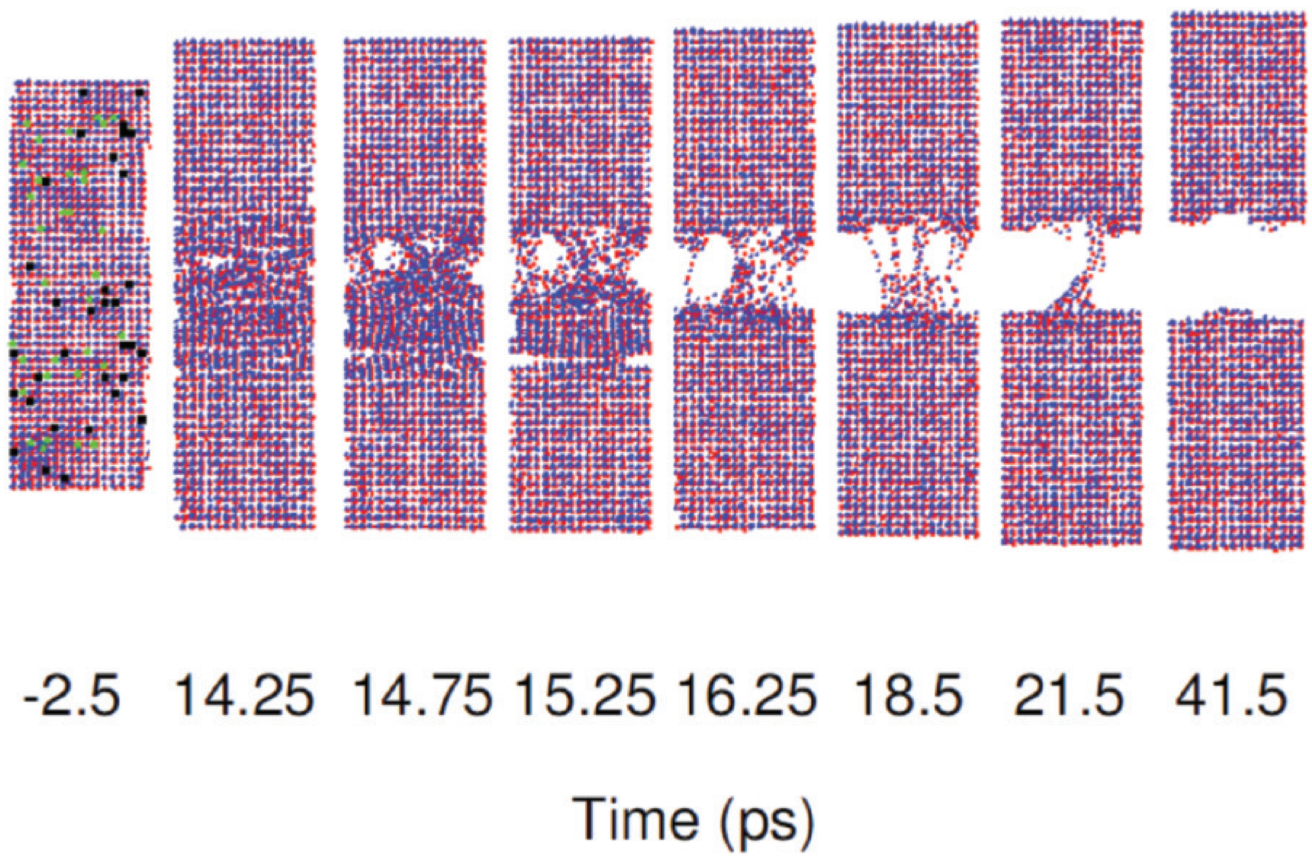
Formation of Li cluster by fast Li^0 diffusion
Condensation heat -> ablation

Experimentally observed under swift-particle irradiation of LiF

„Cold ablation"

Extreme case: assume that laser irradiation
- produces no free electrons (no target heating)
- only produces defects (potential energy)

Here defect concentration 0.57%



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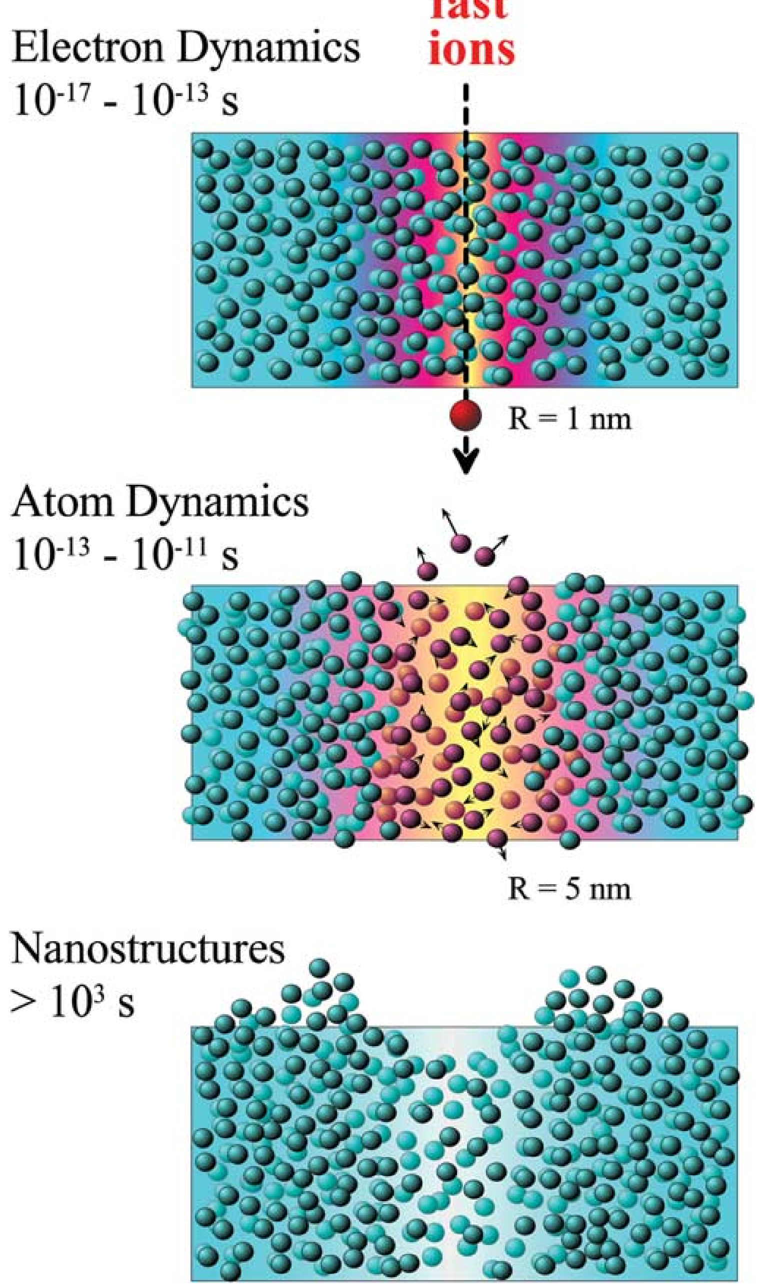
Cherednikov et al., Phys Rev B 87, 245424 (2013)

Swift ions deposit their energy
in the form of electronic excitation
in the target

„Similar physics“ as in
laser irradiation

How to treat system in MD:

After ion passage:
F- -> F+ + 2e- in track cylinder



Schiwietz et al 2004

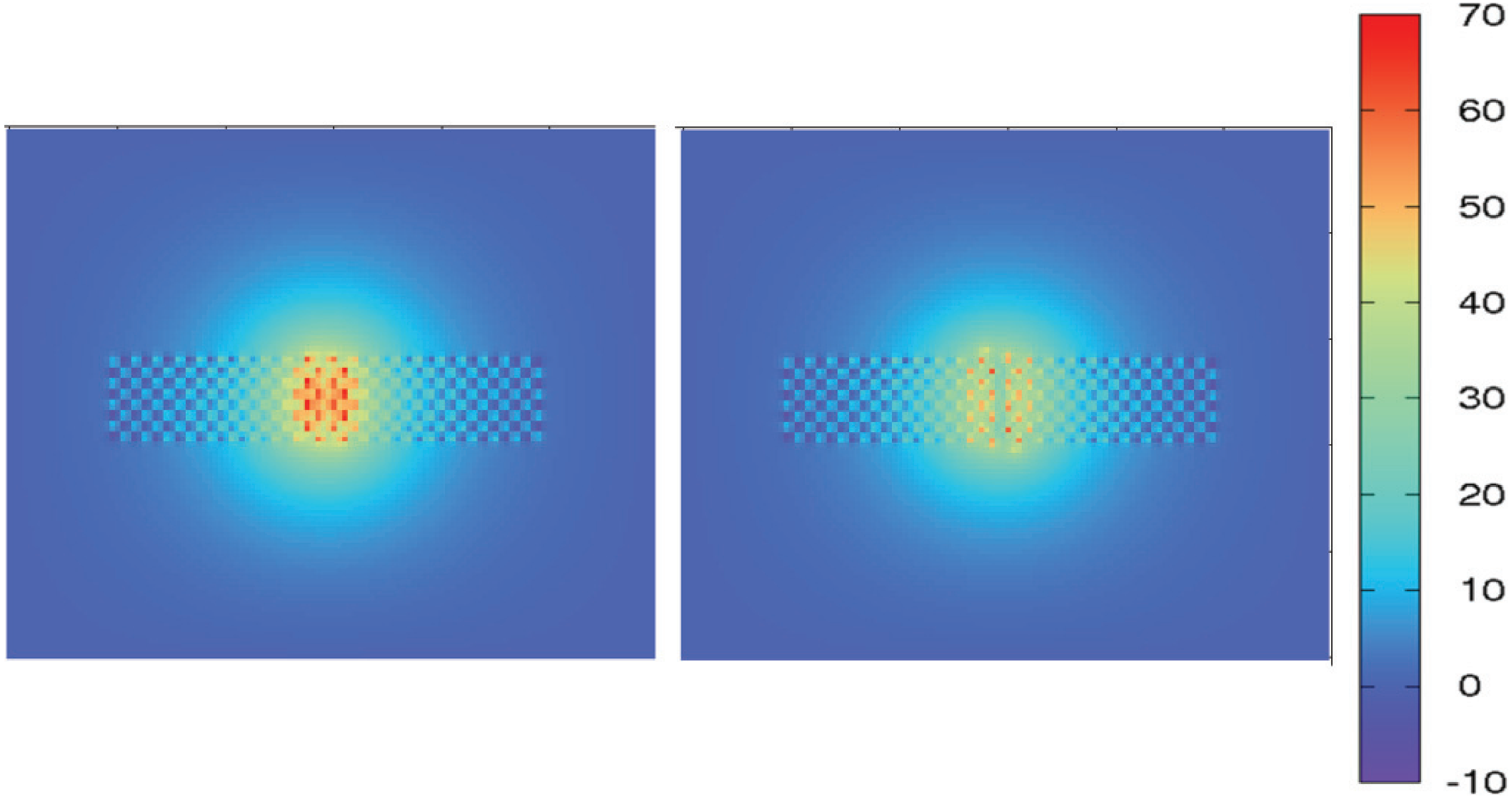
Here:
couple MD with a particle-in-cell (PIC) code for electron dynamics

Equations for electrons:

charge density: $$\rho = e(Z_i n_i - n_e)$$

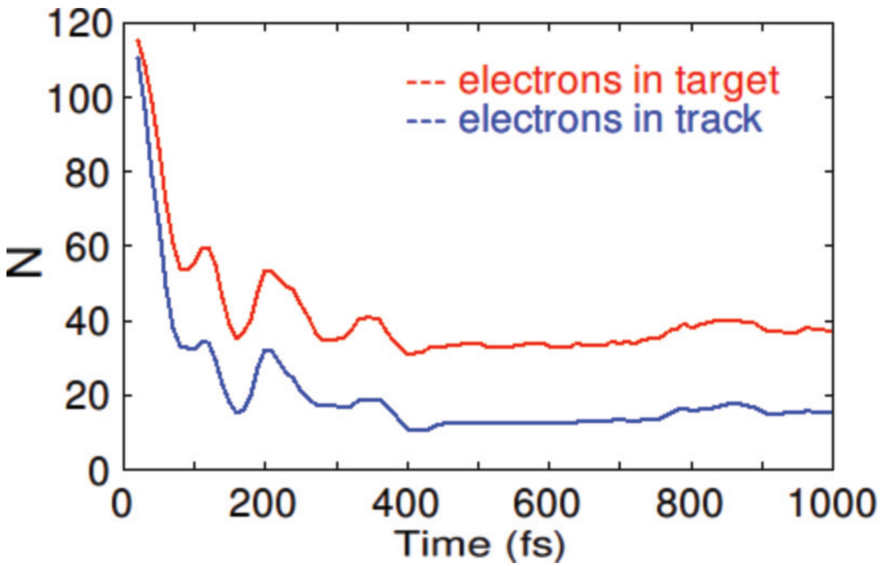
electric potential: $$\nabla^2 \phi = -\frac{\rho}{\varepsilon_0}$$

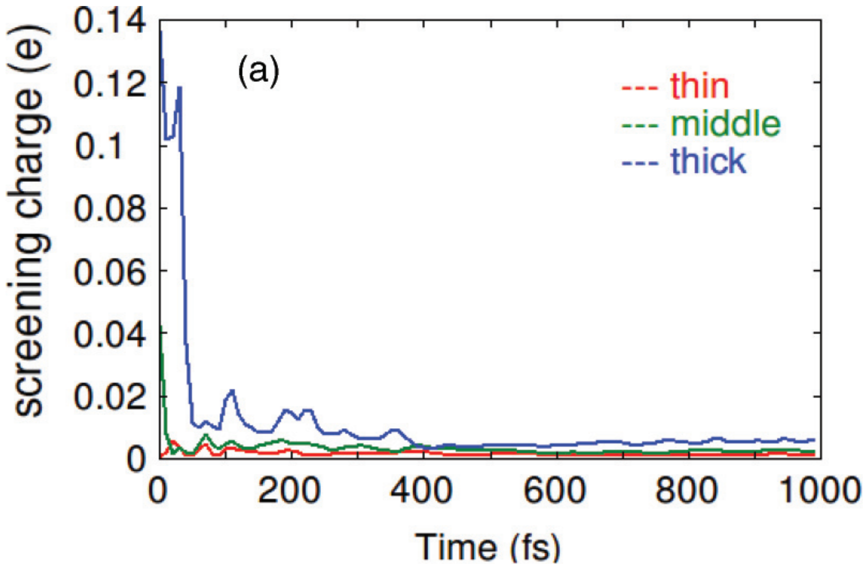
number density: $$n_e = n_0 \exp\left[\frac{e(\phi - \phi_0)}{k_B T_e}\right]$$



Electric potential:
at passage of ion

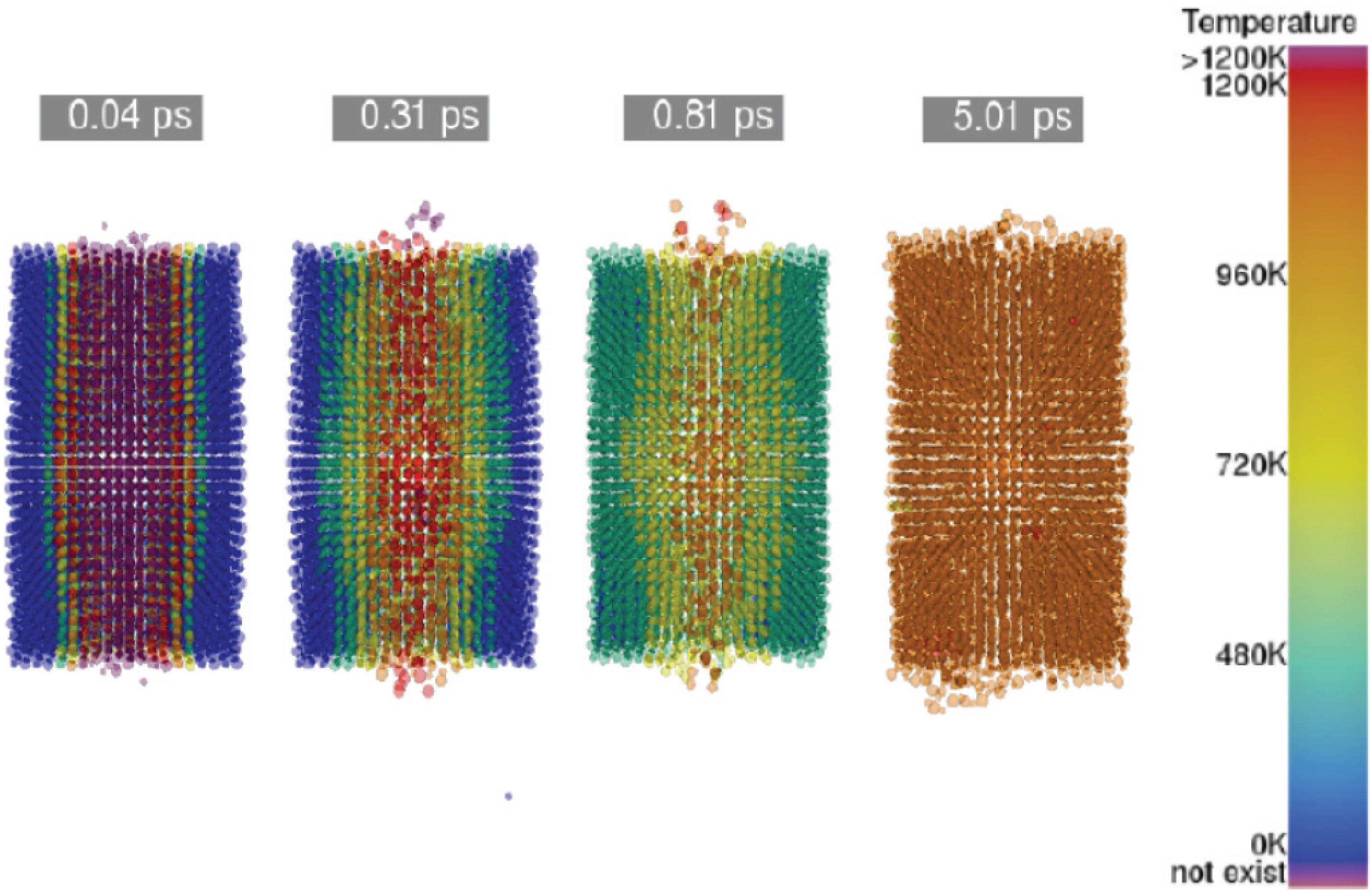
10 fs later





number of electrons remaining in track

shielding of F^+ ions



Evolution of ionization track: sputtering

Conclusions

Two-temperature model for LiF: need for plasma equations for
electron density and energy

- Ablation mechanism similar as in metals: spallation in molten state

- Role of longlived defects
- Defects de-stabilize lattice -> lower ablation threshold
- even „cold" ablation is possible
- role of metallic clusters:
  - form due to high Li^0 diffusion rate
  - destablilize lattice due to condensation heat
- efficient: potential energy introduced by defects small compared to laser energy (or thermal energy of electrons)

Cherednikov et al., J. Opt. Soc. Am. B 28, 1817 (2011)
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