16–21 July 2012

НОЦ

Computer simulation of advanced materials

International School of the European University network

PCAM

MSU Nanotechnology Education and Research Center

Materials in extreme states: from *WARM DENSE MATTER* to laser induced surface nanostructuring

Norman G.E.

JOINT INSTITUTE for HIGH TEMPERATURES RAS

MOSCOW INSTITUTE of PHYSICS and TECHNOLOGY

ОБЪЕДИНЕННЫЙ ИВТАН

МФТИ

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JIHT junior researcher
born **1988**

Contents

Warm dense matter (WDM), examples

Two temperature WDM. ***Exotic properties***

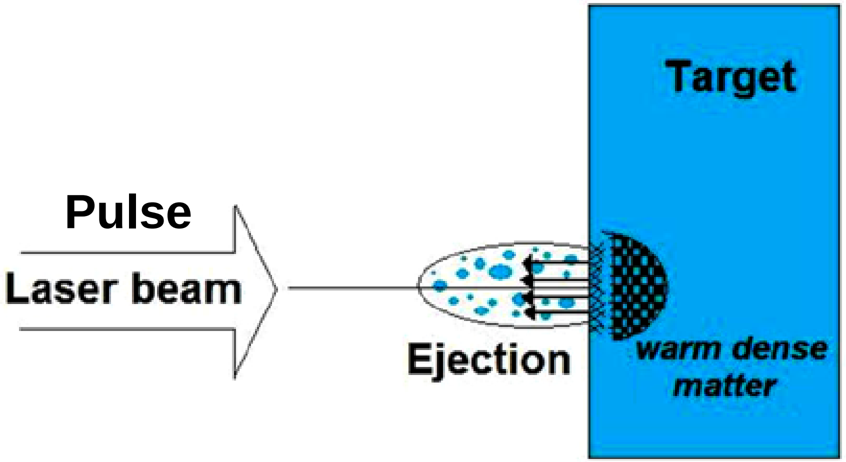
Basic equations for WDM relaxation

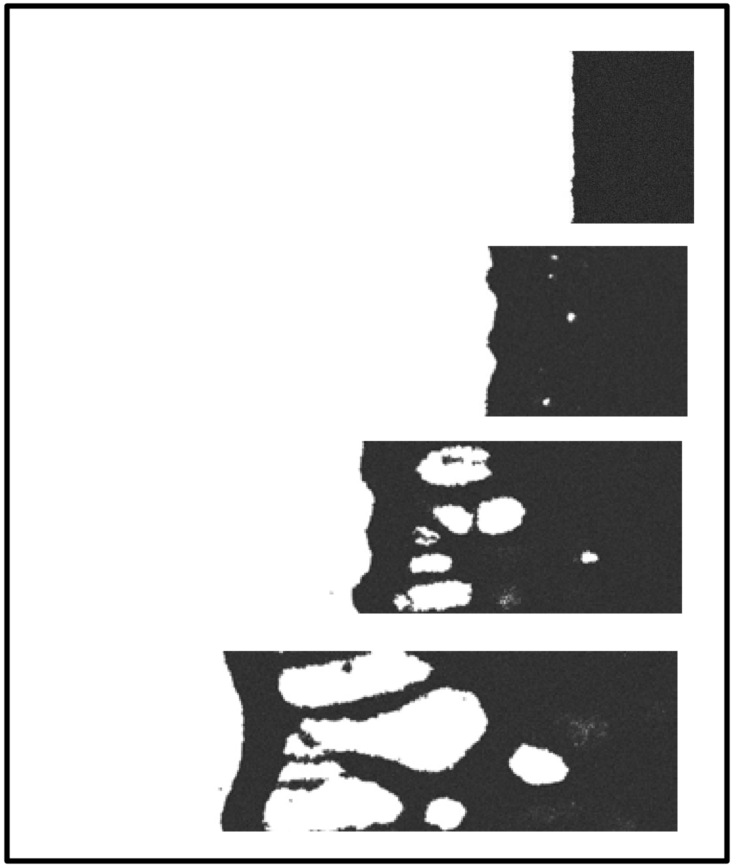
Electron-temperature dependent
interionic potential

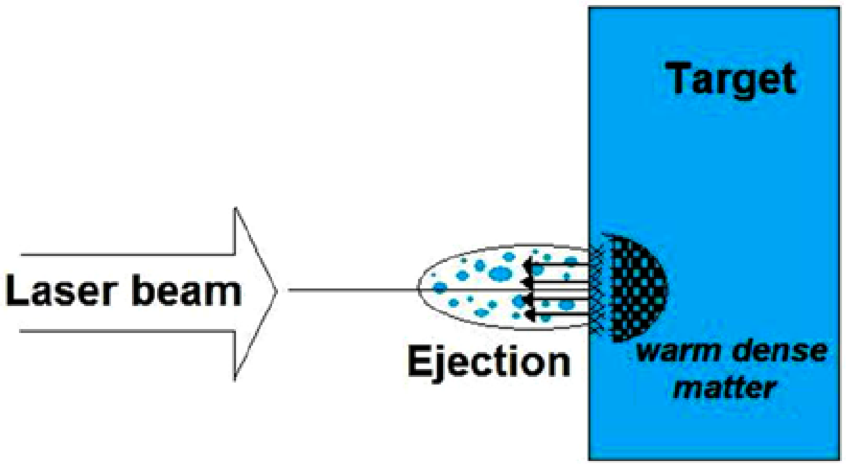
Example 1: Laser ablation of gold

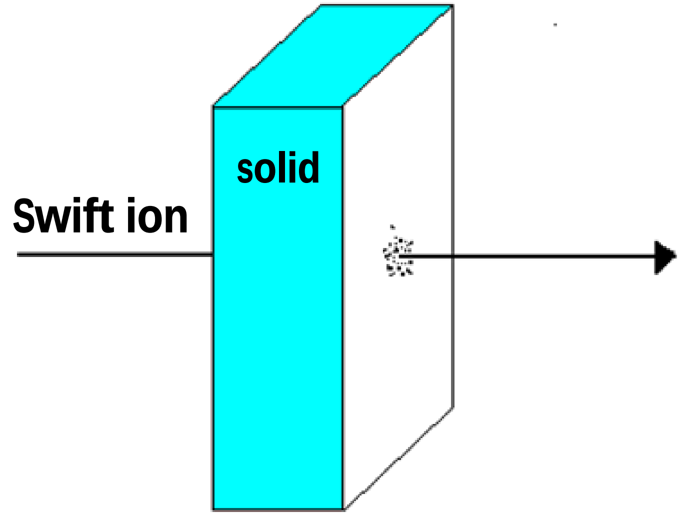
Example 2: Tracks formation in metals

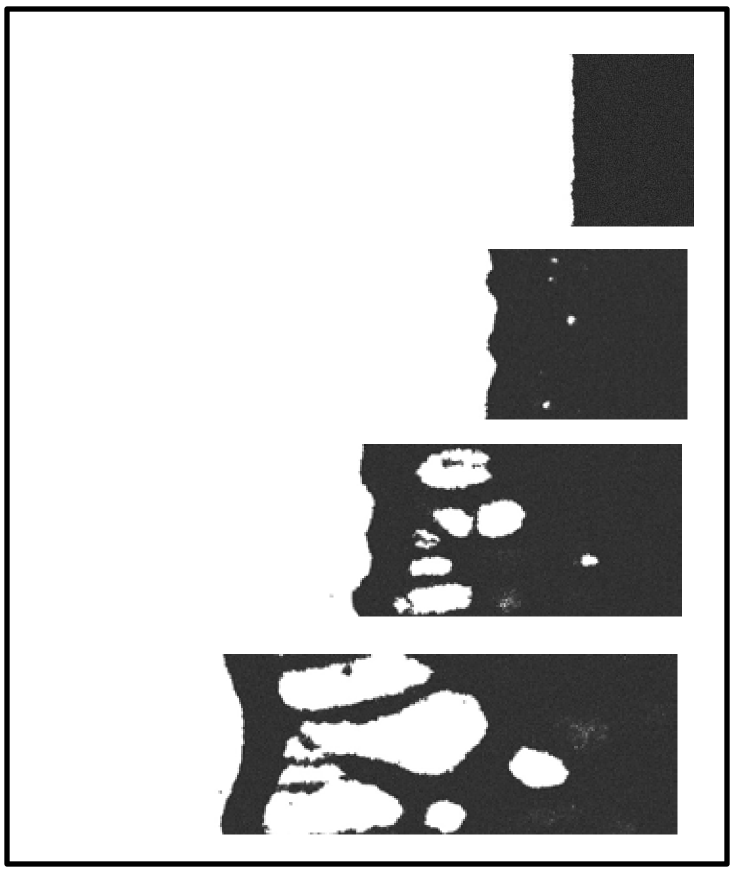
Warm dense matter
Examples

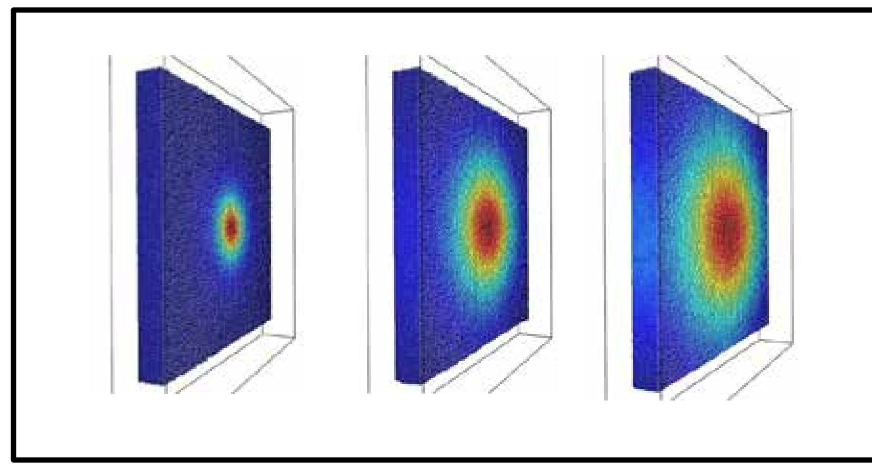

Target
Pulse
Laser beam
Ejection
warm dense
matter



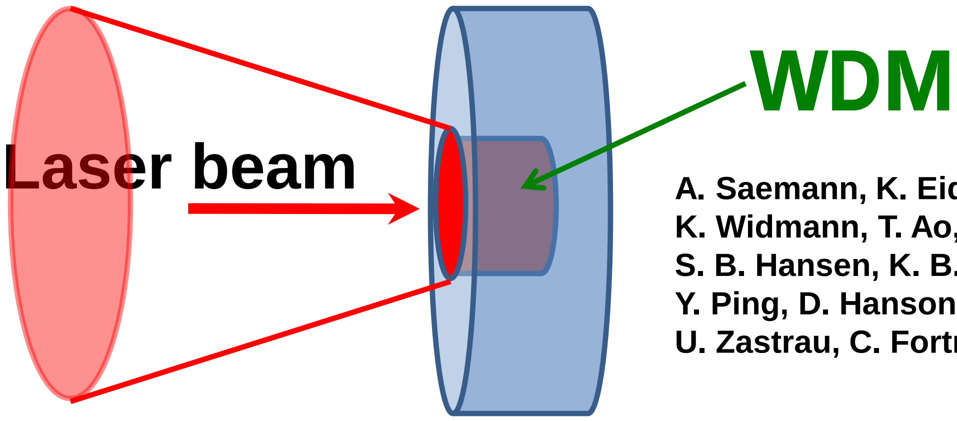

Target
Laser beam
Ejection
warm dense
matter


solid
Swift ion



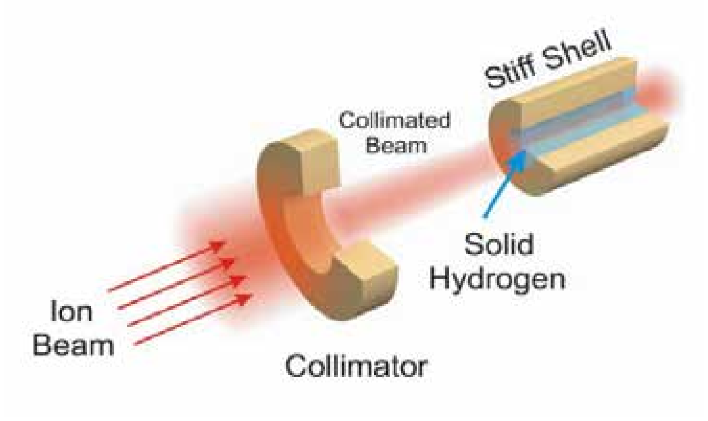


Diverse sources of generation:



IR, visible, UV, XUV lasers

A. Saemann, K. Eidmann, I. E. Golovkin, et al, PRL 82, 4843 (1999)
K. Widmann, T. Ao, M. E. Foord, et al, PRL 92, 125002 (2004)
S. B. Hansen, K. B. Fournier, A.Ya. Faenov, et al, PRE 72, 036408 (2005)
Y. Ping, D. Hanson, I. Koslow, et al, PRL 96, 255003 (2006)
U. Zastrau, C. Fortmann, R. R. Fäustlin, et al, PRE 78, 066406 (2008)



fast single ions, ion beams in condensed matter

A.V.Lankin, I.V.Morozov, G.E.Norman, S.A. Pikuz Jr., I.Yu.Skobelev Phys. Rev. E 79, 036407 (2009)

Nanosecond Electric Explosion of Wires

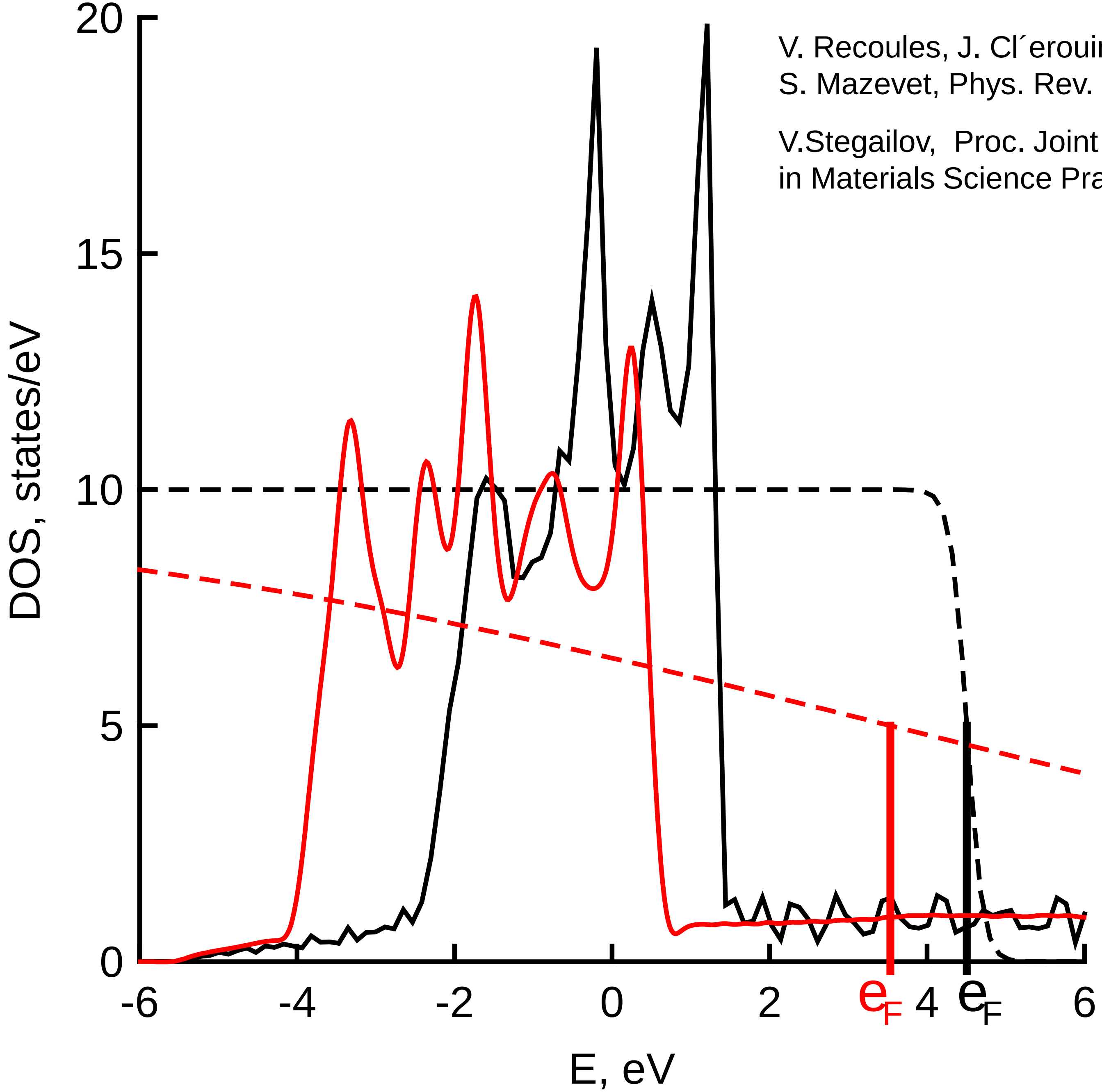
G.E. Norman, V.V. Stegailov, A.A. Valuev. Contrib. Plasma Phys. 43, 384 (2003)

Explosions of tips (protrusion, whisker) on cathode surface

Two temperature warm dense matter.

Exotic properties

Gold: electron DOS (d- and s-electrons)



V. Recoules, J. Cl´erouin, G. Z´erah, P. M. Anglade, S. Mazevet, Phys. Rev. Lett. 96, 55503 (2006)

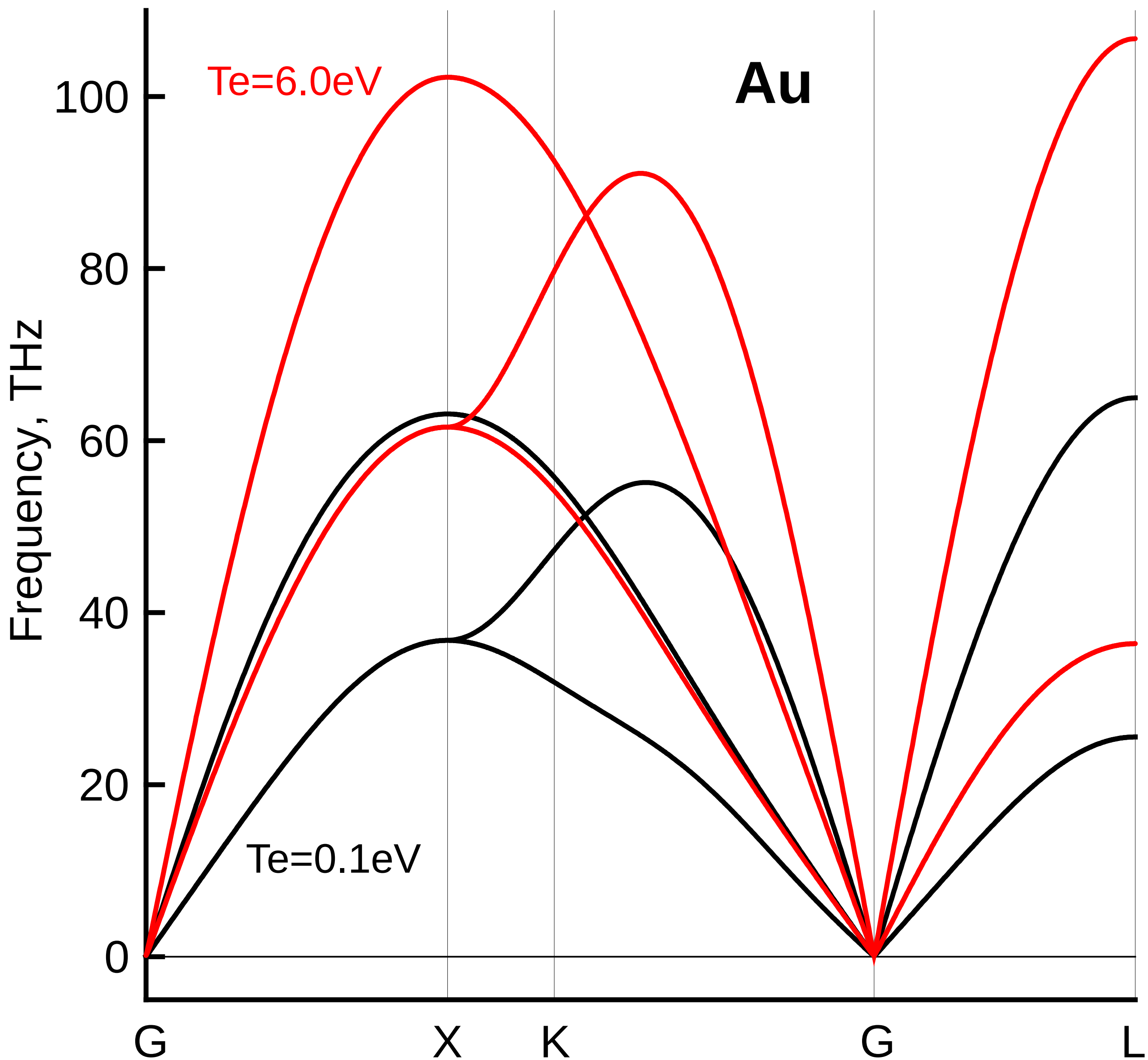
V.Stegailov, Proc. Joint U.S. Russia Conf. on Advances in Materials Science Prague, August 2009

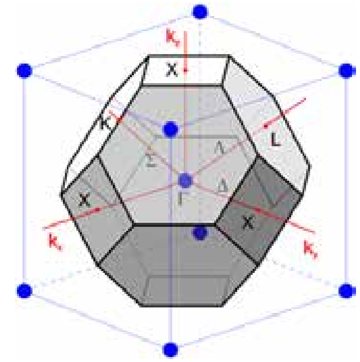
Au = [Xe] 4f[14] **5d[10] 6s[1]**

Te=0.1eV

Te=6.0eV

FCC gold phonon spectrum after pulse irradiation



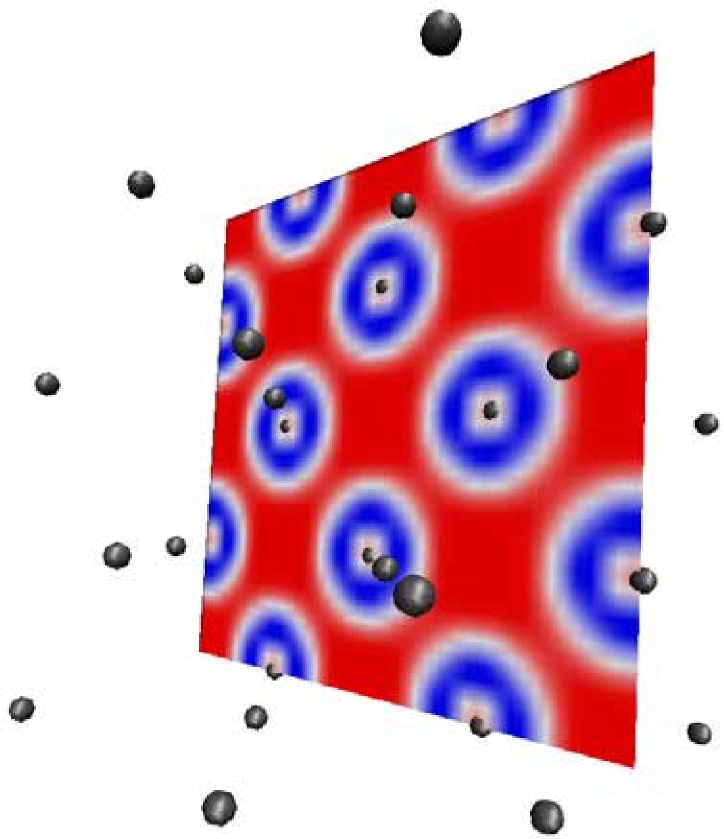


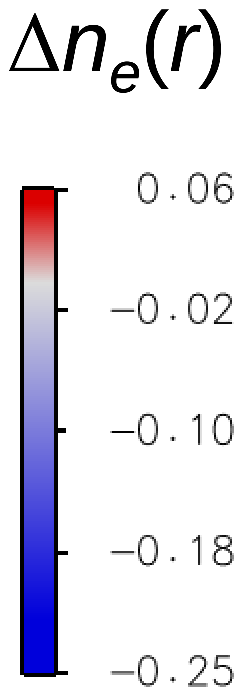
V.Recoules, J.Cl´erouin, G.Z´erah,
P.Anglade, S.Mazevet,
Phys. Rev. Lett. 96, 55503 (2006)

V.Stegailov, Proc. Joint
U.S. Russia Conf. on
Advances in Materials Science,
Prague, August 2009

Spatial redistribution of the electron density after the electron temperature increase

f.c.c. Au





$$\Delta n_e(r) = n_e(r)\big|_{T_e=6eV} - n_e(r)\big|_{T_e=0.1eV}$$

Basic equations
for WDM relaxation

MD simulation for *IONS* :

ETD-potential + Langevin thermostat + P_e

$$m\frac{d\vec{v}_i}{dt} = \vec{F}_i(T_e) - \beta\vec{v}_i + \vec{\xi}(T_e) - \frac{\nabla P_e^{deloc}}{\rho_{ion}}$$

Electron-ion relaxation
Langevin thermostat

Electron-temperature dependent
interionic potential

Electronic pressure
(delocalized electron energy)

Thermal conductivity equation for *ELECTRONS* at continuum media approximation

$$C_e \frac{\partial T_e}{\partial t} = \nabla\left(K_e \nabla T_e\right) - g_p\left(T_e - T_i\right) + \nabla Q$$

Electron thermal conductivity

Electron-ion relaxation

Source of heating at ablation

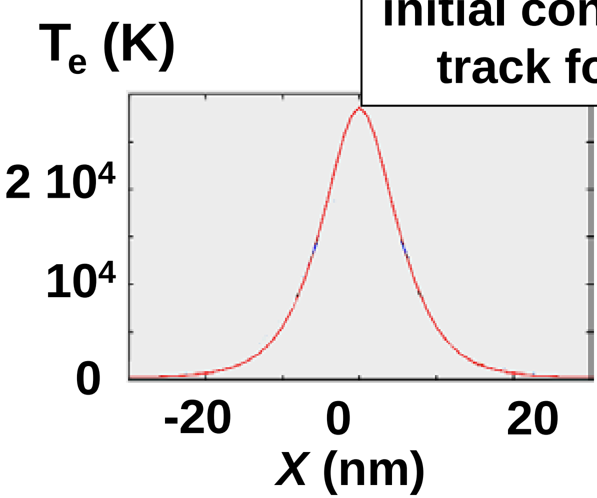
Thermal conductivity equation for *ELECTRONS* at continuum media approximation

$$C_e \frac{\partial T_e}{\partial t} = \nabla\left(K_e \nabla T_e\right) - g_p\left(T_e - T_i\right) + \nabla Q$$

Electron thermal conductivity

Electron-ion relaxation

Source of heating at ablation



initial conditions for track formation

$$Q = I_0 \cdot \exp[-x/d] \qquad t < \text{pulse width}$$
$$Q = 0 \qquad t \geq \text{pulse width}$$

laser ablation

Complete model

ELECTRONS: thermal conductivity equation at continuum media

MD simulation for *IONS*: ETD-potential + Langevin thermostat + P_e

modification of LAMMPS

$$C_e \frac{\partial T_e}{\partial t} = \nabla\left(K_e \nabla T_e\right) - \boxed{g_p\left(T_e - T_i\right)} + \nabla Q$$

$$m\frac{d\vec{v}_i}{dt} = \vec{F}_i\left(T_e\right) - \boxed{\beta\vec{v}_i + \vec{\xi}\left(T_e\right)} - \frac{\nabla P_e^{deloc}}{\rho_{ion}}$$

$$Q = I_0 \cdot \exp[-x/d] \qquad t < \text{pulse width}$$

$$Q = 0 \qquad t \geq \text{pulse width}$$

laser ablation

initial distribution of T_e

track formation

g_p – factor of electron-ion relaxation

C_e – electron heat capacity

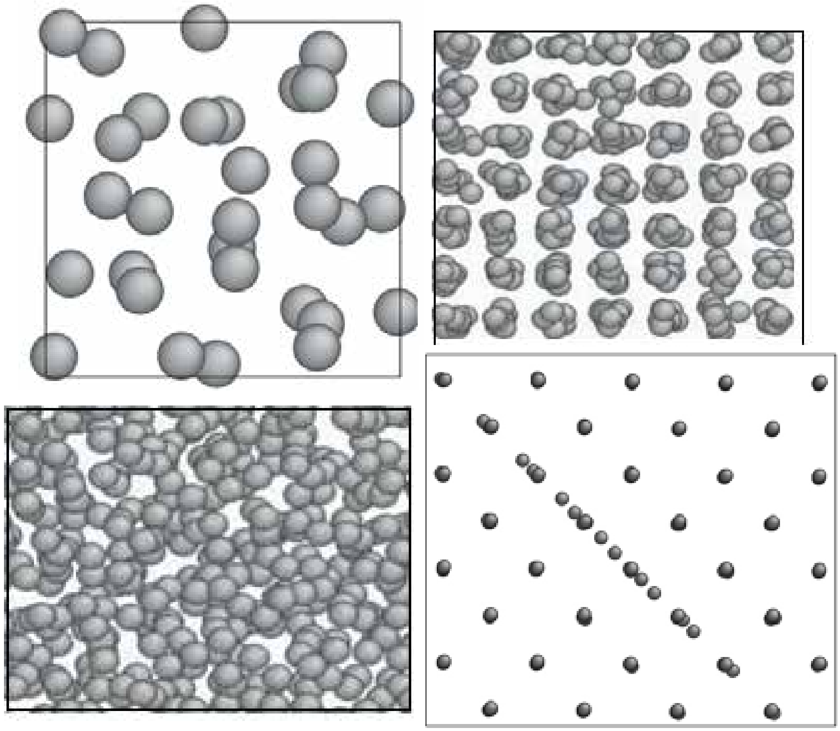
K_e – electron heat conductivity

$$\beta = \beta(g_p, C_e) \qquad \xi \sim T_e^{1/2}$$

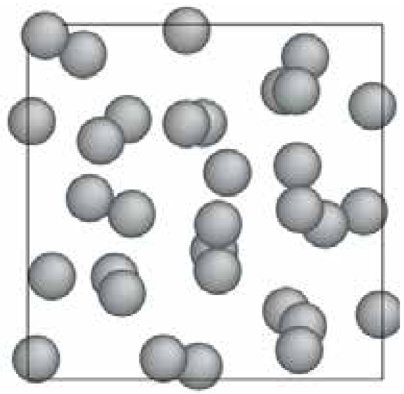
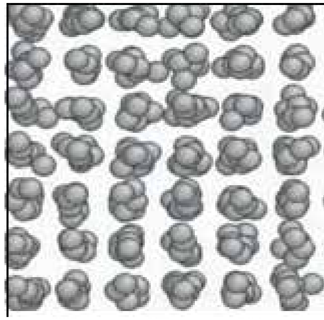
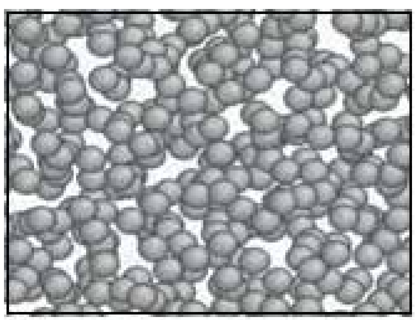
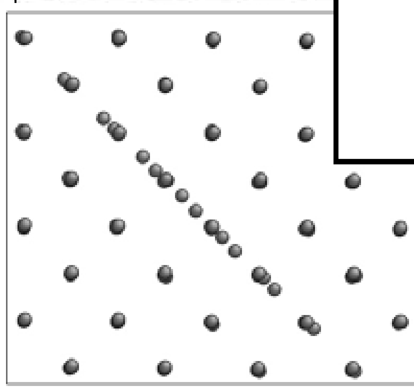
$$\tau = \frac{m}{\beta} \qquad \tau \sim 20 \text{ ps}$$

Electron-temperature dependent interionic potential

Force matching method for development of potential



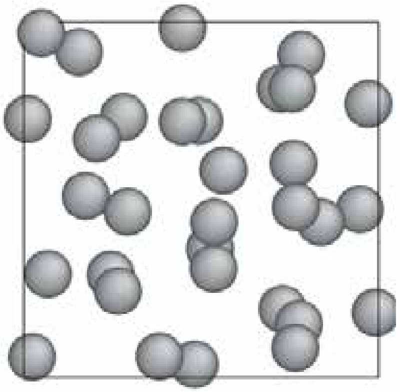
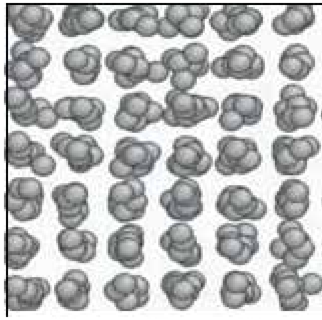
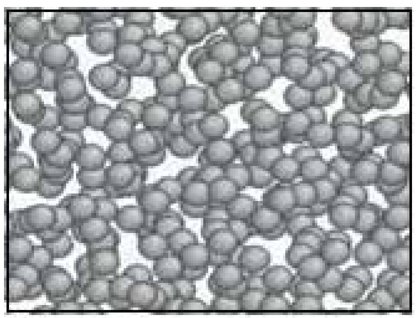
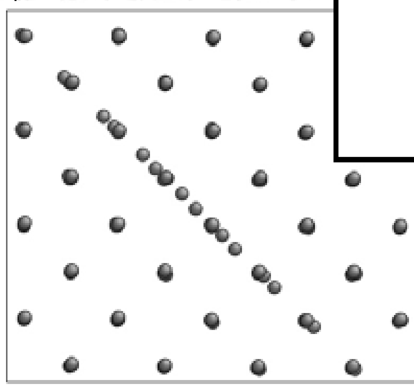
Force matching method for development of potential

$$E = \frac{\langle \varphi_n | \mathbf{H} | \varphi_n \rangle}{\langle \varphi_n | \varphi_n \rangle}$$

$$\mathbf{F}_i = \frac{\partial E}{\partial \mathbf{R}_i}$$

Force matching method for development of potential

$$E = \frac{\langle \varphi_n | \mathbf{H} | \varphi_n \rangle}{\langle \varphi_n | \varphi_n \rangle}$$

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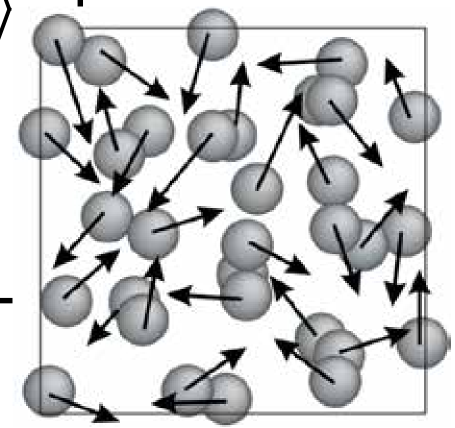
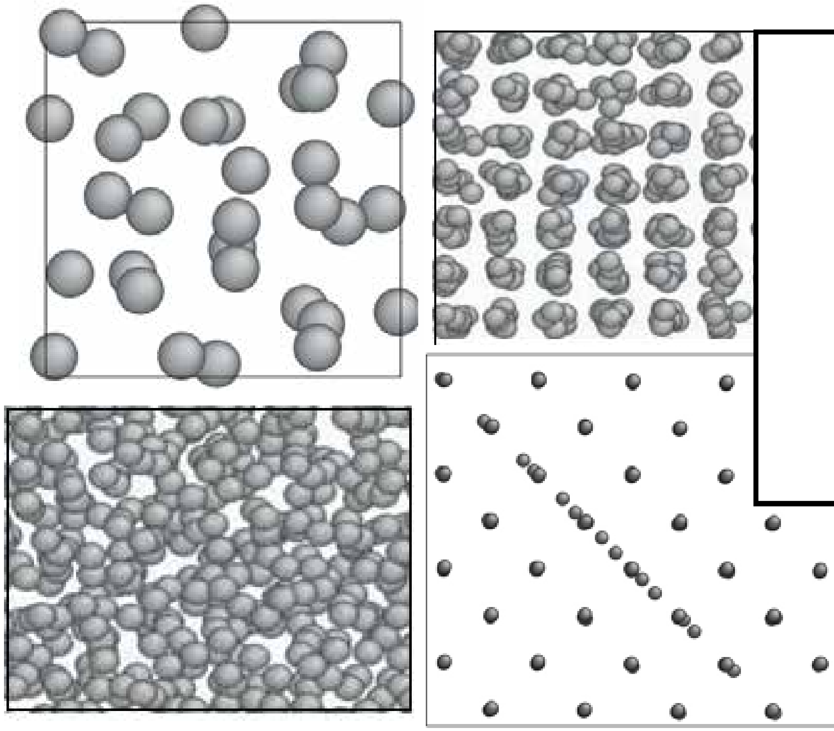


Force matching method for development of potential



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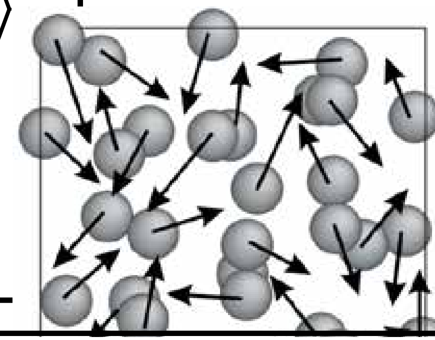
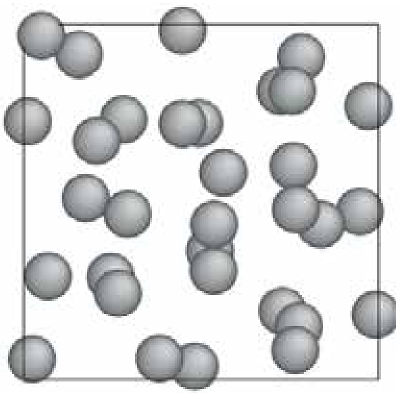


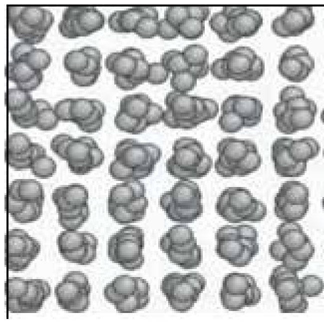
$$U = \sum_{i,j<i} \phi_{ij}(r_{ij}) + \sum_i F(\rho_i),$$

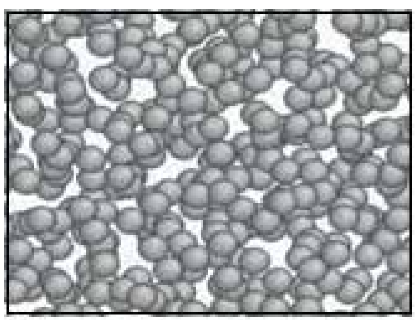
$$\rho_i = \sum_{j \neq i} \rho(r_{ij})$$

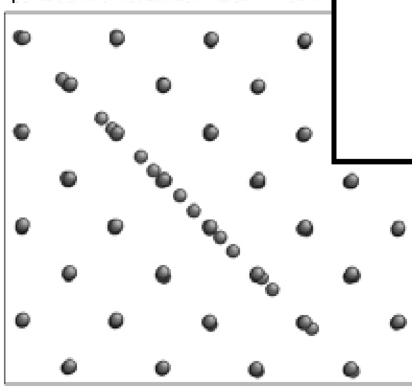
(EAM)

Force matching method for development of potential









$$E = \frac{\langle \varphi_n | \mathbf{H} | \varphi_n \rangle}{\langle \varphi_n | \varphi_n \rangle}$$

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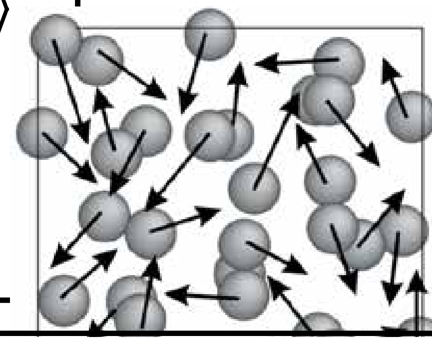


$$U = \sum_{i,j<i} \phi_{ij}(r_{ij}) + \sum_i F(\rho_i),$$

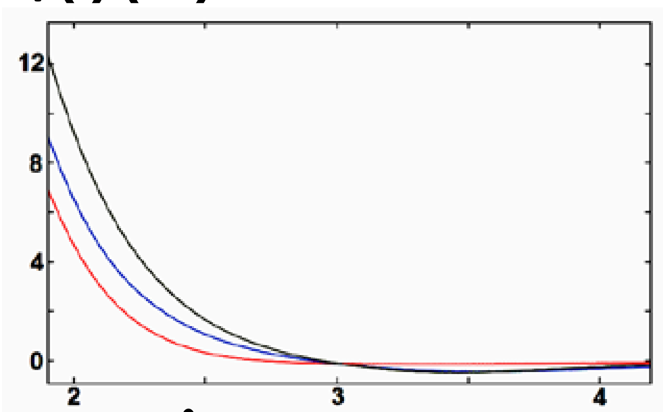
$$\rho_i = \sum_{j \neq i} \rho(r_{ij})$$

(EAM)

$$Z = \sum \frac{(f_{quantum} - f_{EAM})^2}{f^2_{quantum}}$$

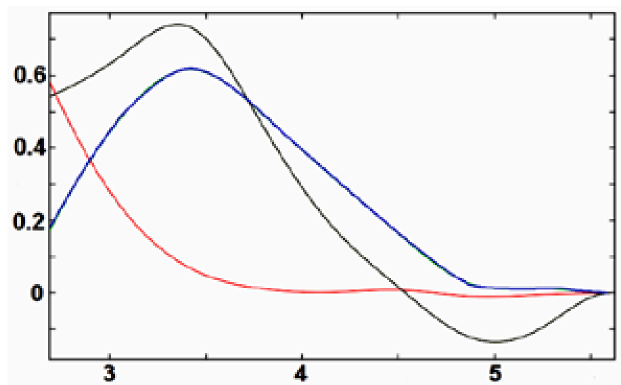
ETD-potential for gold

φ(r) (eV)



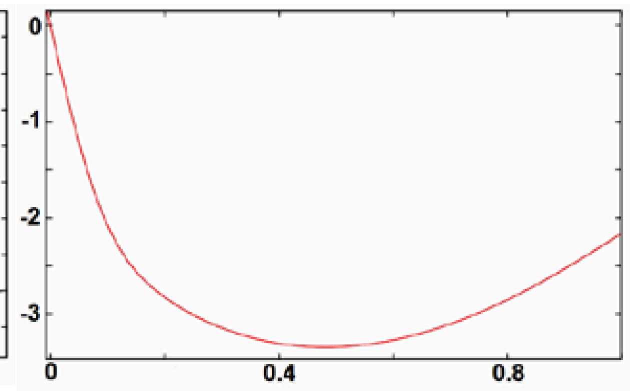
r (Å)

ρ(r)



r (Å)

F (eV)



ρ

$$U = \sum_{i,j<i} \phi_{ij}(r_{ij}) + \sum_i F(\rho_i),$$

$$\rho_i = \sum_{j \neq i} \rho(r_{ij})$$

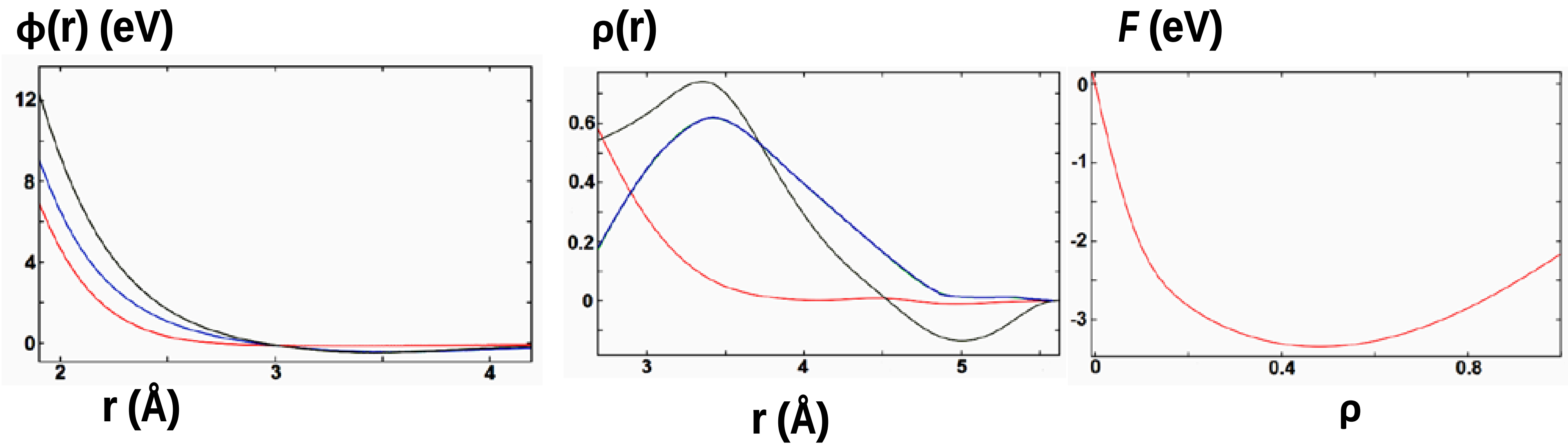
(EAM)

T_e=0.1 eV

T_e=3 eV

T_e=6 eV

ETD-potential for gold



$$E = \sum_{j}^{N} \left(\varphi_0(r_j) + \varphi_1(r_j) \cdot T_e + \varphi_2(r_j) \cdot T_e^{\,2} \right) +$$

$$+ F\left(\sum_{j}^{N} \left\{ \rho_0(r_j) + \rho_1(r_j) \cdot T_e + \rho_2(r_j) \cdot T_e^{\,2} \right\} \right)$$

T_e=0.1 eV

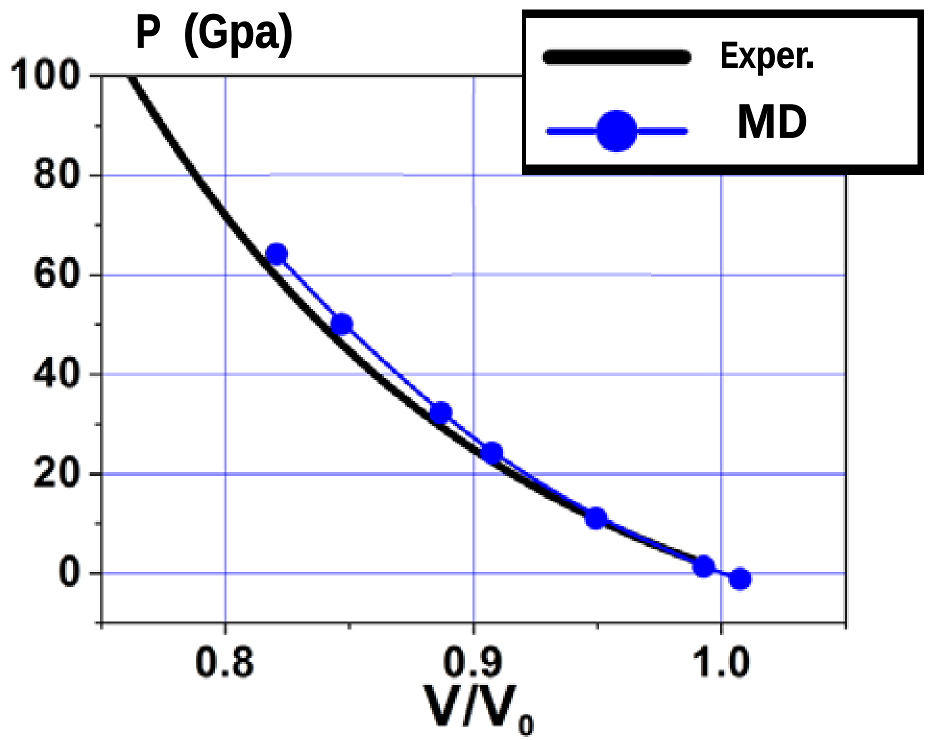
T_e=3 eV

T_e=6 eV

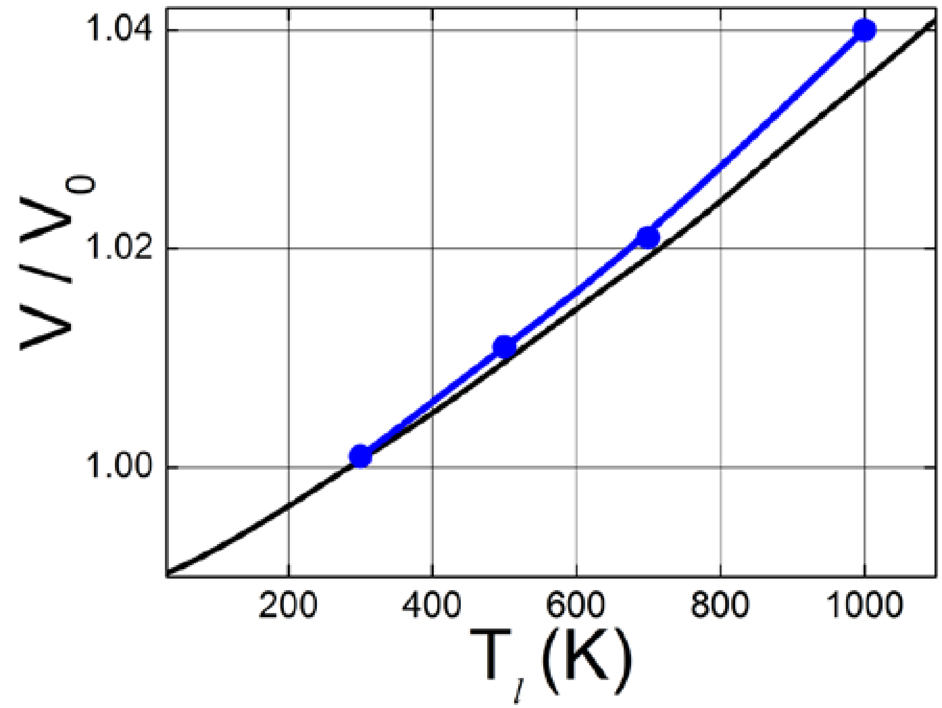
Starikov S.V., Stegailov V.V., Norman G.E. et al. JETP Lett., V. 93, pp. 642-647 (2011)

Норман Г.Э., Стариков С.В., Стегайлов В.В. ЖЭТФ, том 141, выпуск 5, С. 910-917 (2012)

Verification of ETD-potential for Au at T_e = 0.05 eV



Cold curve Au



Thermal expansion Au

	V_0 , Å^3	E_c , eV	C_{11} , GPa	C_{12} , GPa	T_{melt} , K
experiment	10.22	3.8	202	170	1338
MD	10.23	4.1	225	180	1210

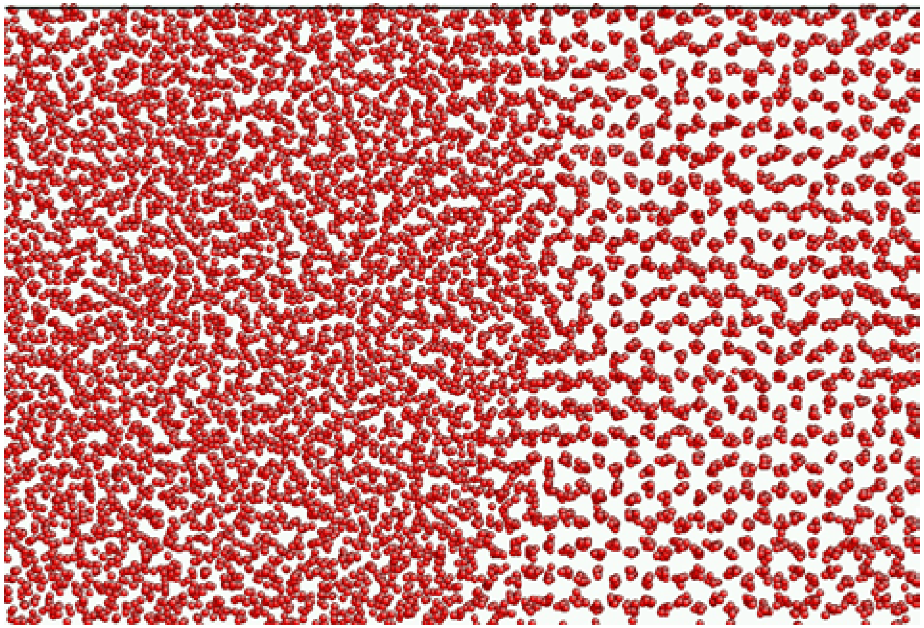
Melting curve calculation, **Fe**
Two-phase 3D modeling

$$E_{conf} = \sum_i E_i$$

$$E_i = \frac{1}{2}\sum_{j \neq i}^{N} \varphi(r_{ij}) + F(\rho_i)$$

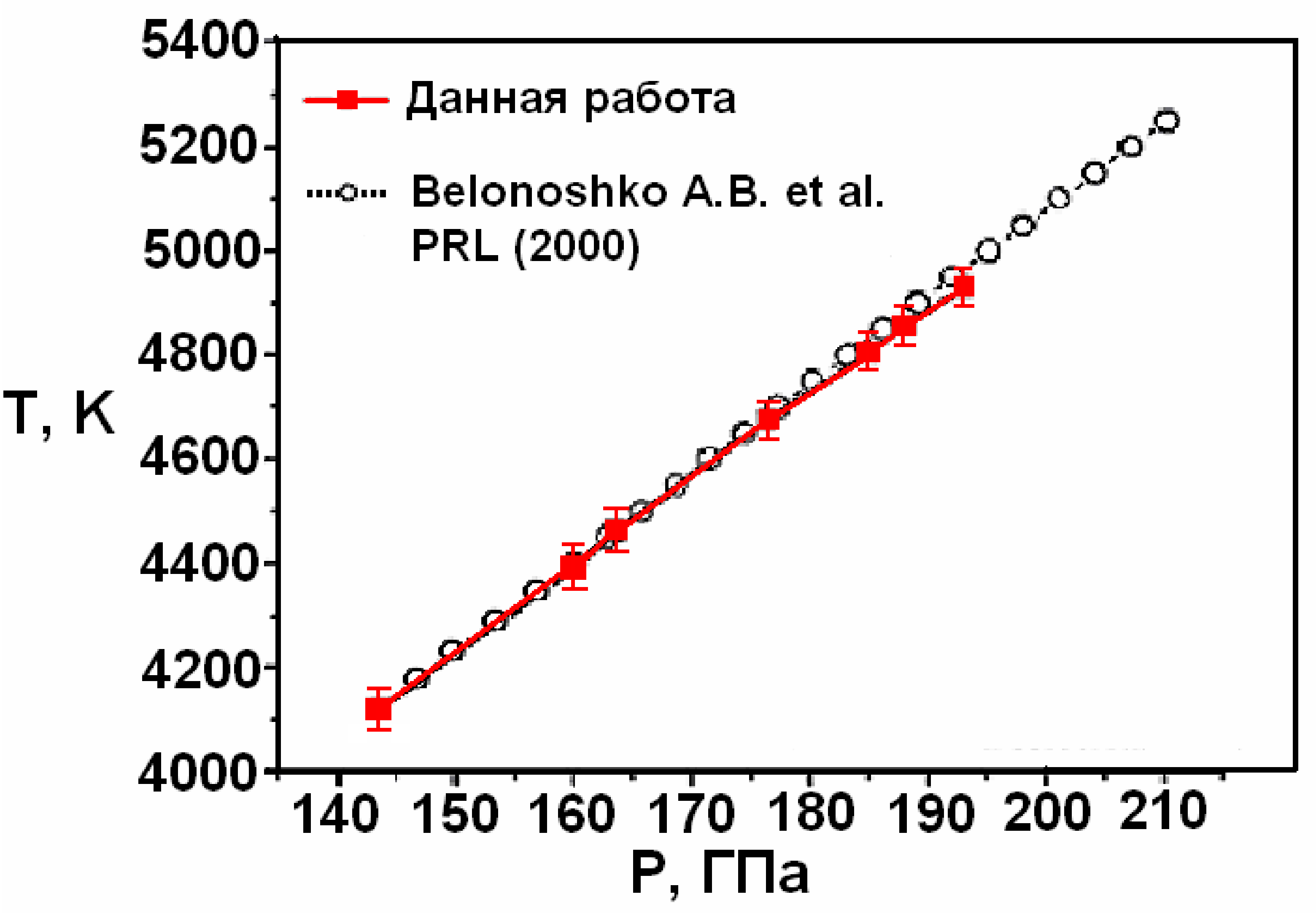
$$\rho_i = \sum_{j \neq i}^{N} \rho(r_{ij})$$

EAM potential

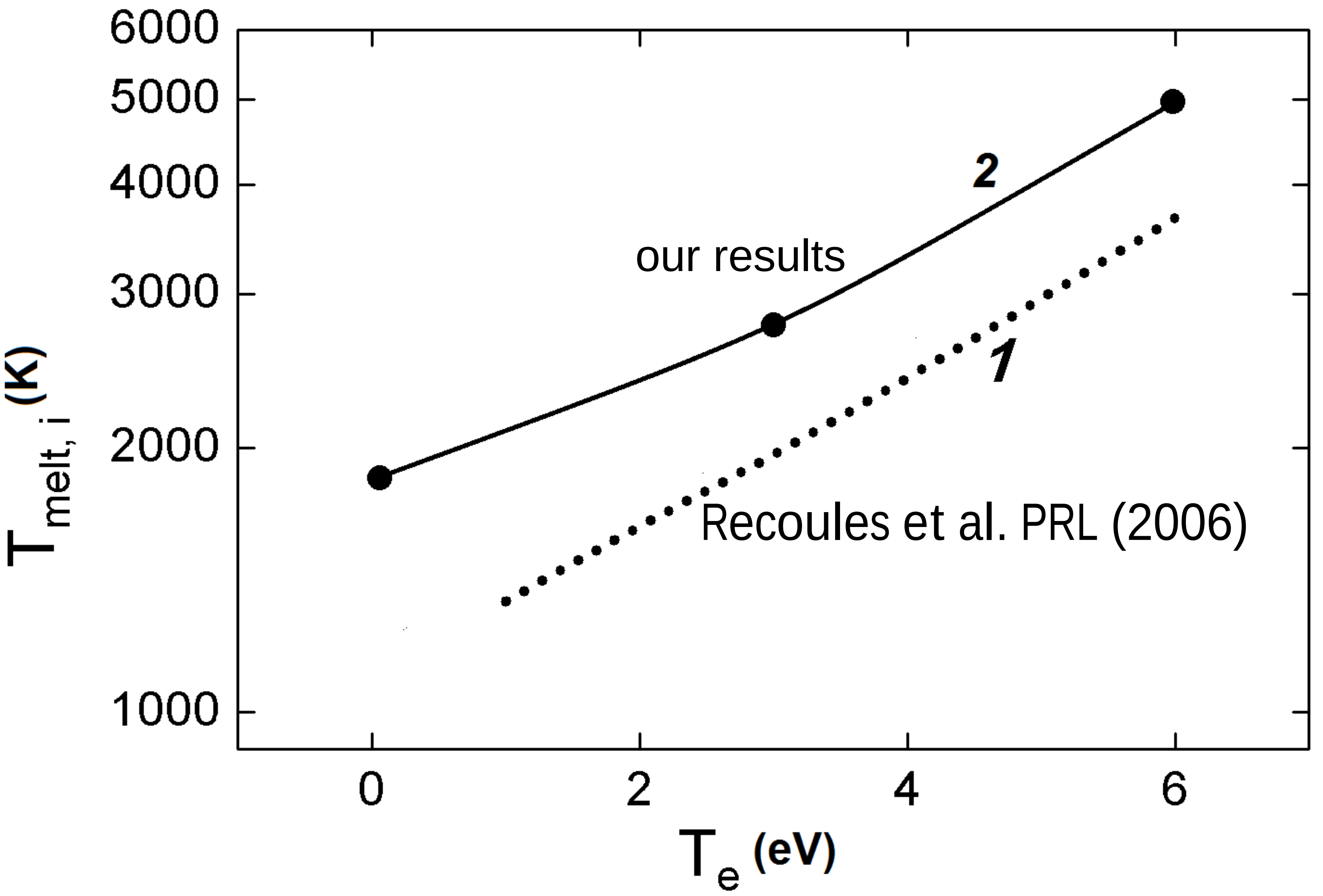


Projection onto xy plane

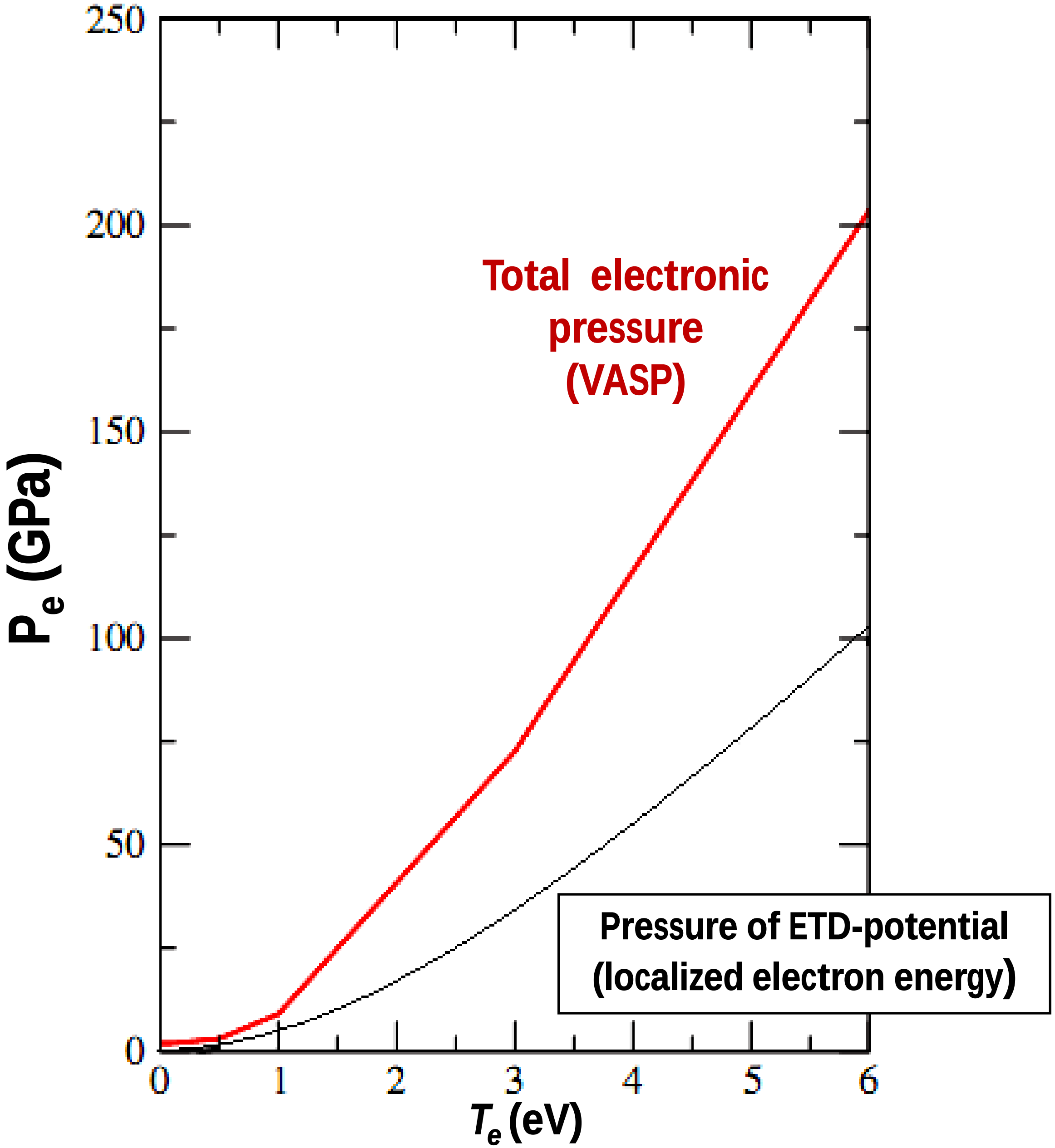
Melting curve $T_m(P)$ for Fe



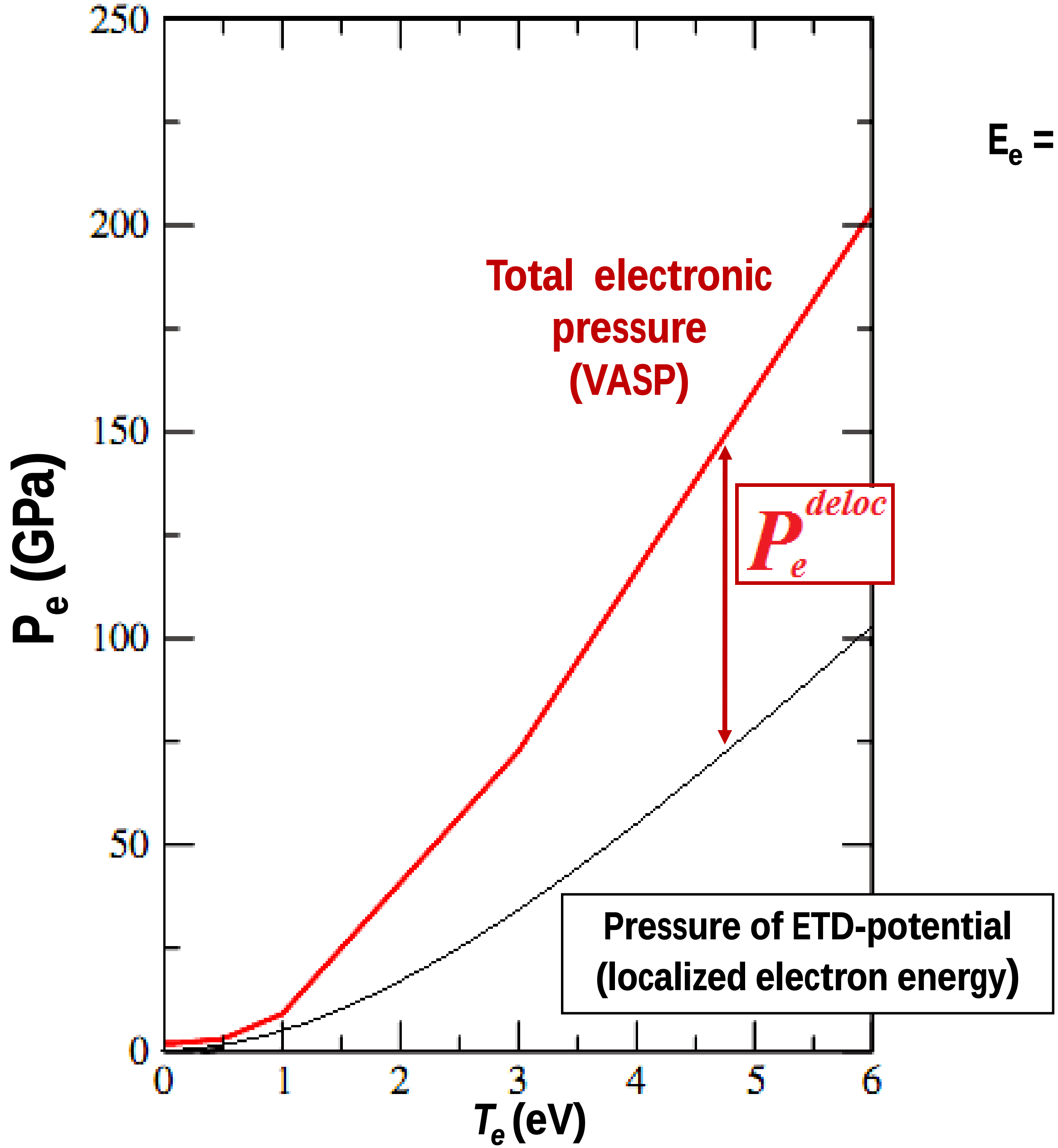
T_e-dependence of **Au** crystal melting temperature



Electronic pressure in gold



Electronic pressure in gold



$E_e = E(R_1 \ldots R_i \ldots R_N, T_e) + E(\text{volume}, T_e)$

Like gas of free-electrons

$$P_e^{\,deloc} = A \cdot T_e^{\,2}$$

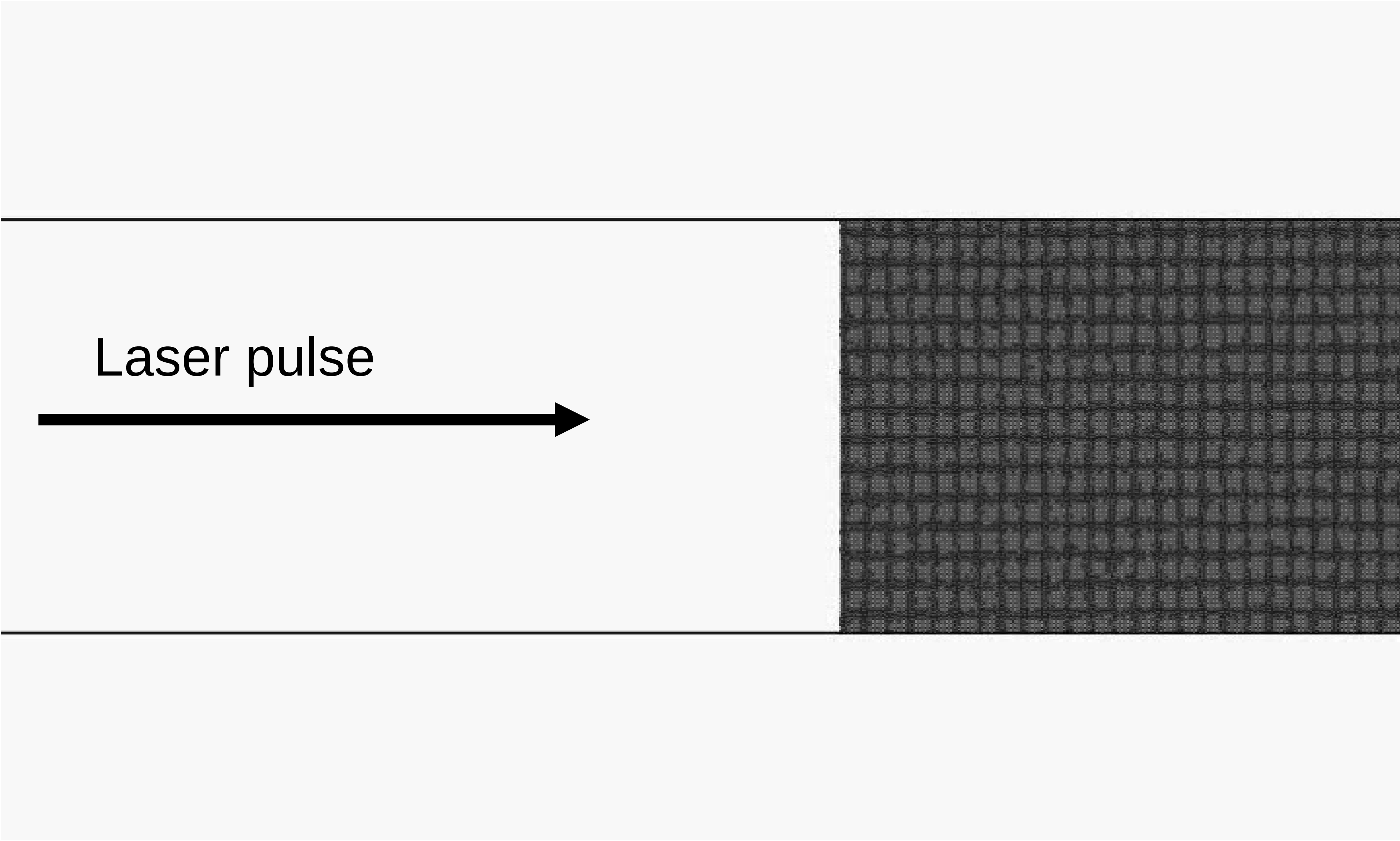
$$\mathbf{B} = \frac{\tilde{N} P_e^{deloc}}{r_{ion}}$$

Electron blast force

[Gan, Chen // APL (2009)]

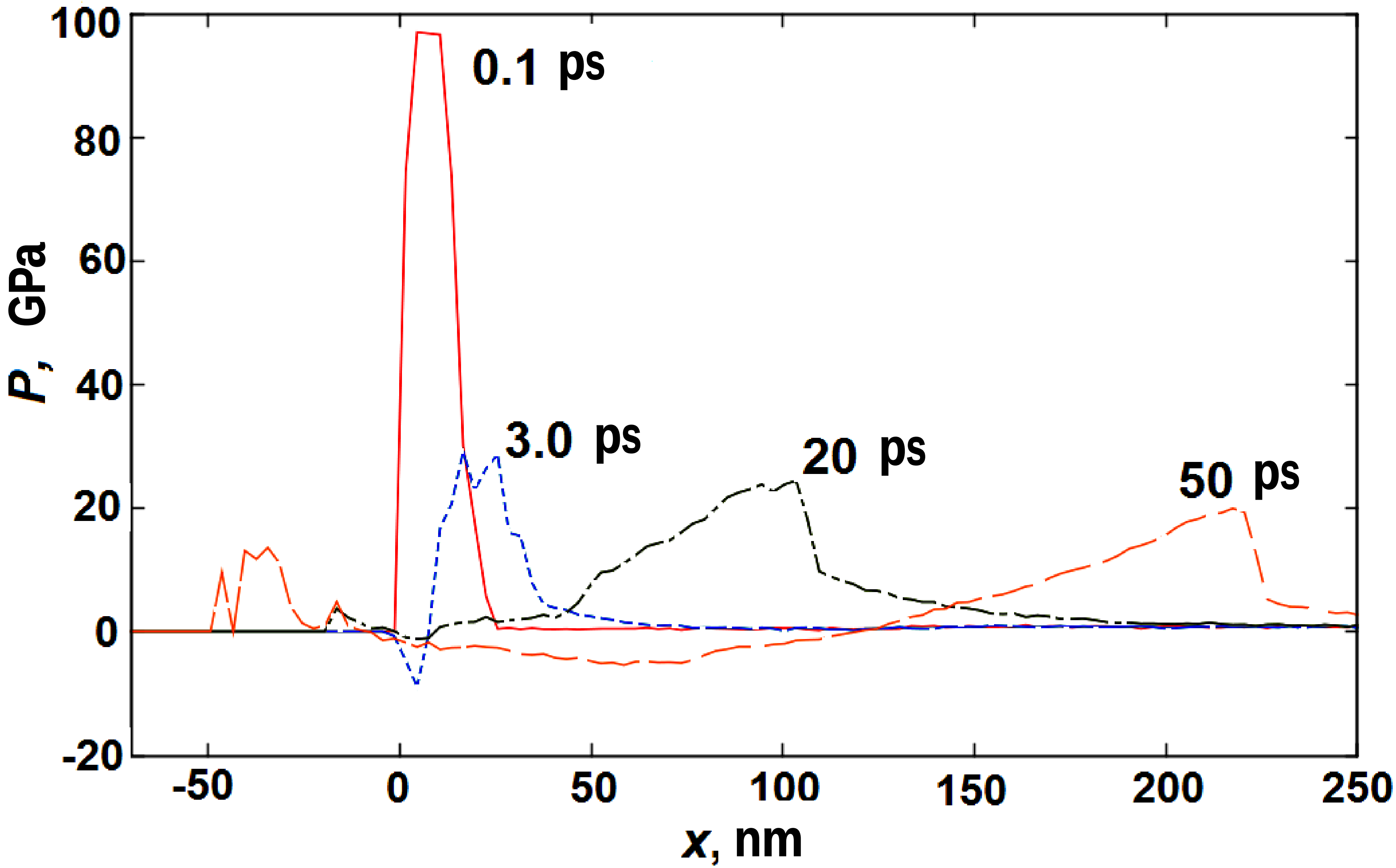
Laser ablation of gold

“short” and “long” ablation mechanisms for sub-*ps* pulses



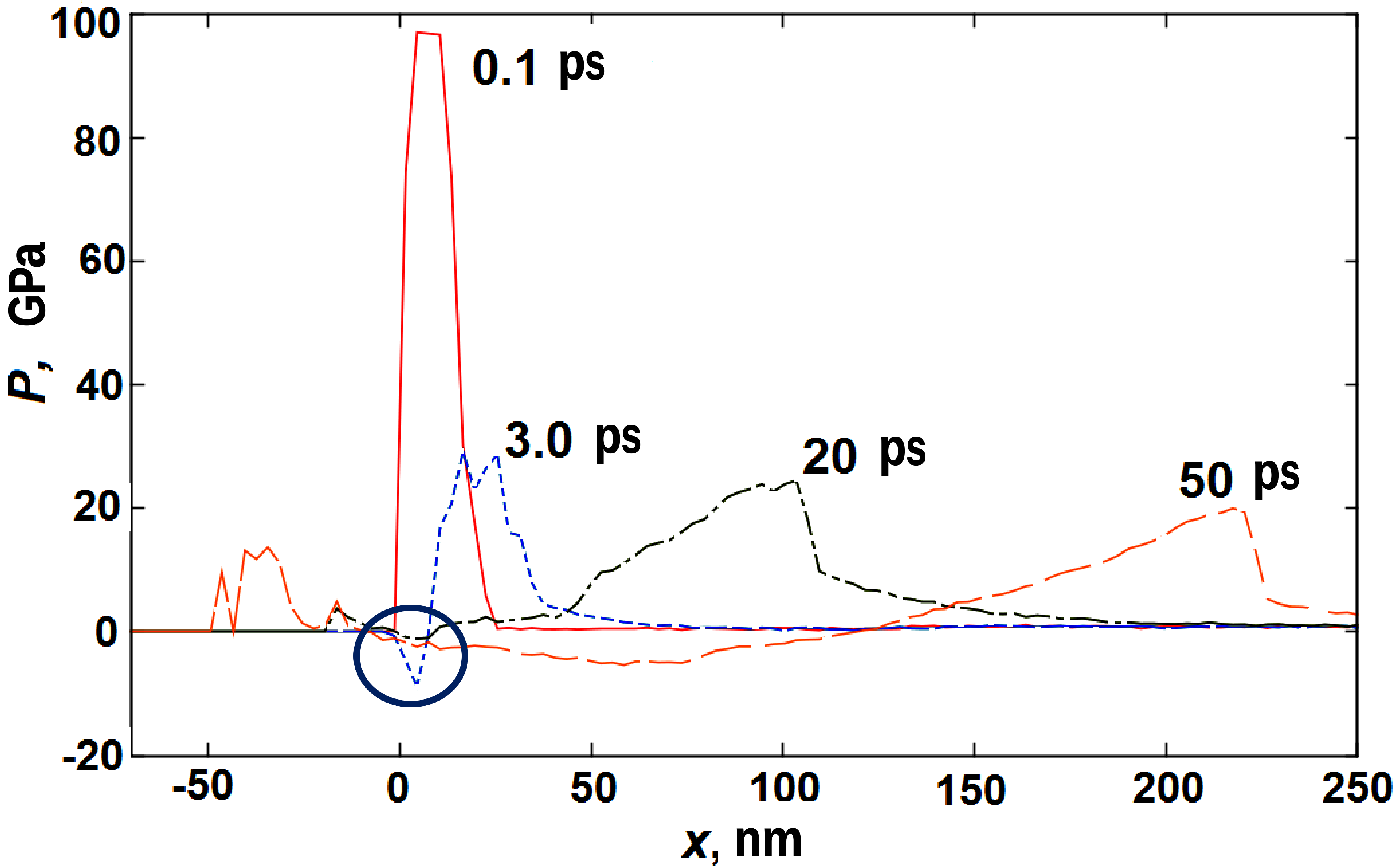
F = 1600 J/m²

ablation mechanism



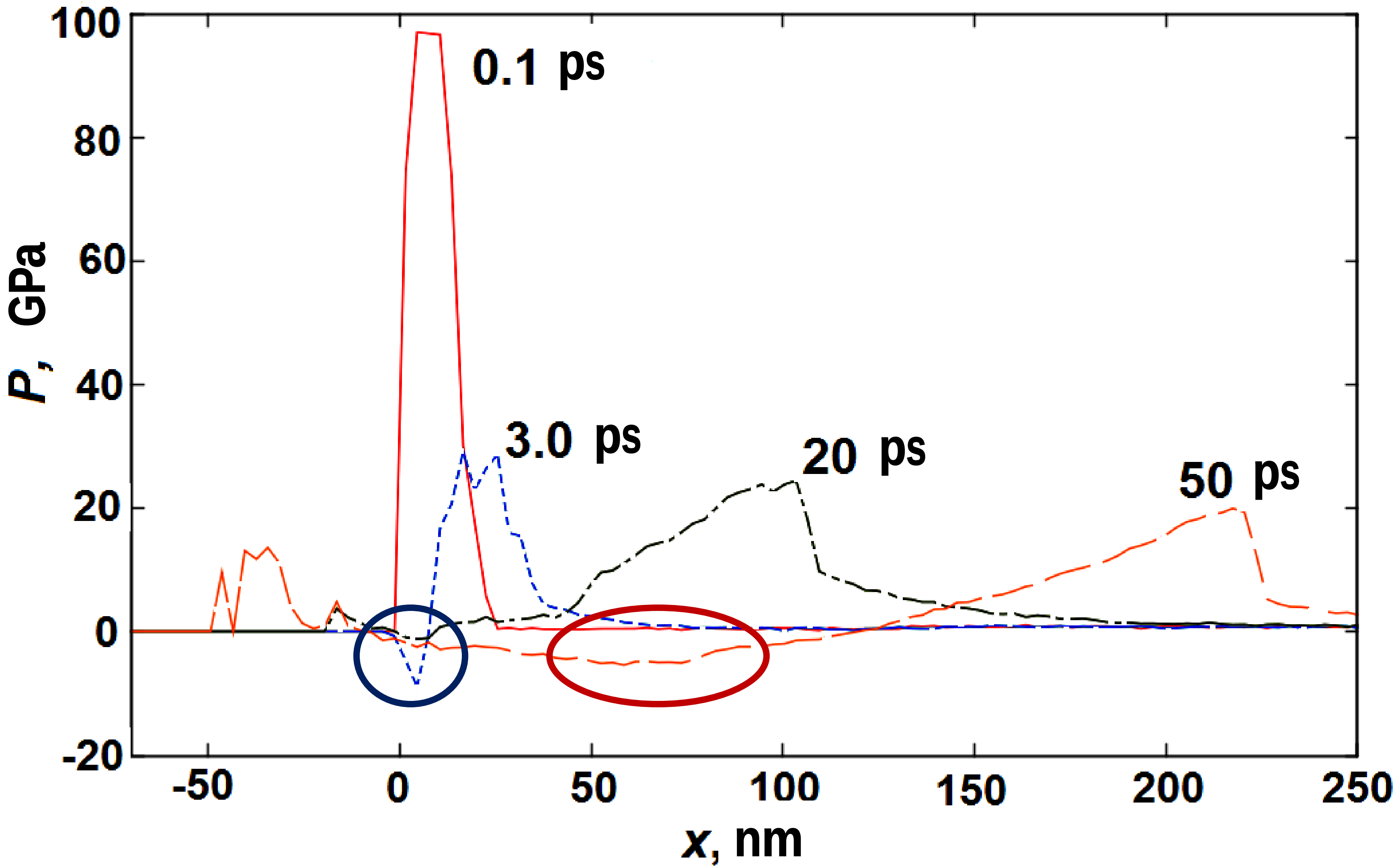
F = 1600 J/m^2

“short” ablation mechanism



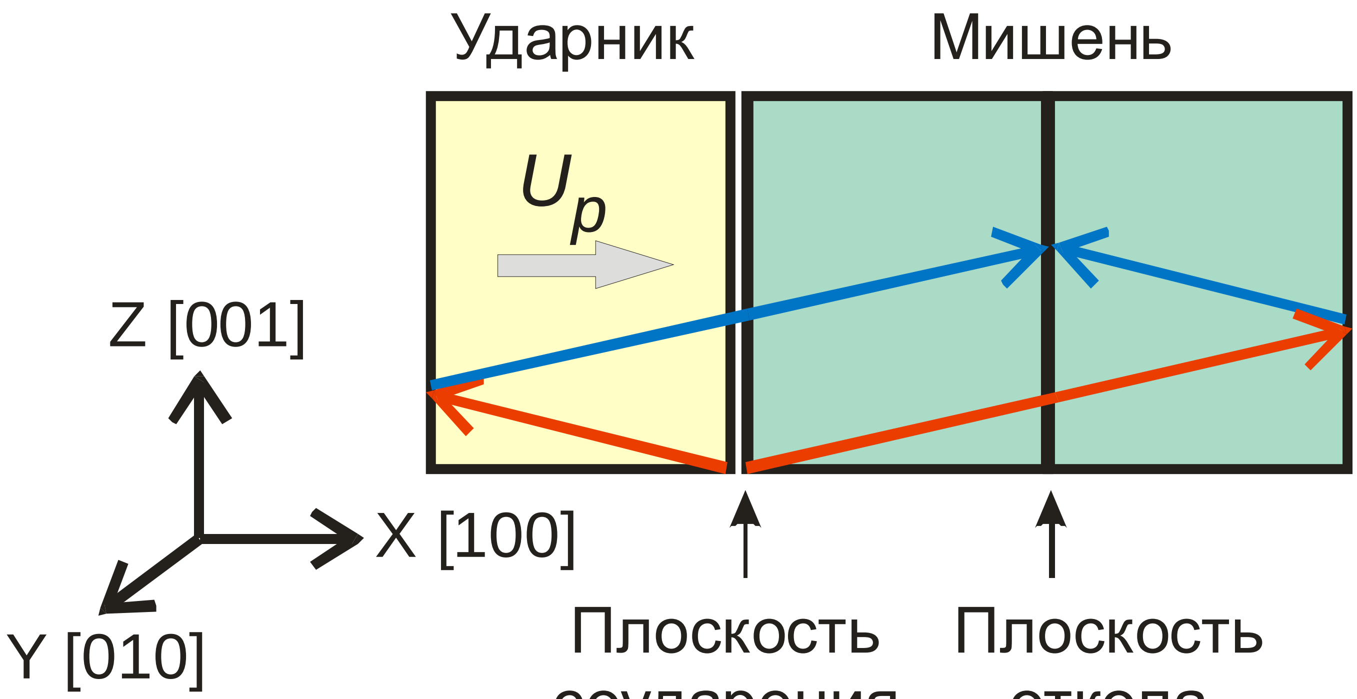
F = 1600 J/m^2

“long” and “short” ablation mechanisms

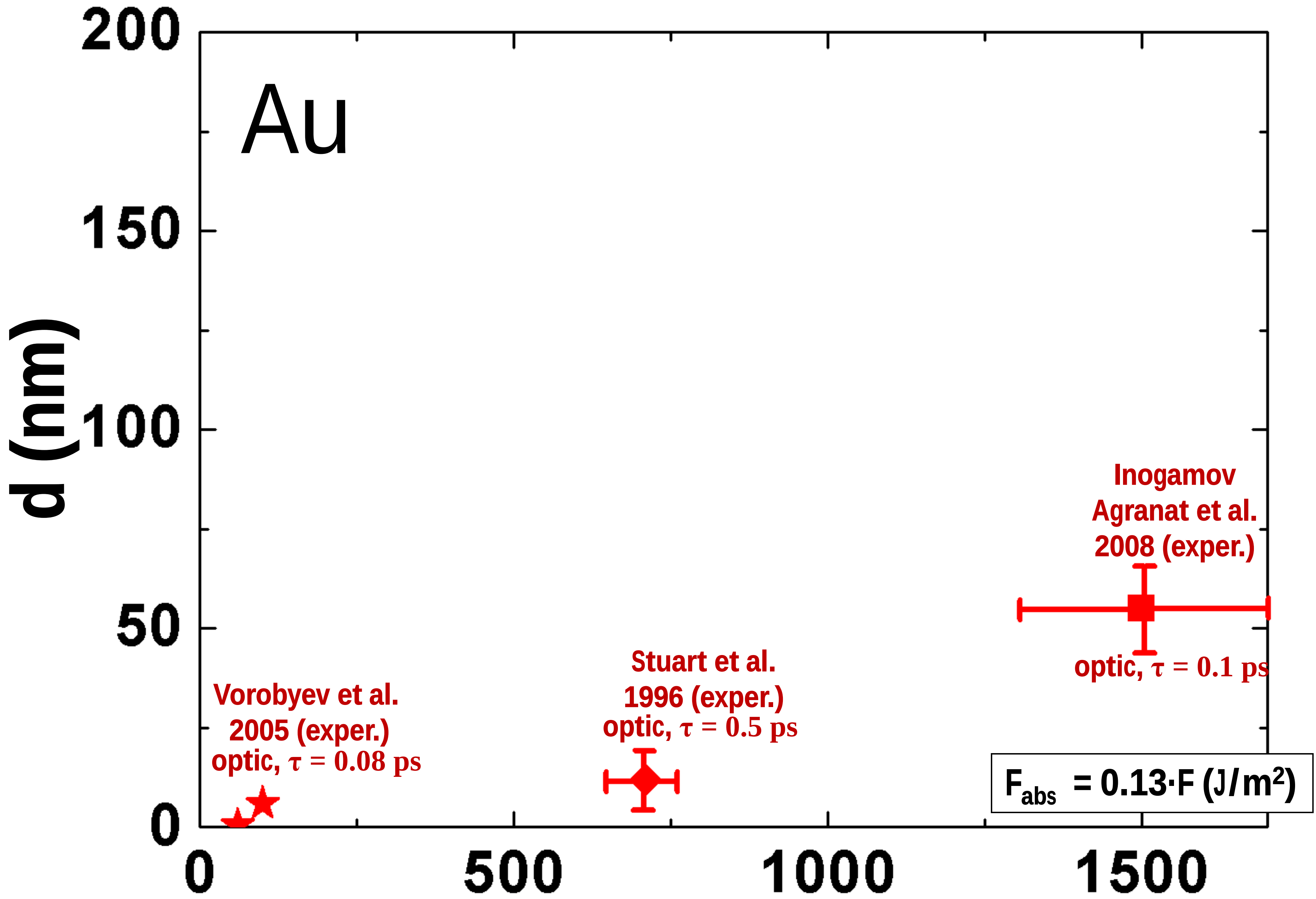


F = 1600 J/m²

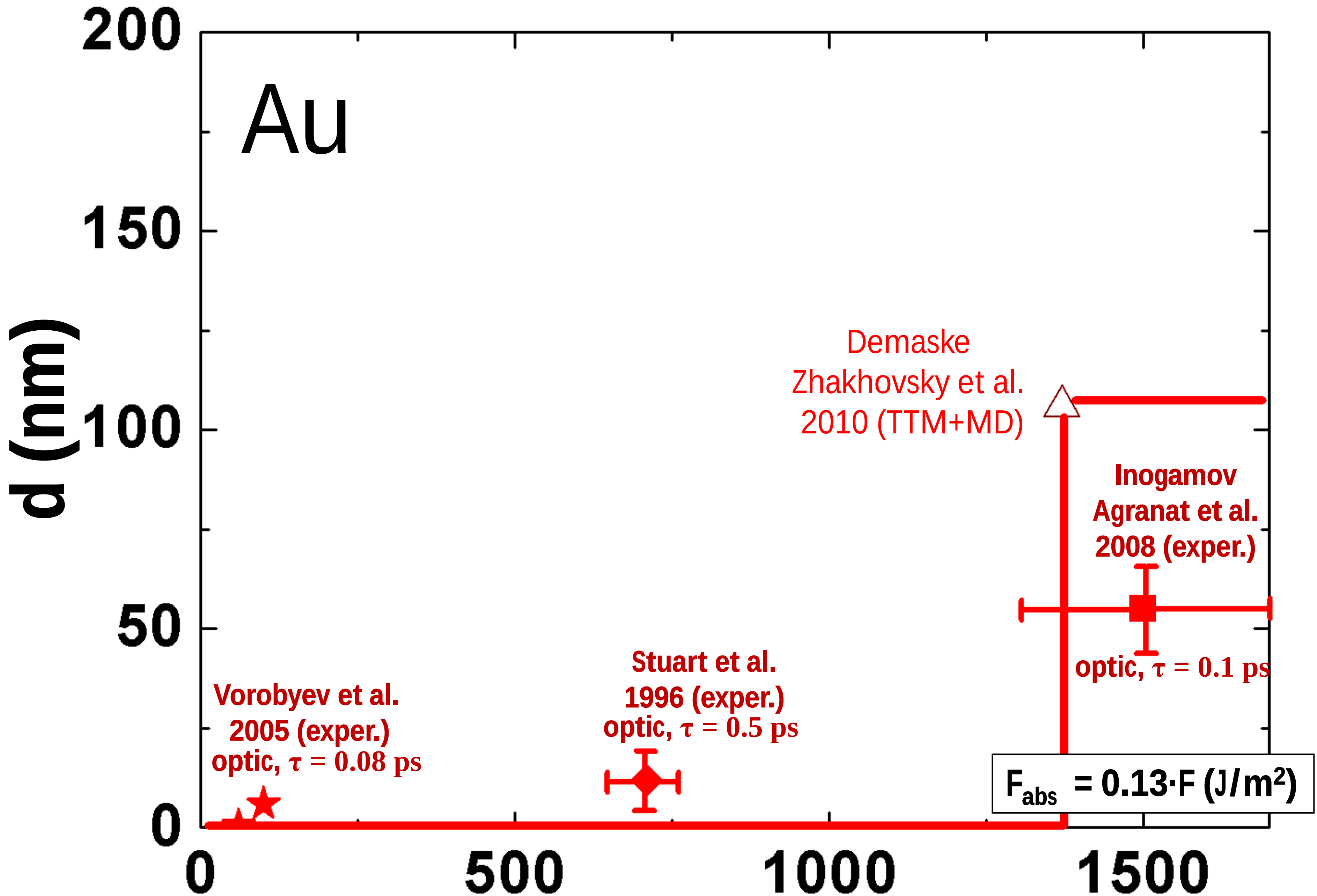
Схема откола при УВ нагружении



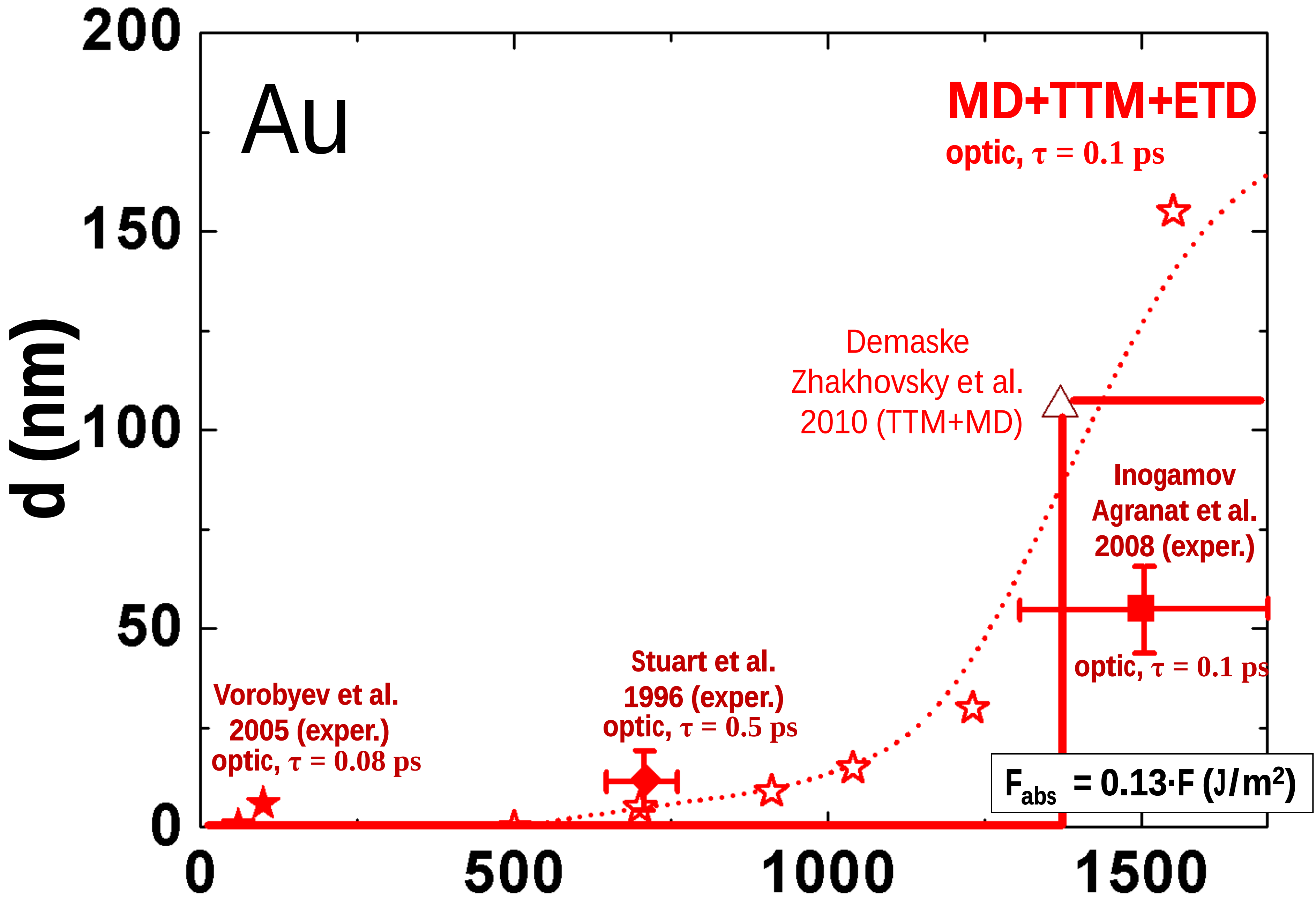
Dependence of crater depth on absorbed fluence (sub-ps pulses)



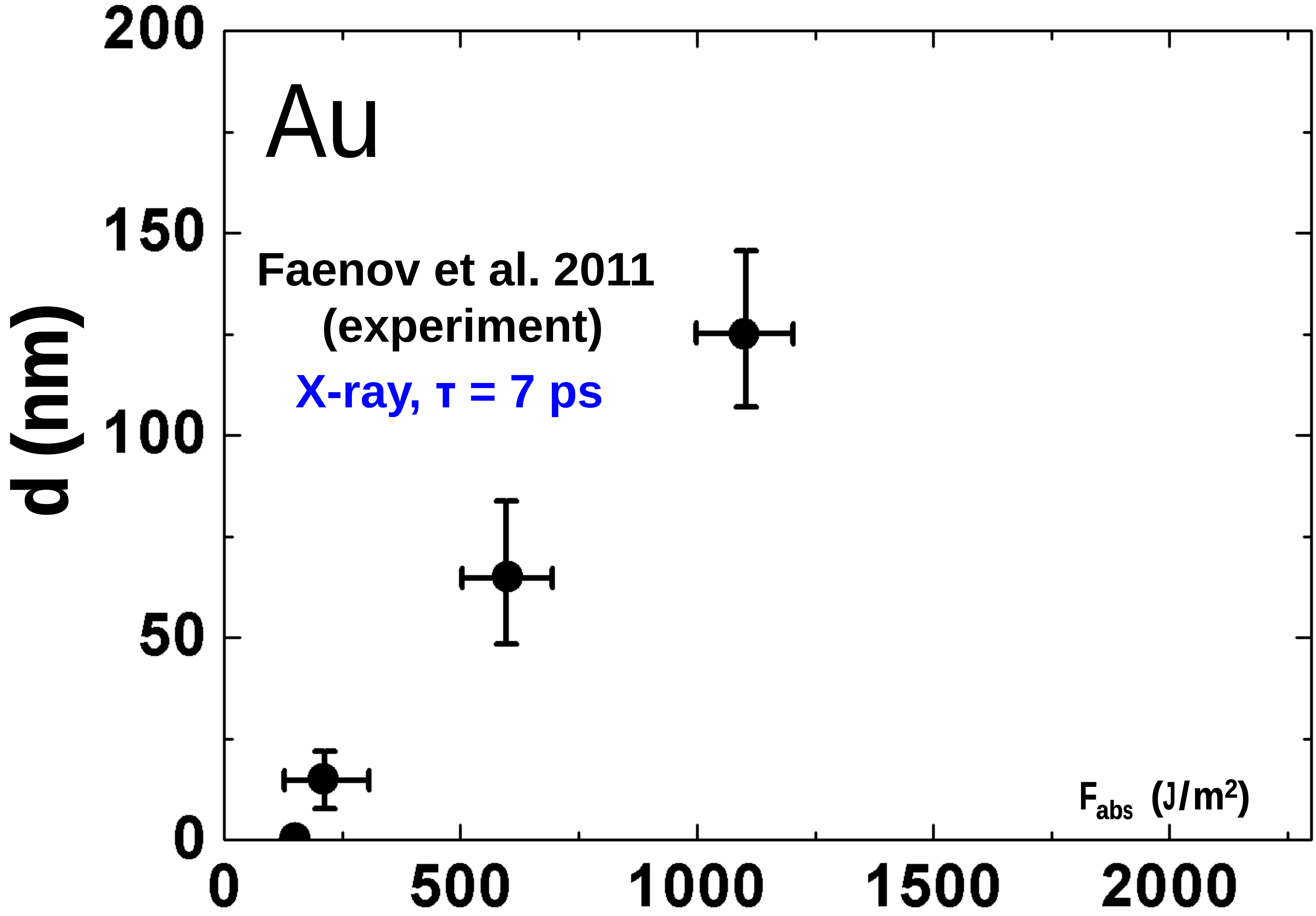
Dependence of crater depth on absorbed fluence (sub-ps pulses)



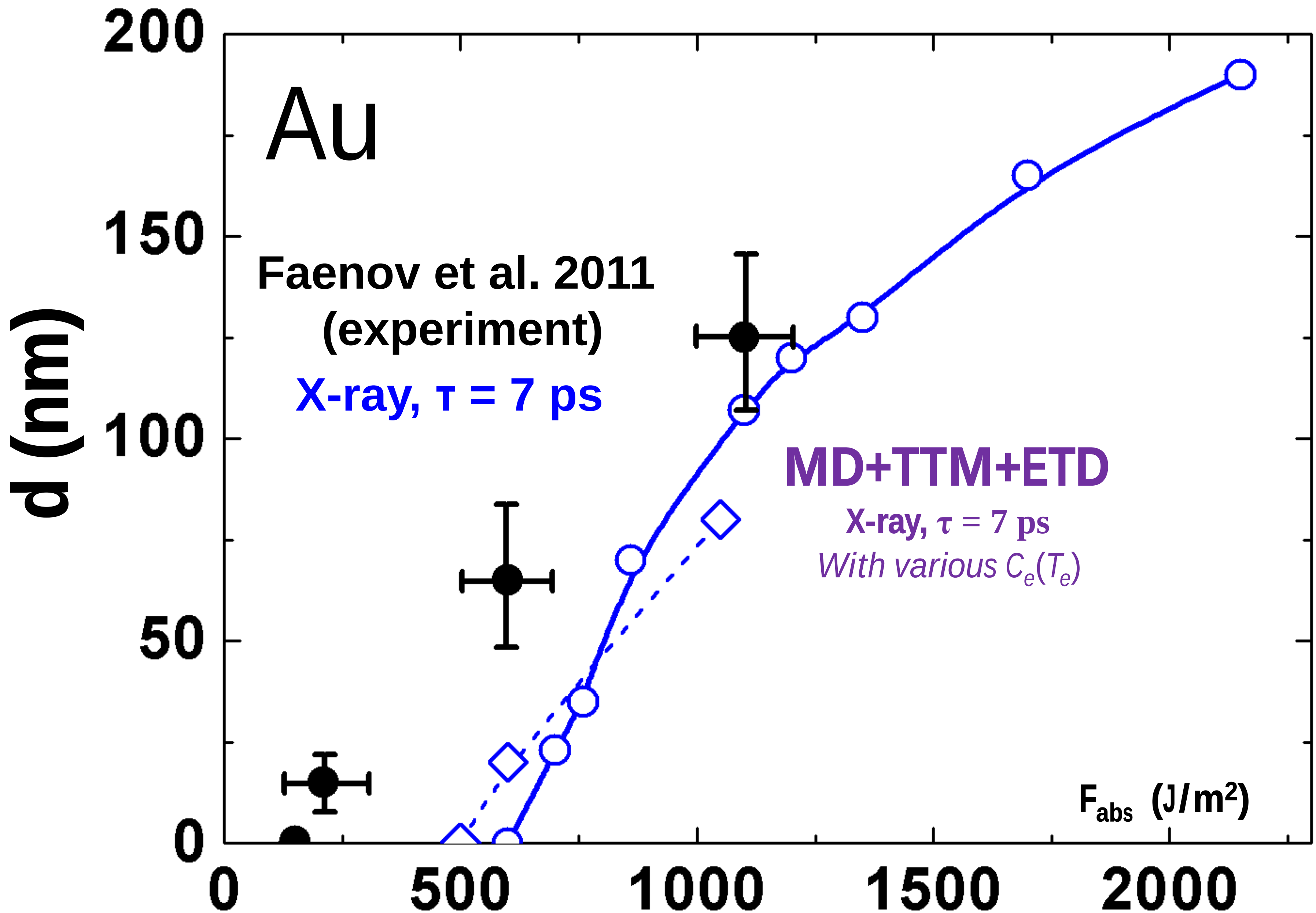
Dependence of crater depth on absorbed fluence (sub-ps pulses)



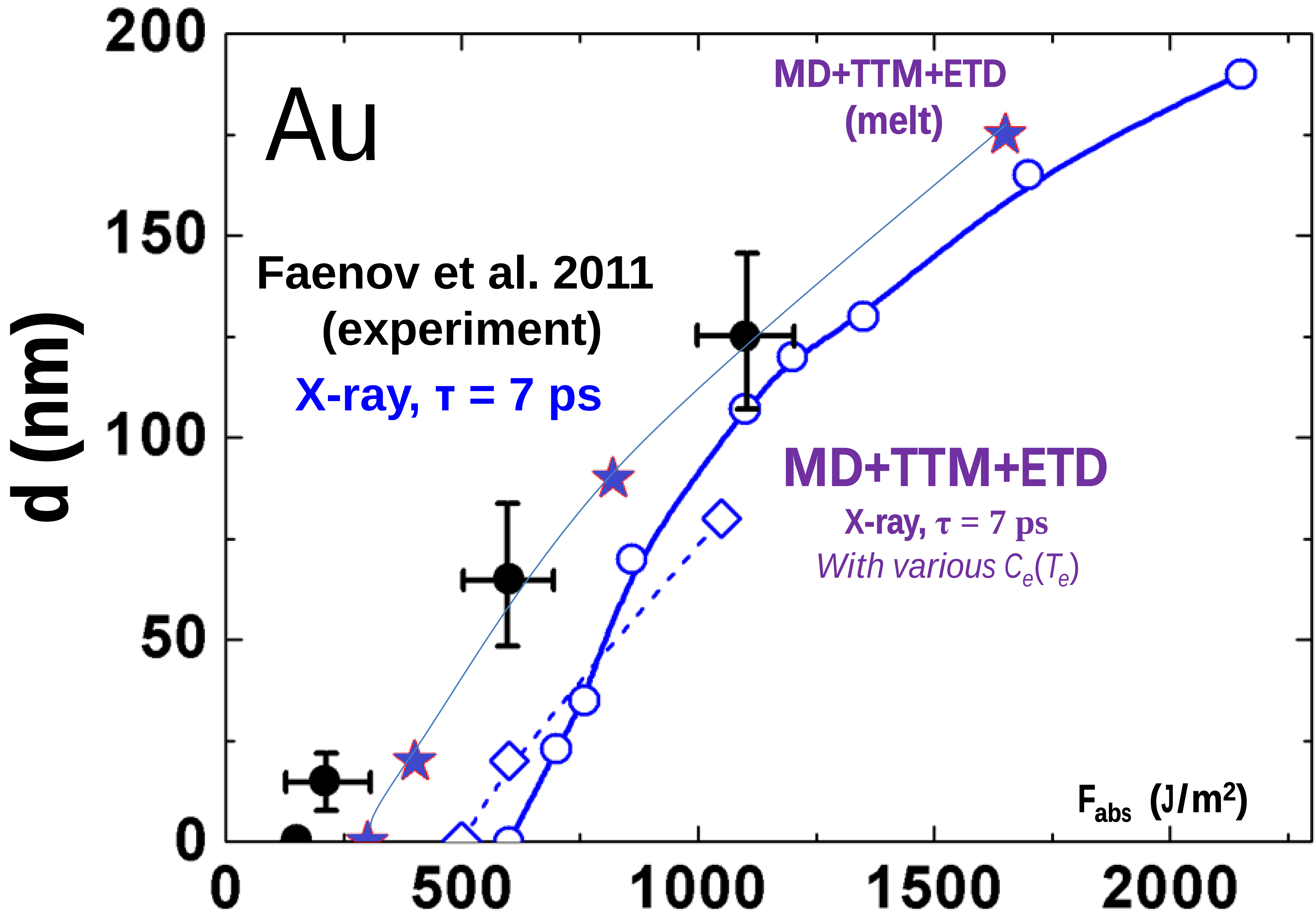
Laser ablation of gold (ps pulses)
Dependence of crater depth on absorbed fluence



Laser ablation of gold
Dependence of crater depth on absorbed fluence (ps pulse)



Laser ablation of gold
Dependence of crater depth on absorbed fluence (ps pulse)



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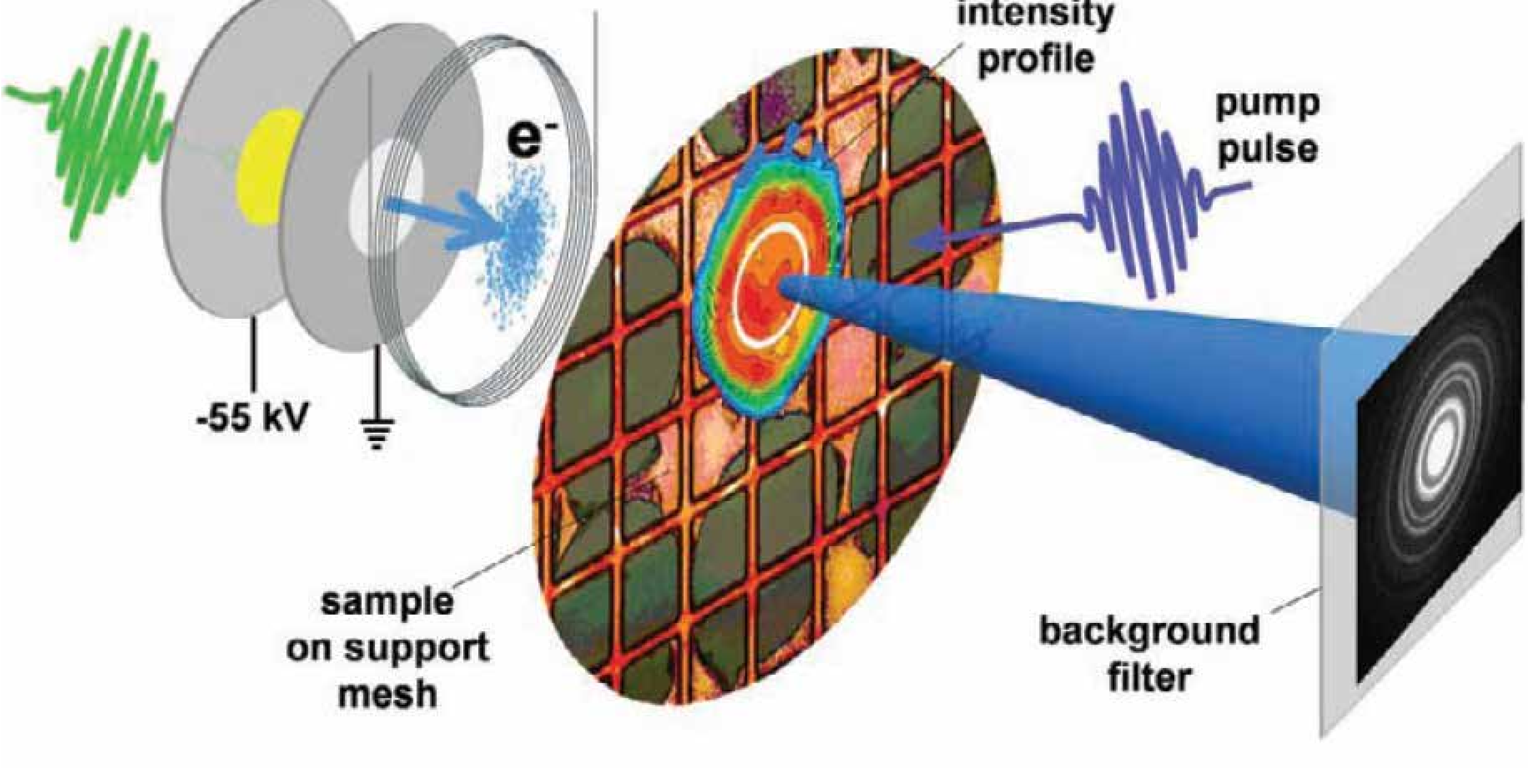
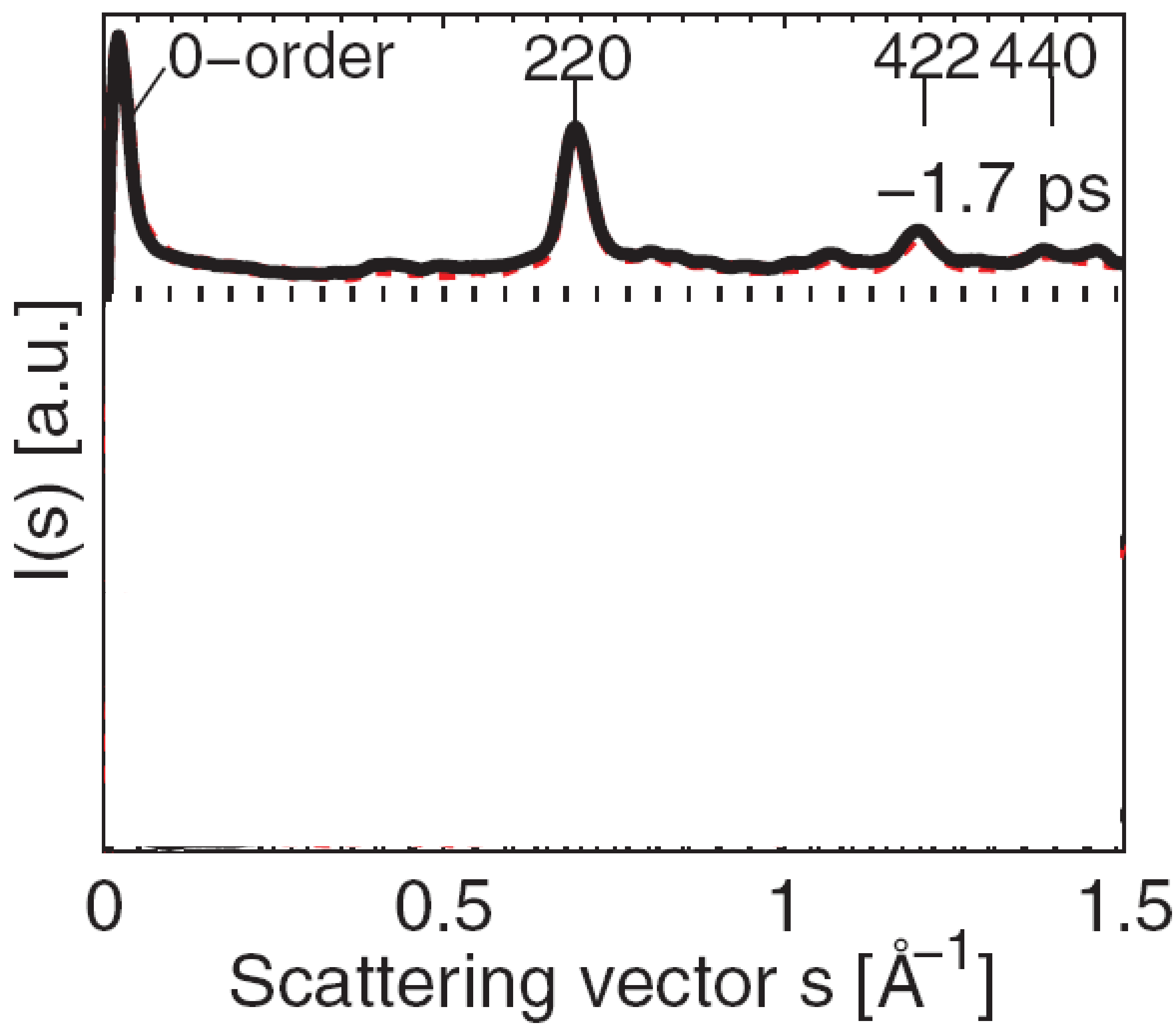
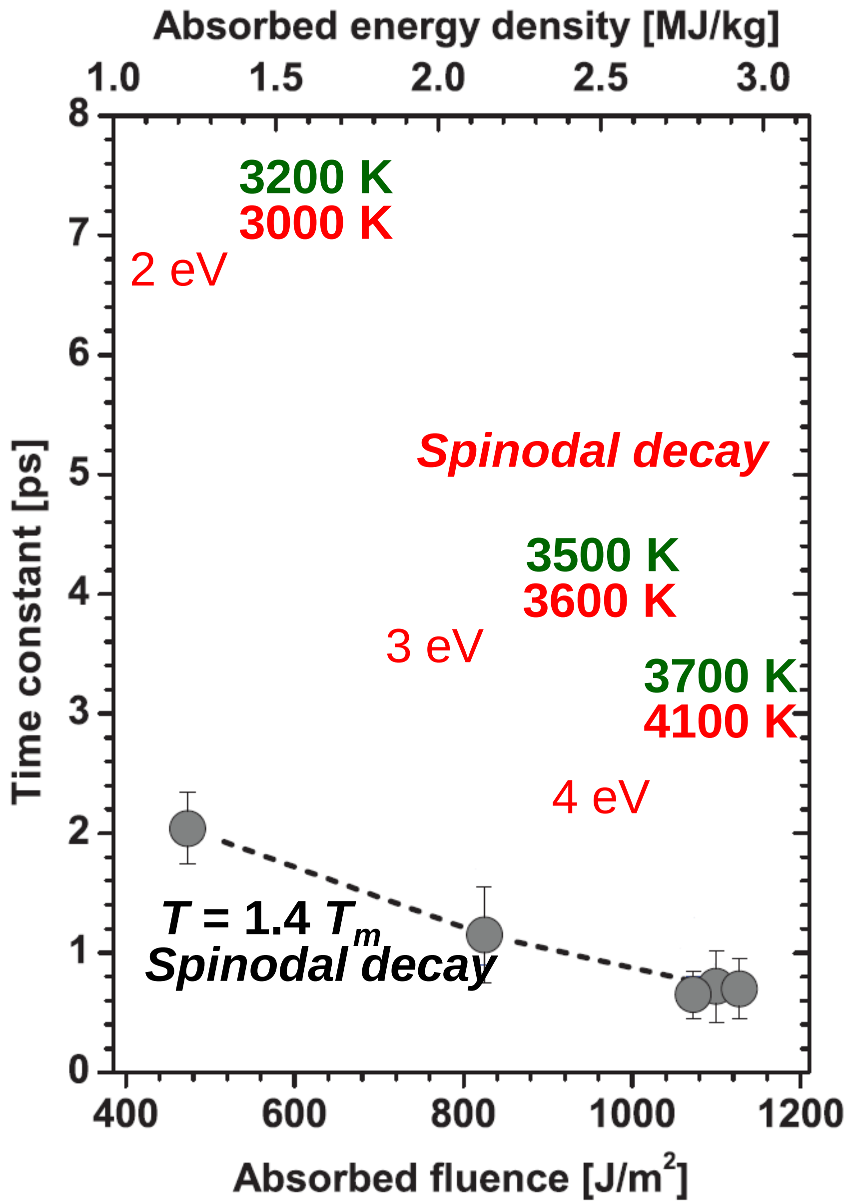
The Formation of Warm Dense Matter: Experimental Evidence for Electronic Bond Hardening in Gold.

R.Ernstorfer, M.Harb, C.Hebeisen, G.Sciaini, T.Dartigalongue, R.Miller

Science. Vol. 323. no. 5917, pp. 1033 – 1037. ***20 February 2009***

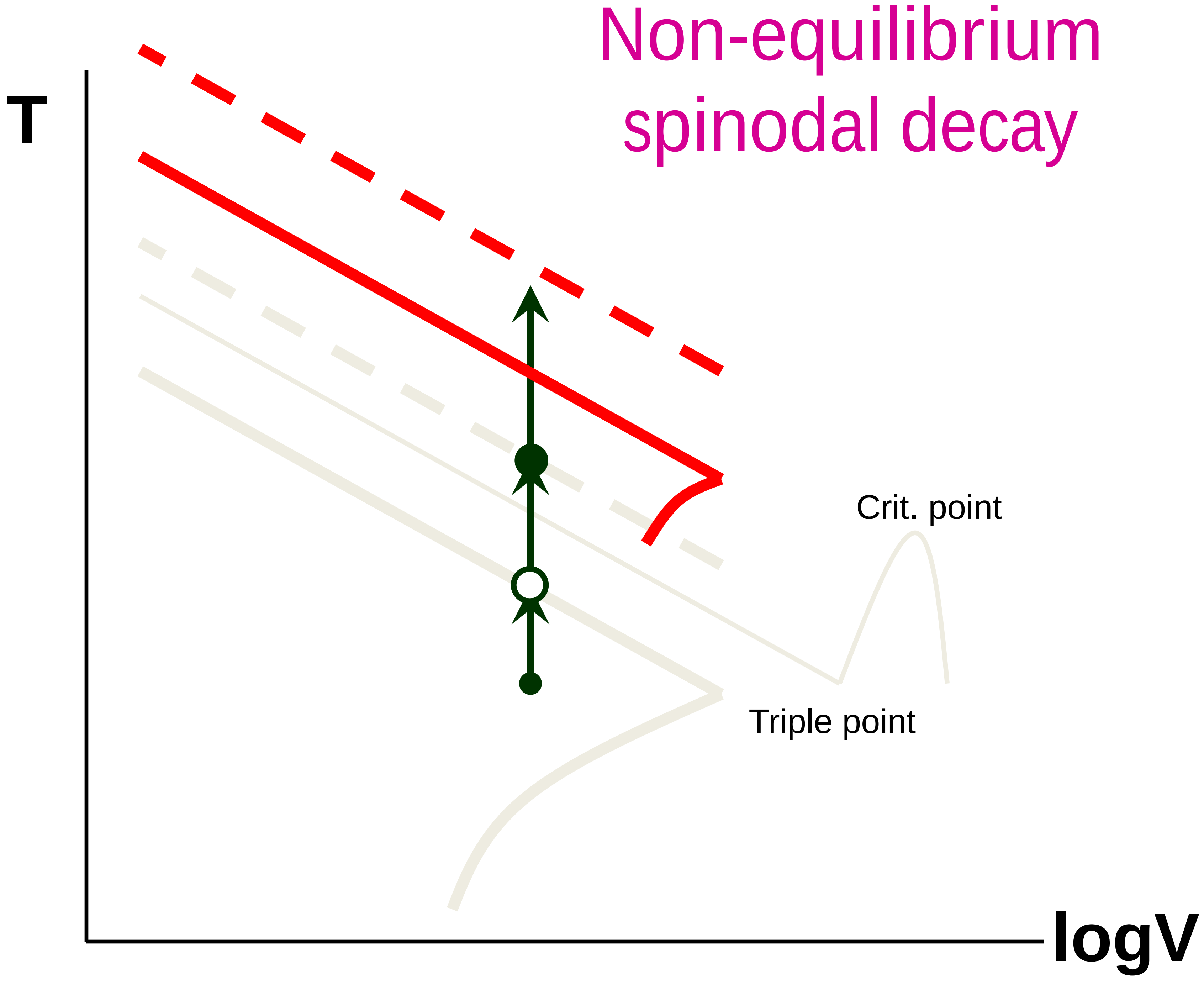

intensity
profile
e-
pump
pulse
-55 kV
sample
on support
mesh
background
filter


0–order
220
422
440
–1.7 ps
I(s) [a.u.]
0
0.5
1
1.5
Scattering vector s [Å^{-1}]



Absorbed energy density [MJ/kg]
1.0
1.5
2.0
2.5
3.0
Time constant [ps]
3200 K
3000 K
2 eV
Spinodal decay
3500 K
3600 K
3 eV
3700 K
4100 K
4 eV
T = 1.4 Tm
Spinodal decay
Absorbed fluence [J/m2]

Tm = 1330 K


Non-equilibrium
spinodal decay
T
Crit. point
Triple point
logV

The energy deposited in WDM
can be by order of magnitude higher
because of the creation of the
two-temperature state and
corresponding ion lattice hardening

The final explosion can be
a spinodal decay of
a particular two-temperature
metastable state

Formation of two-temperature state: experimental evidence for electronic bond hardening in gold

Ernstorfer R. et al. The formation of warm dense matter: experimental evidence for electronic bond hardening in gold // Science. 2009.

Recoules V. et al. Effect of Intense Laser Irradiation on the Lattice Stability of Semiconductors and Metals// PRL. 2006.

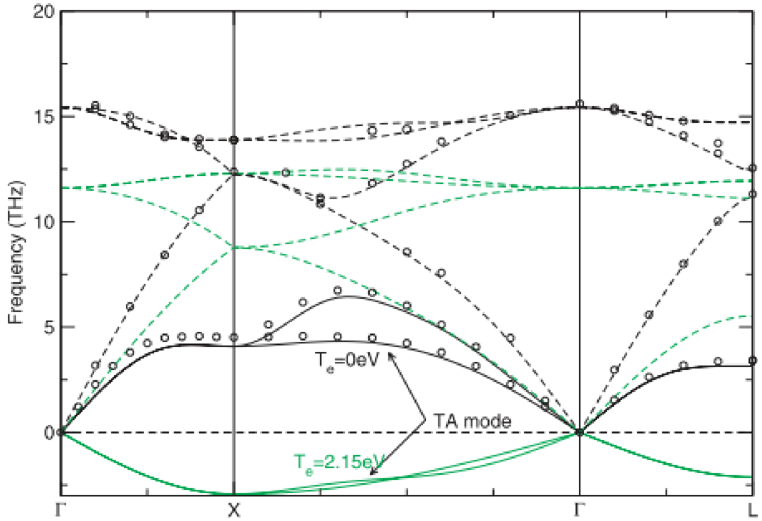


FIG. 1 (color online). Phonon spectrum of Si at different electronic temperature. The black curve is the spectrum for Te = 0 eV. The green curves are for Te = 2.15 eV. Open circles are experimental results from [26].

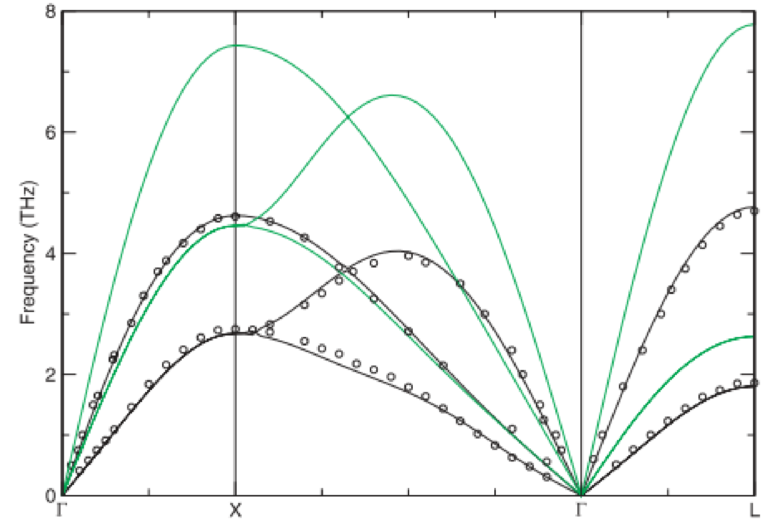
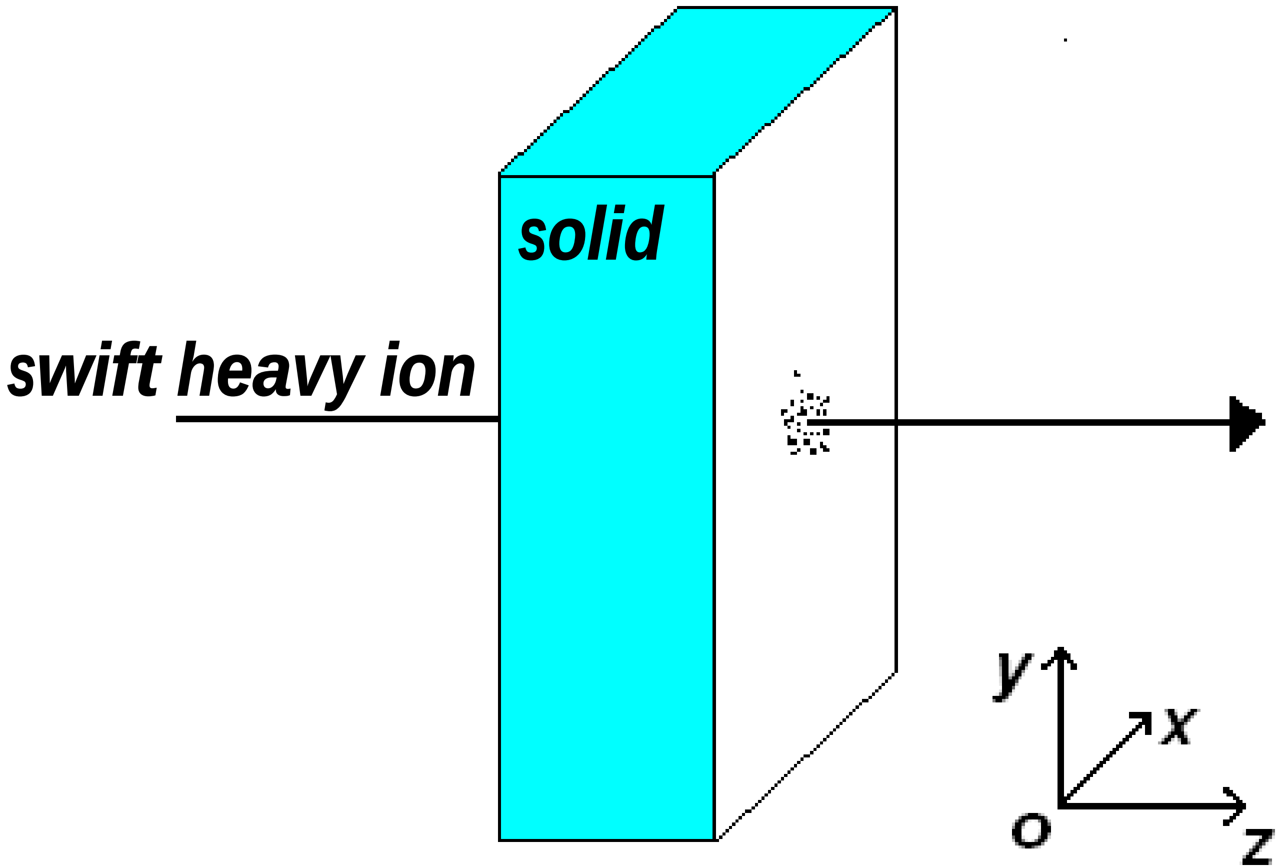


FIG. 2 (color online). Phonon spectrum of Au at different electronic temperatures. The black curves are the spectrum for room temperature. The green curves are for Te = 6 eV. Open circles are experimental results from [27].

Simulation of tracks formation in metals

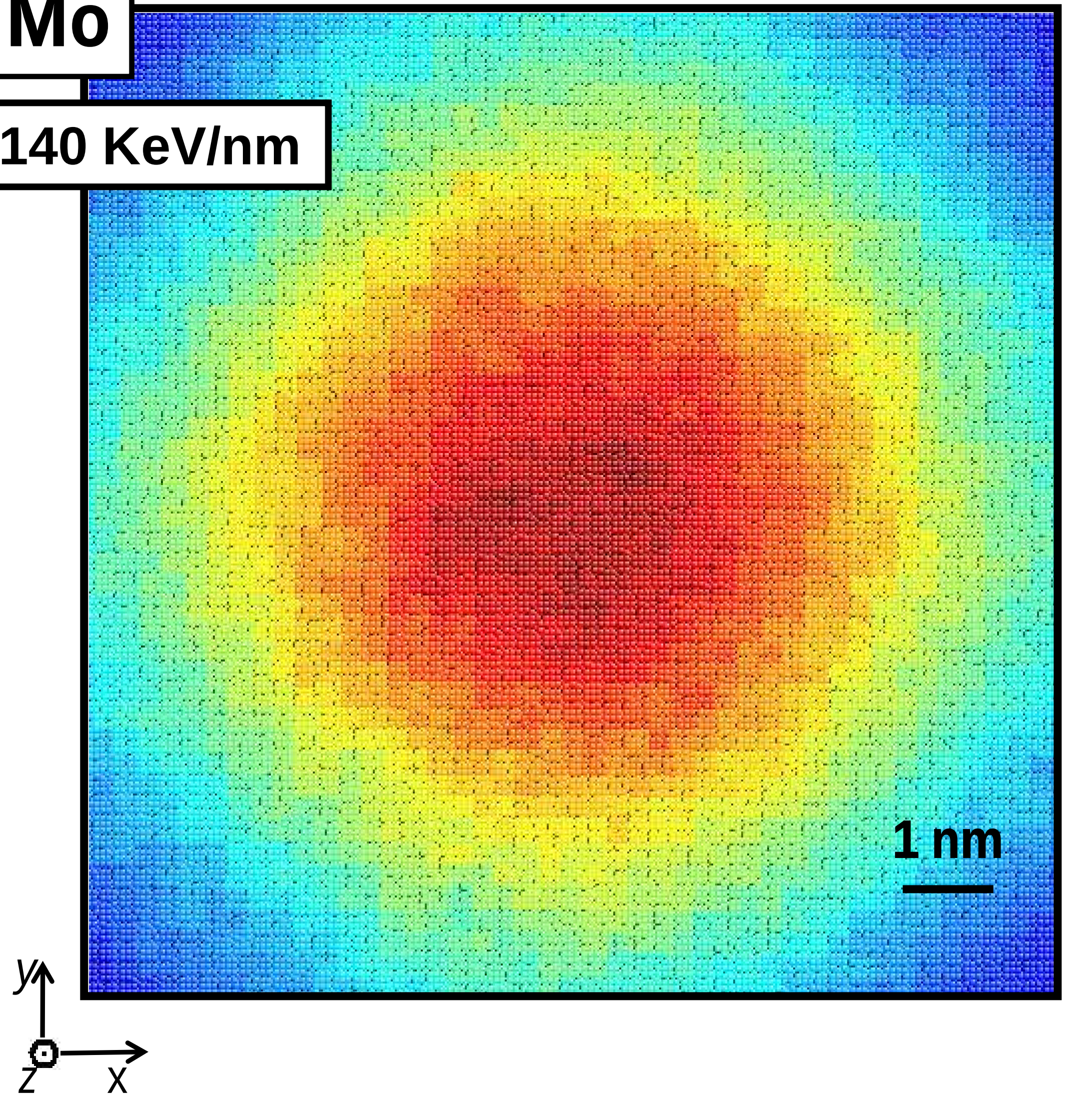
Moving of high energy ion through matter

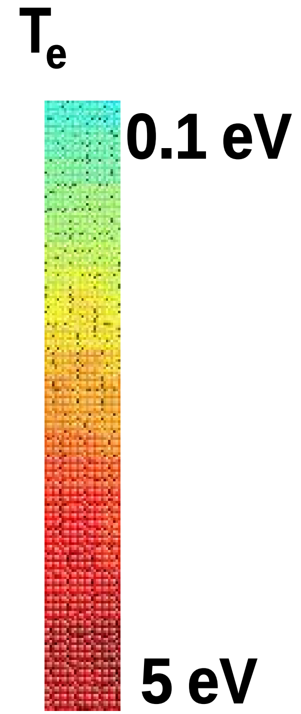


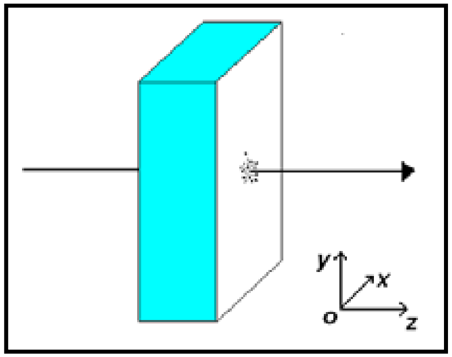
Initial distribution of electronic temperature

Xe in Mo

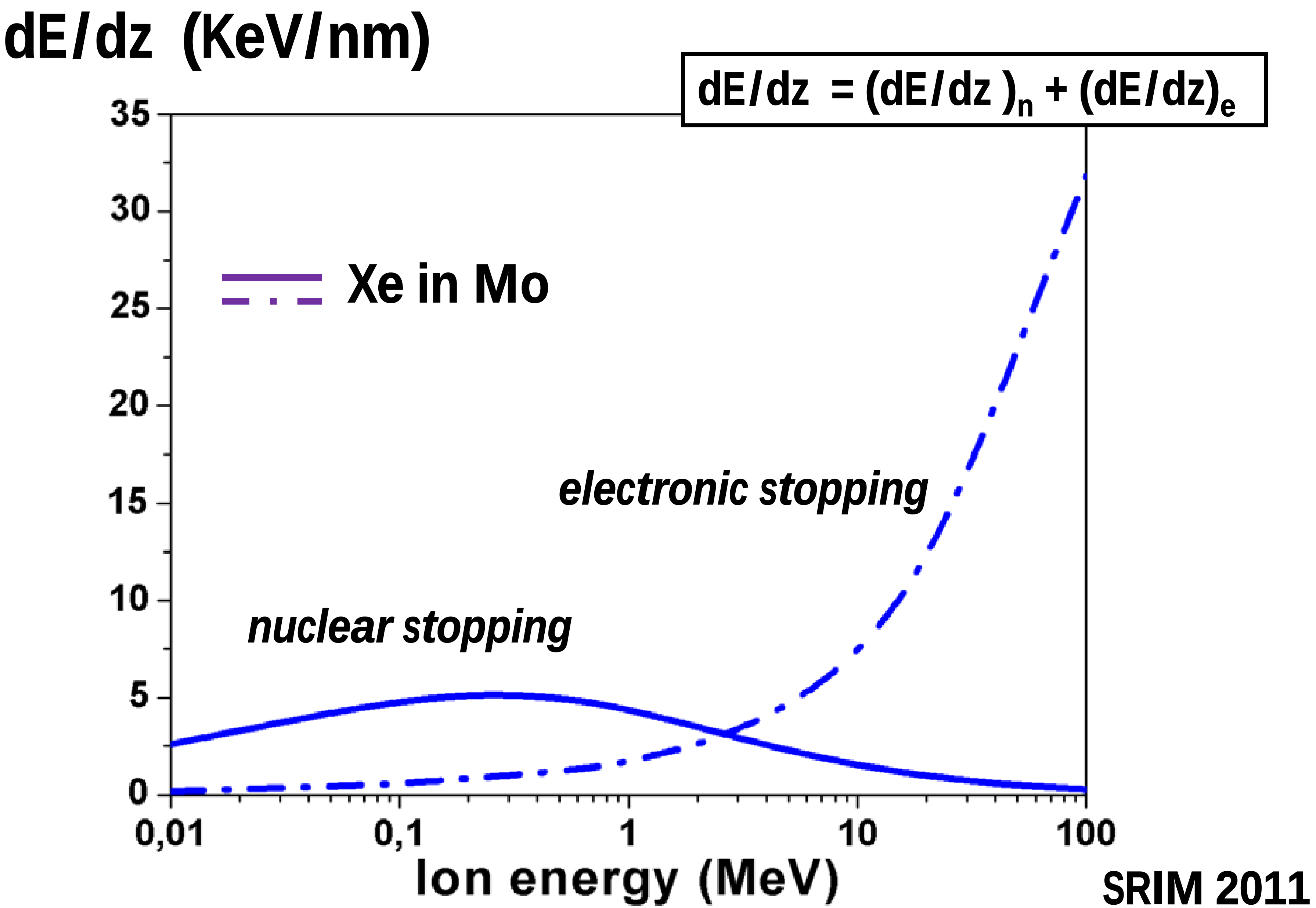
dE/dz = 140 KeV/nm



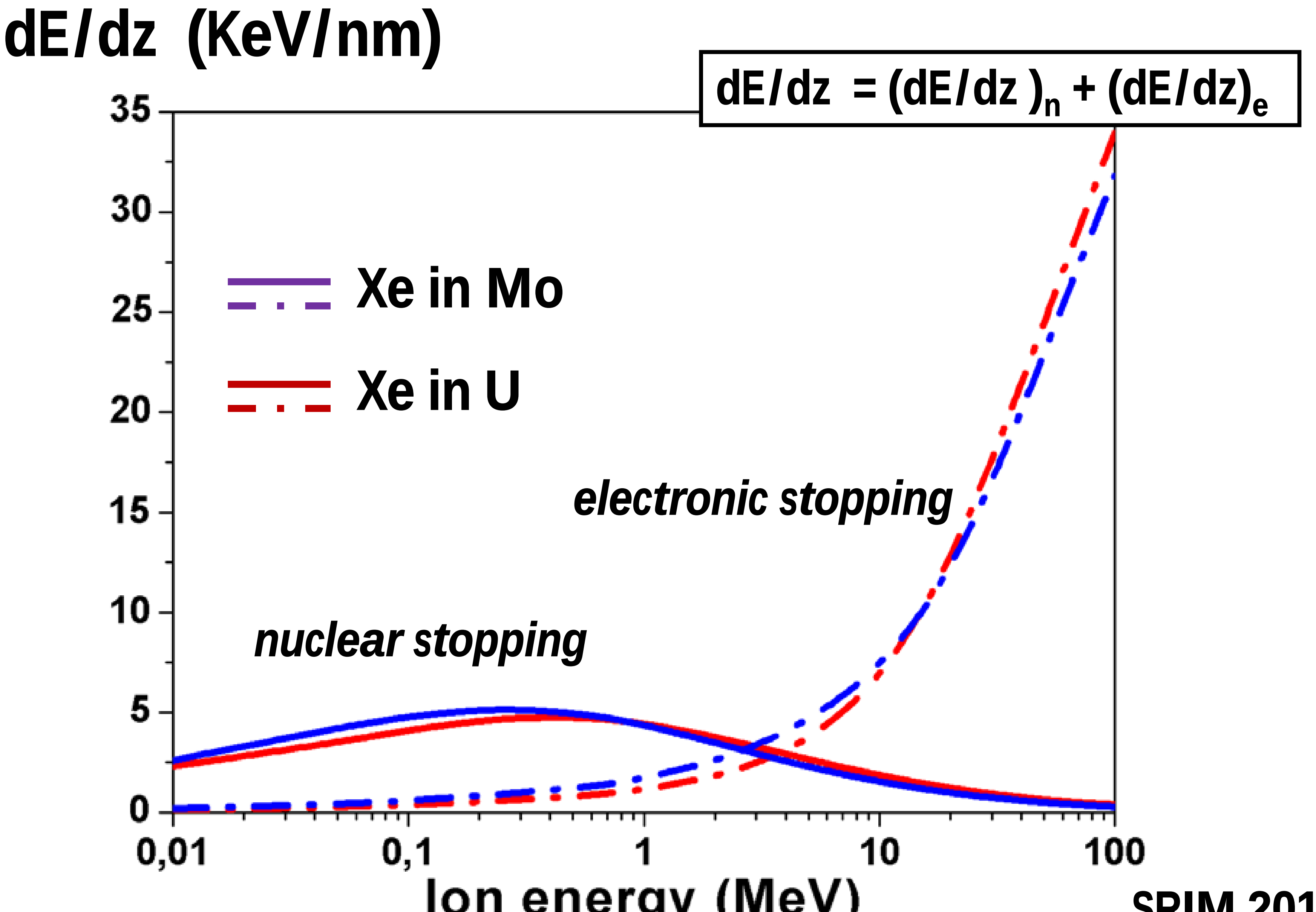




Energy loss of Xe ion in metal



Energy loss of Xe ion in metal



SRIM 2011

Two-temperature model

Ion subsystem

$$m\frac{d\mathbf{v}_i}{dt} = \mathbf{F}_i - \beta\,\mathbf{v}_i + \mathbf{F}_i^{Lang}(T_e)$$

Electronic subsystem

$$C_e\frac{\partial T_e}{\partial t} = \nabla(K_e \nabla T_e) - G_e(T_e - T_i)$$

Interatomic potential

Mo

Starikov S., Insepov Z., Rest J., Kuksin A., Norman G., Stegailov V., Yanilkin A.
// *Phys. Rev. B* **84** 104109 (2011)

U

Smirnova D., Starikov S., Stegailov V.
// *J. Phys.: Condens. Matter* **24** 015702 (2012)

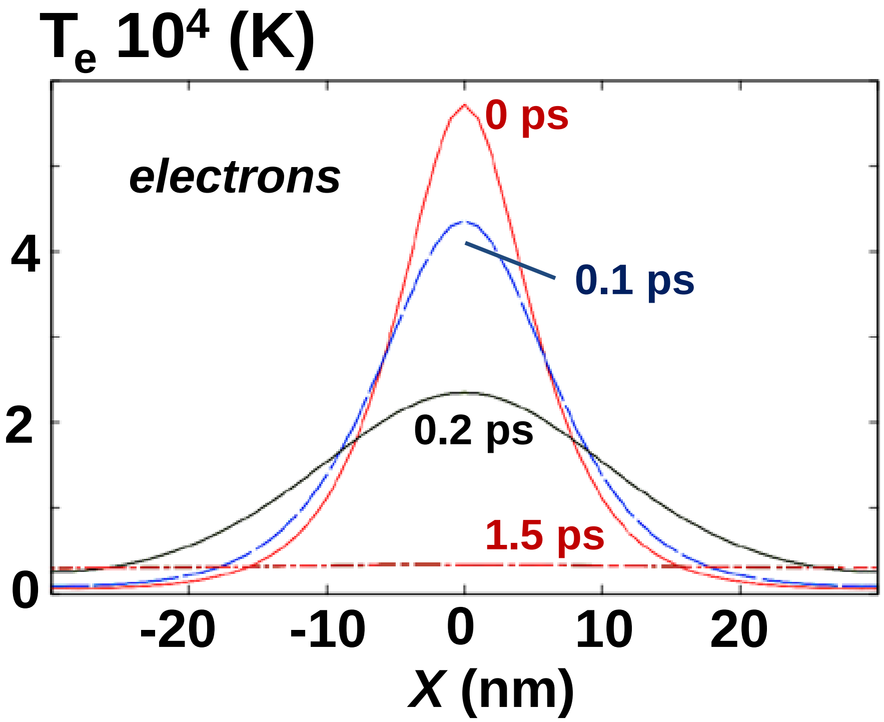
Characteristics of the electronic subsystem

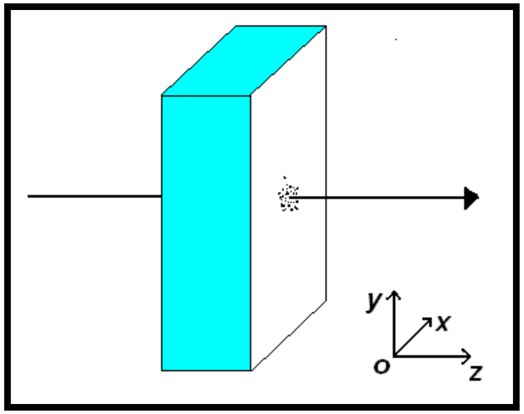
$C_e = \gamma\, T_e$; K_e ; G

Simulation of track formation in Mo

Profiles of electronic and ionic temperatures

dE/dz = 140 KeV/nm

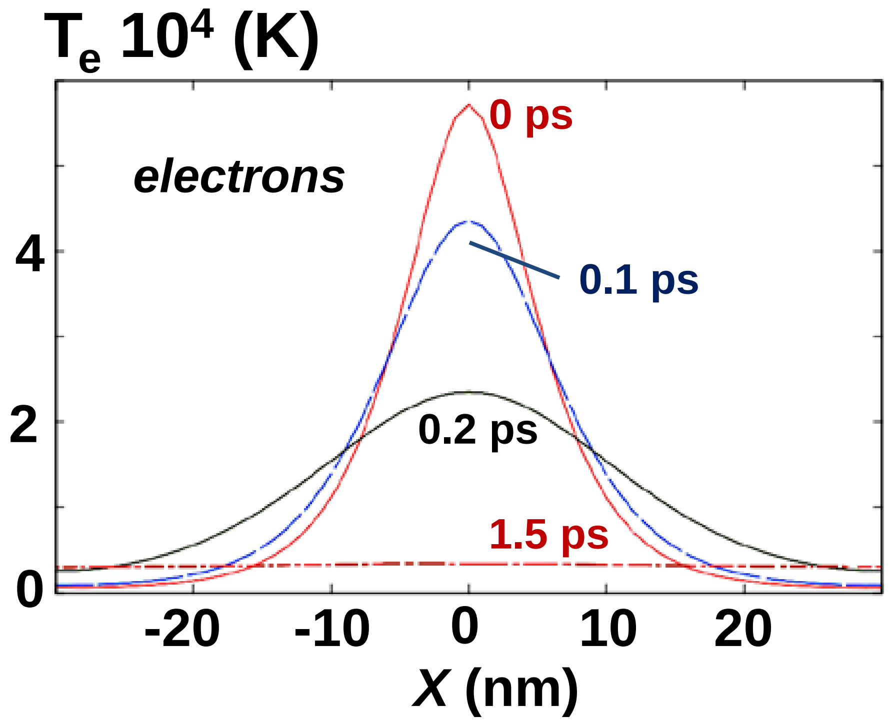


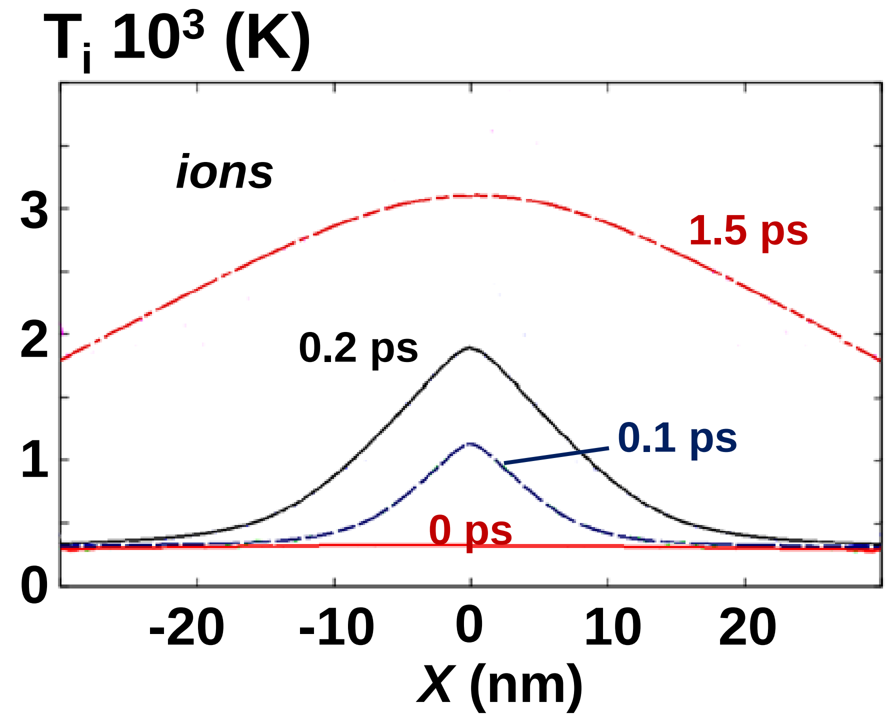


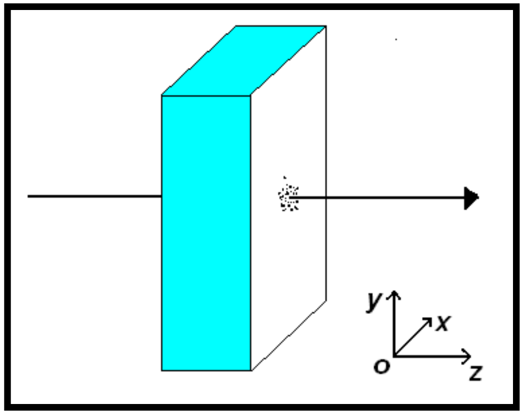
Mo

Profiles of electronic and ionic temperatures

dE/dz = 140 KeV/nm



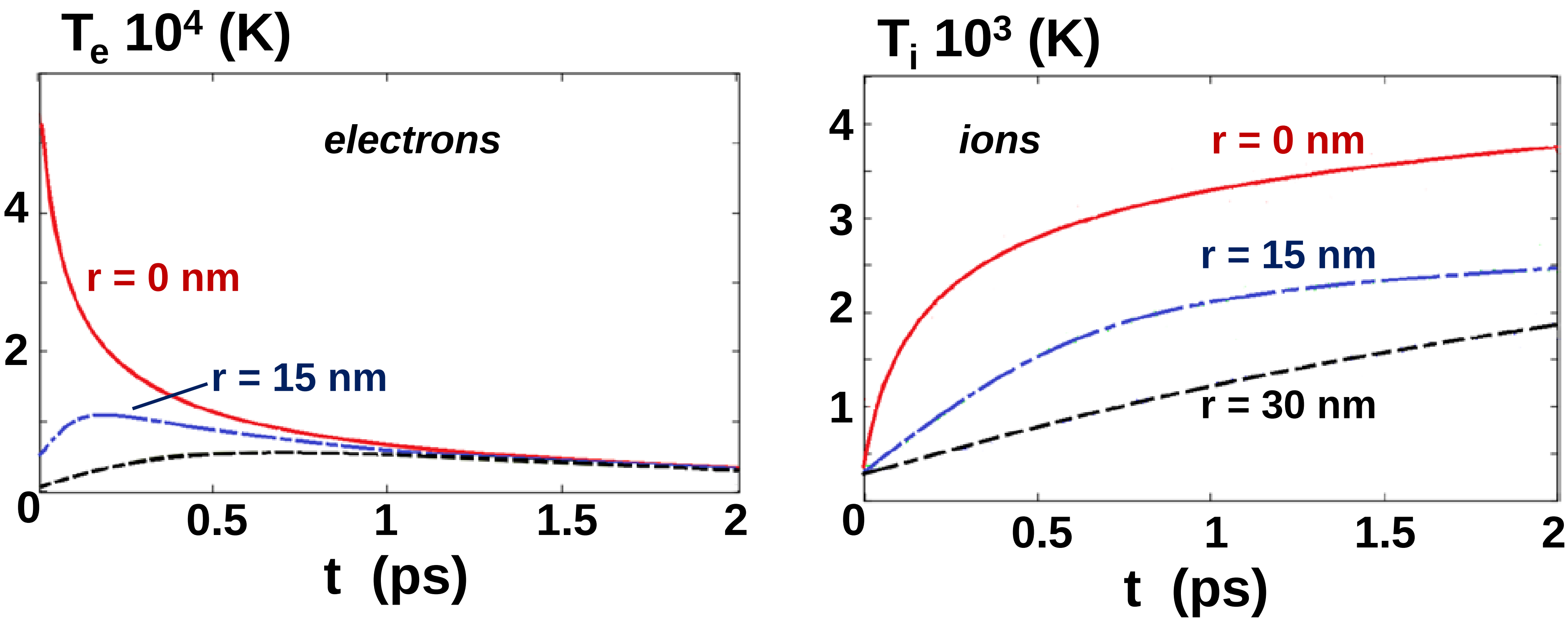




Mo

Dependence of temperature on time

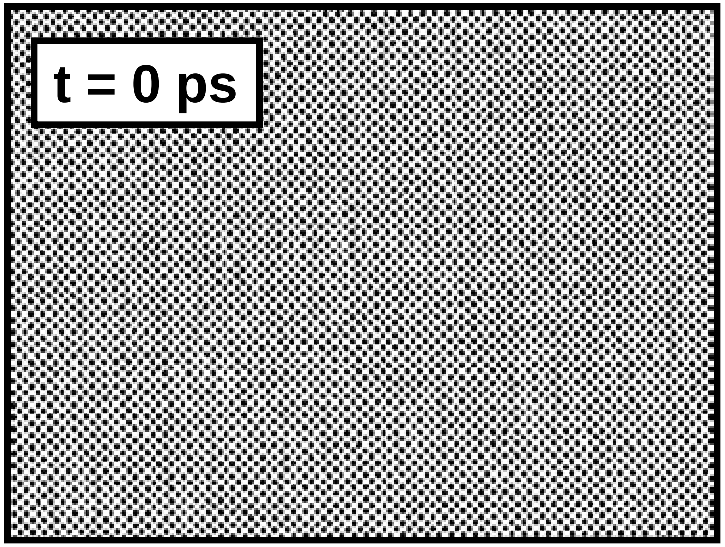
dE/dz = 140 KeV/nm



Mo

Defect formation at heating/melting of ionic subsystem

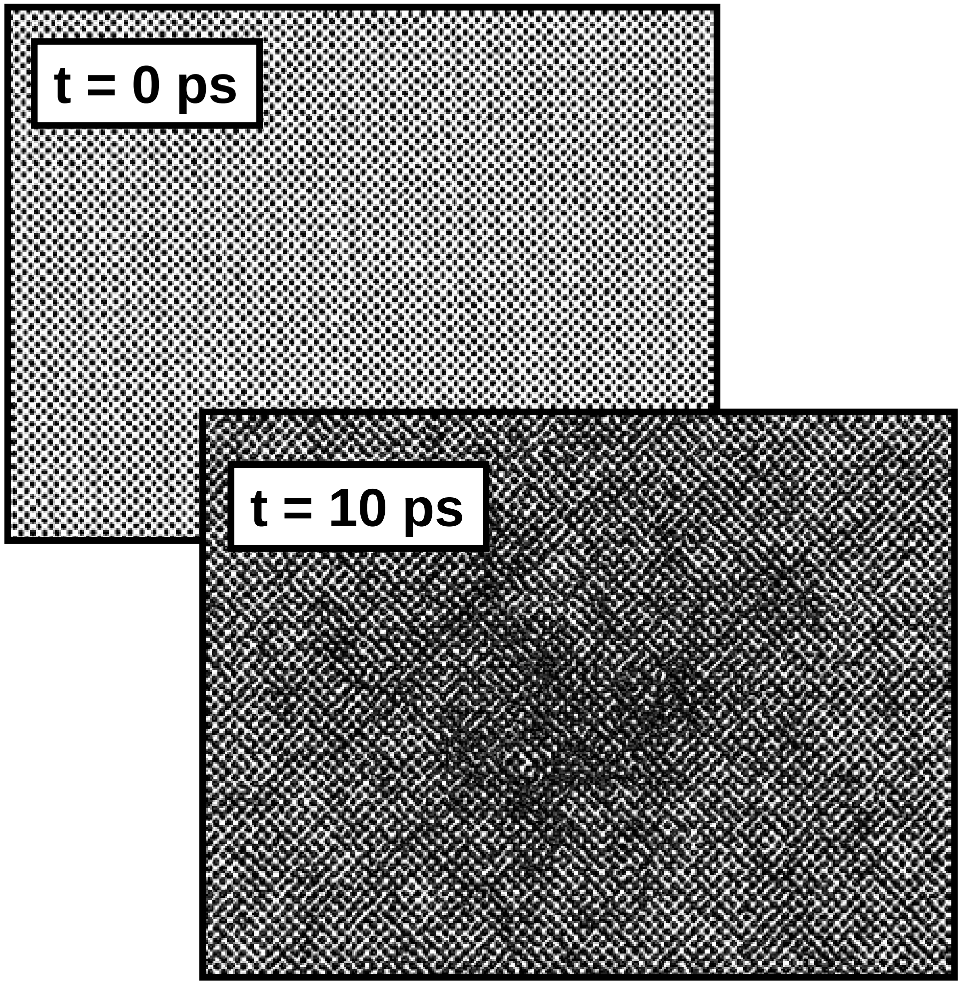
dE/dz = 160 KeV/nm



Mo

Defect formation at heating/melting of ionic subsystem

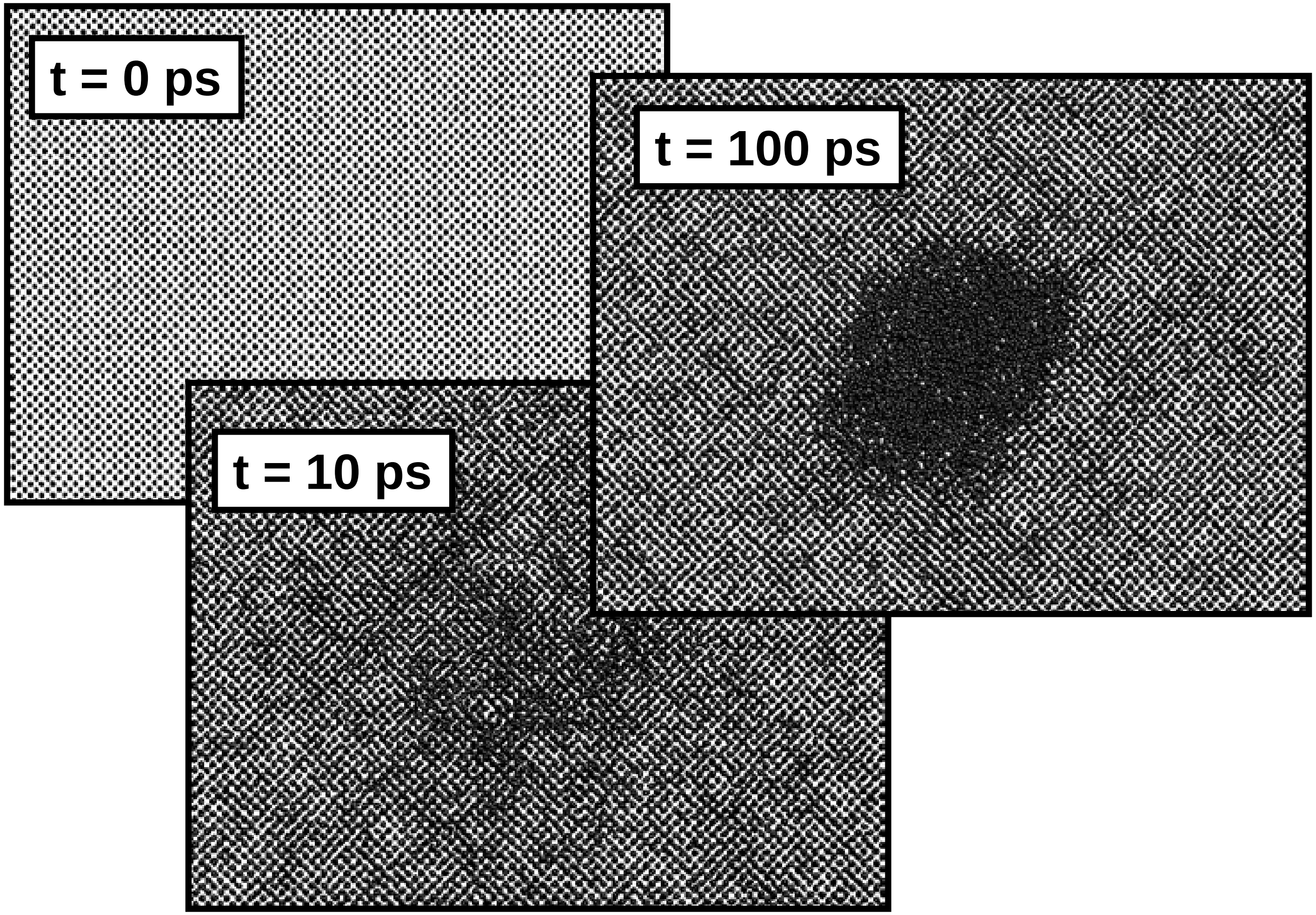
dE/dz = 160 KeV/nm



Mo

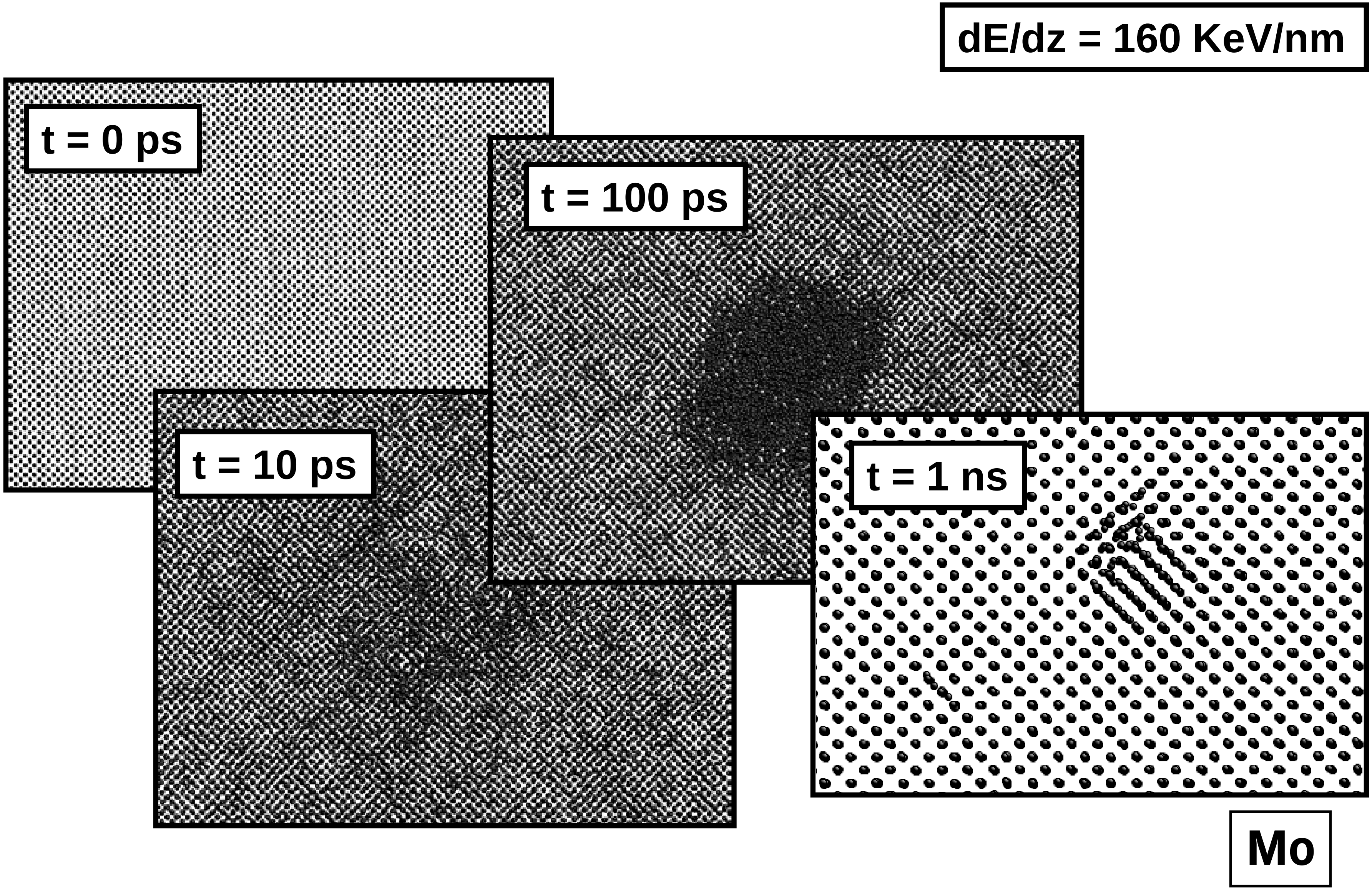
Defect formation at heating/melting of ionic subsystem

dE/dz = 160 KeV/nm

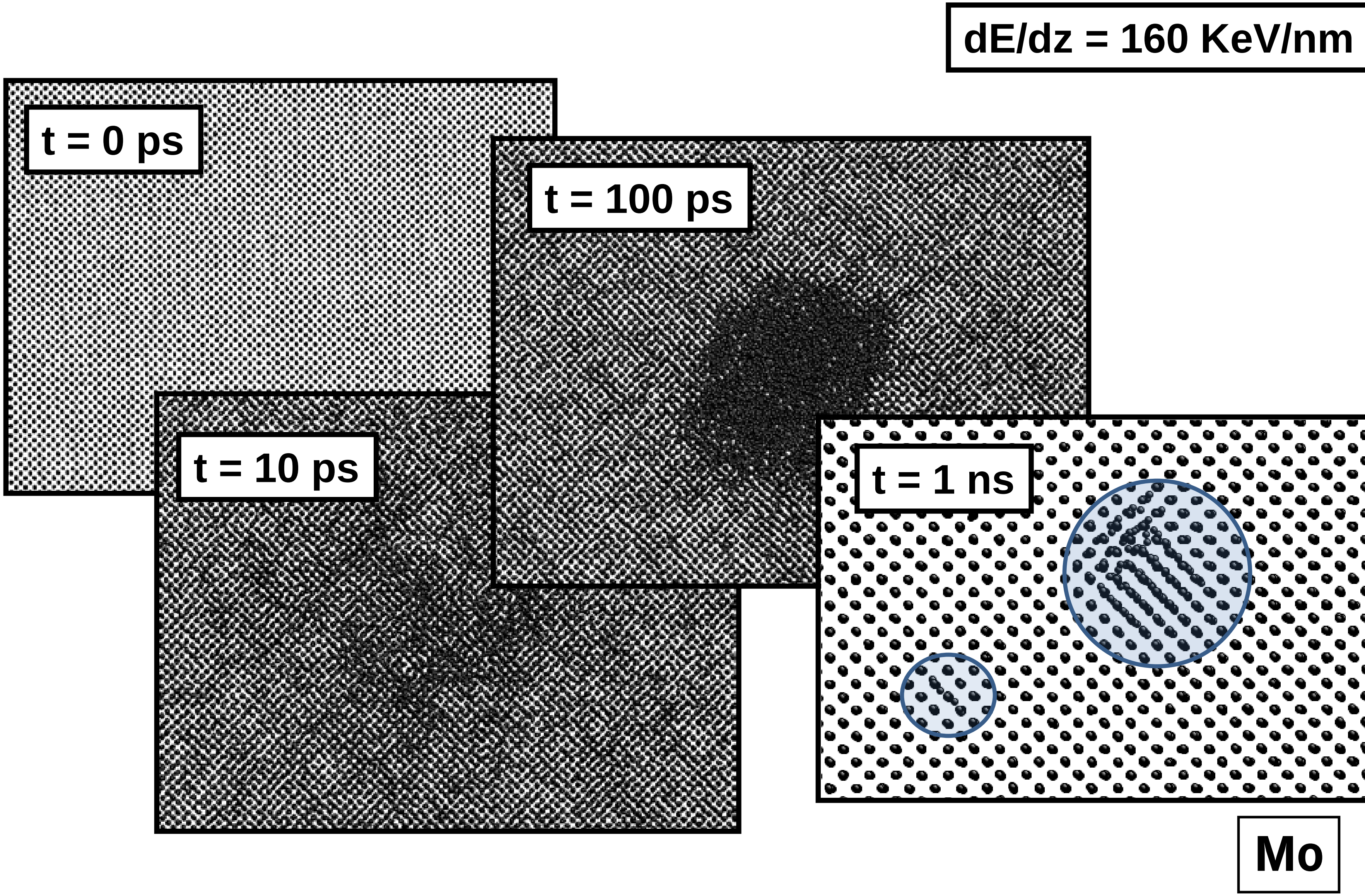


Mo

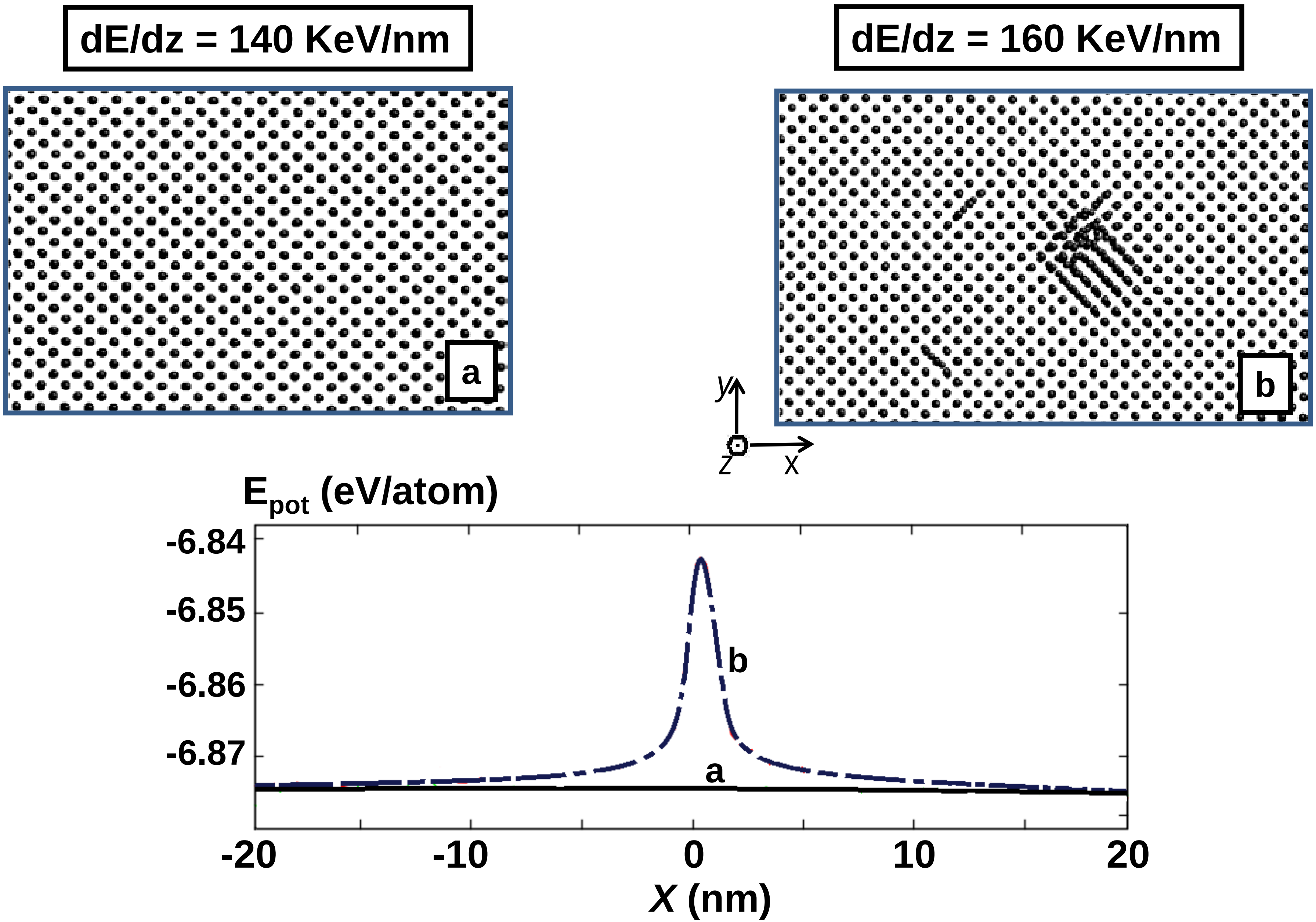
Defect formation at heating/melting of ionic subsystem



Defect formation at heating/melting of ionic subsystem



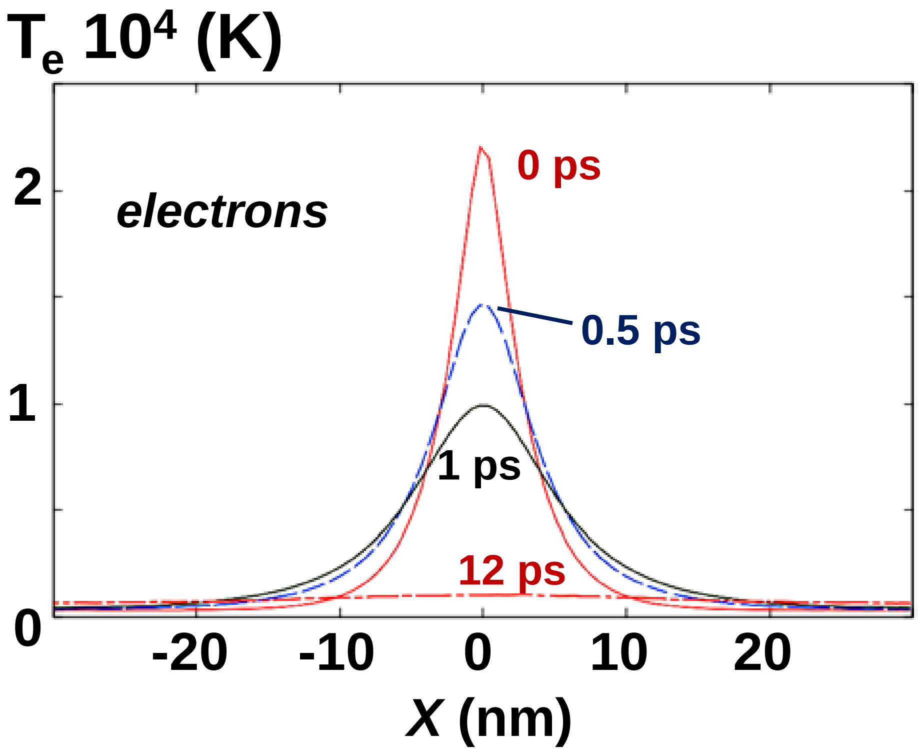
Defect formation at heating/melting of ionic subsystem



Simulation of track formation in U

Profiles of electronic and ionic temperatures

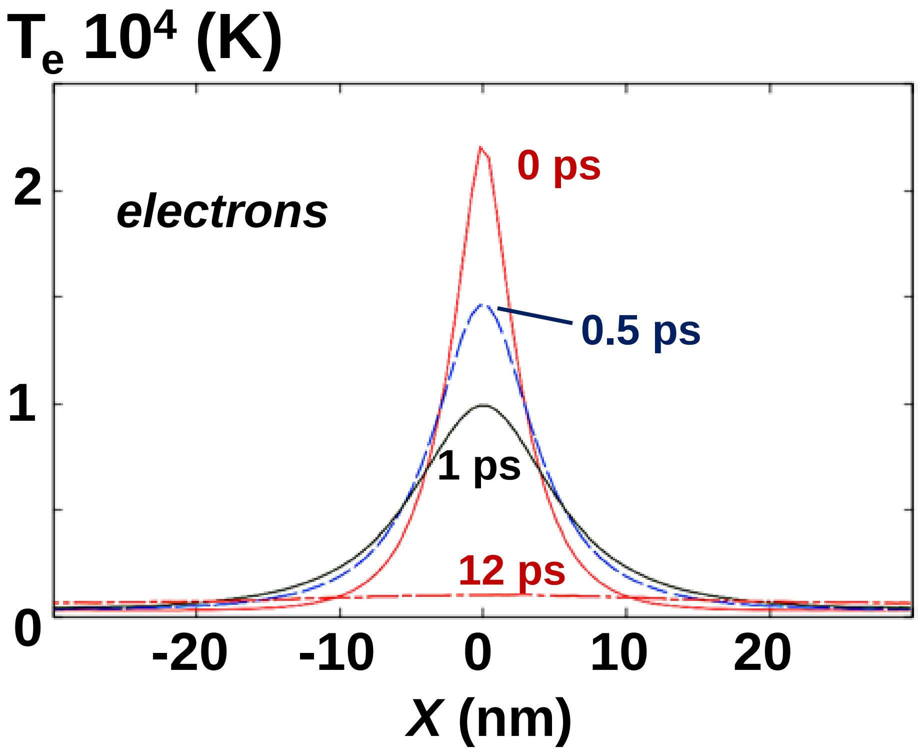
dE/dz = 28 KeV/nm

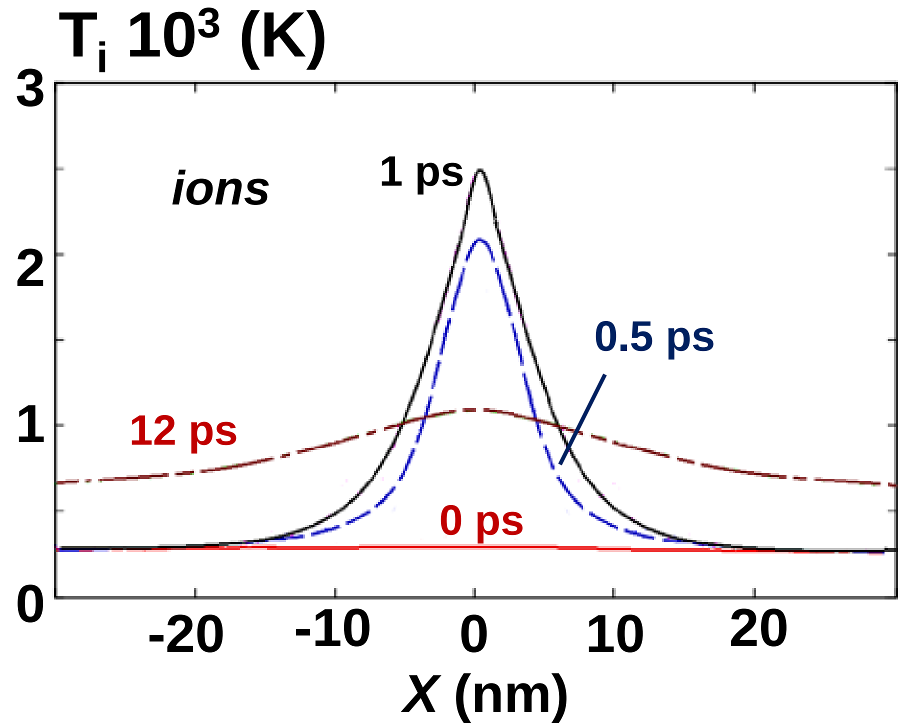


U

Profiles of electronic and ionic temperatures

dE/dz = 28 KeV/nm

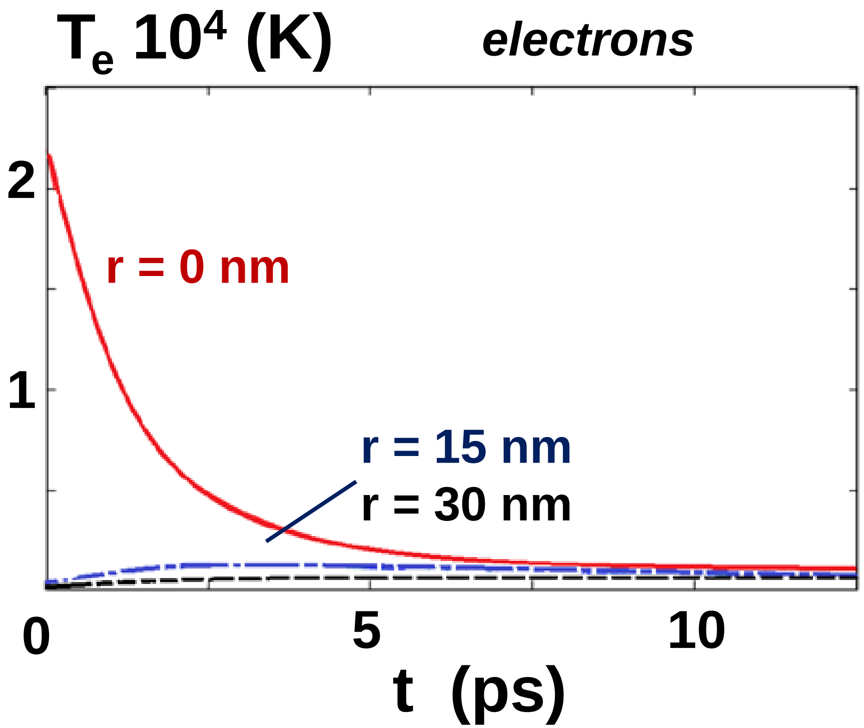


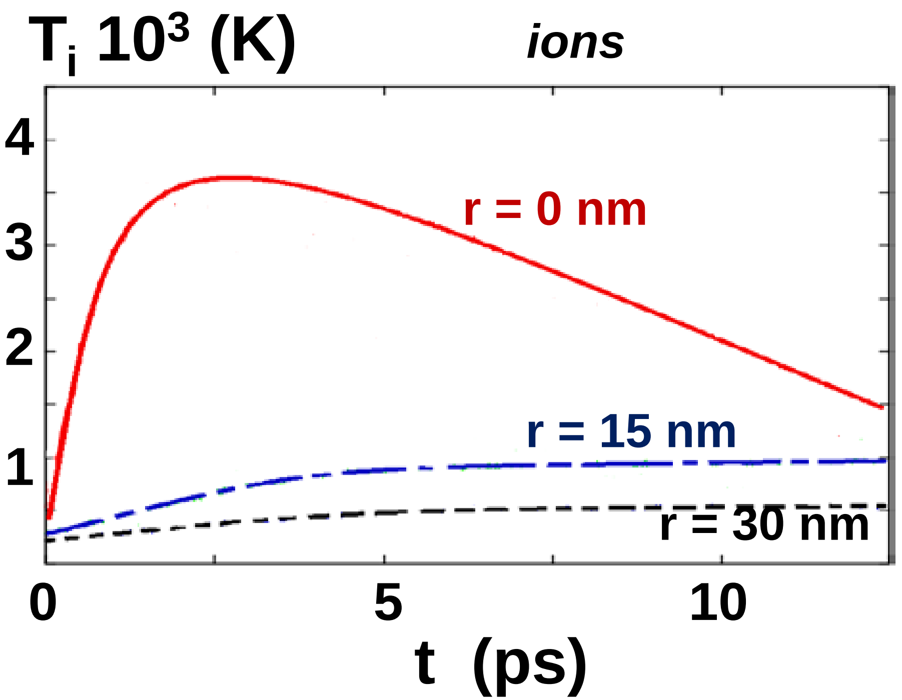


U

Dependence of temperature on time

dE/dz = 28 KeV/nm

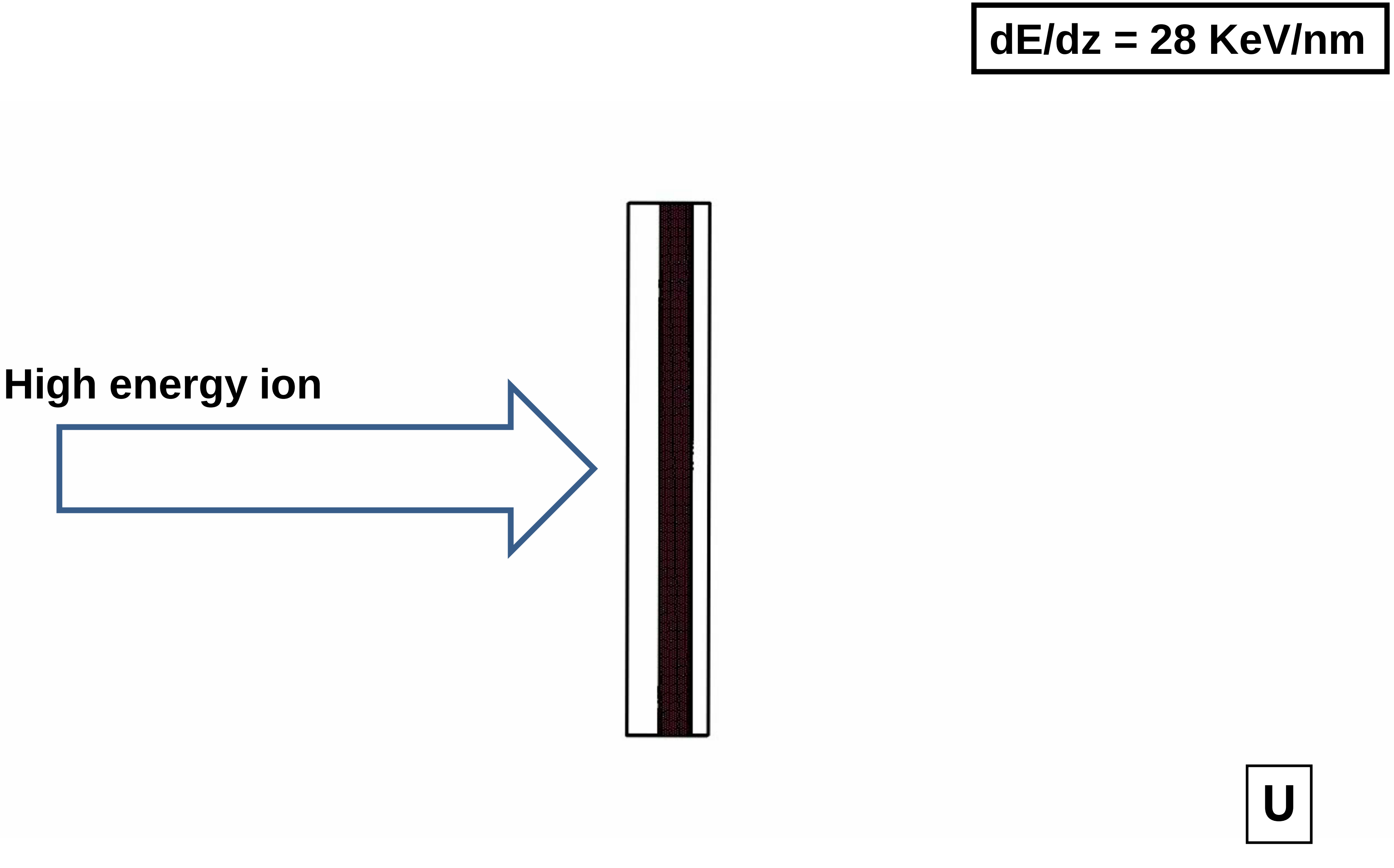




U

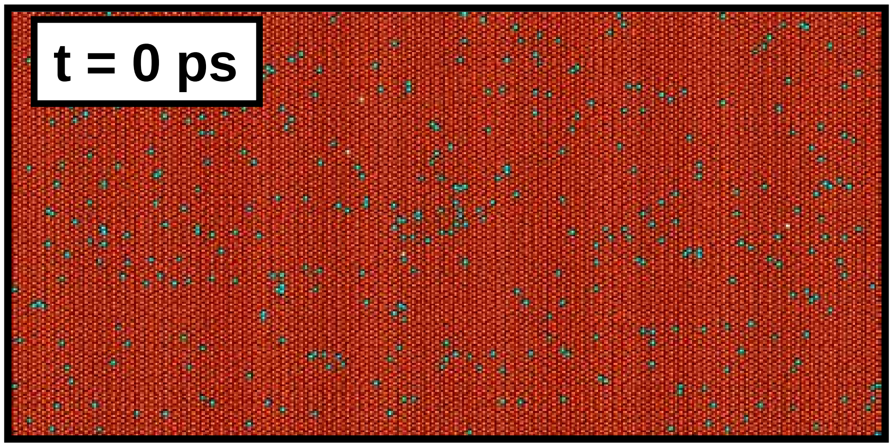
Образование дефектов при нагреве/плавлении ионной подсистемы

(*атомы раскрашены в соответствии с координационным числом*)



Образование дефектов при нагреве/плавлении ионной подсистемы

(*атомы раскрашены в соответствии с координационным числом*)



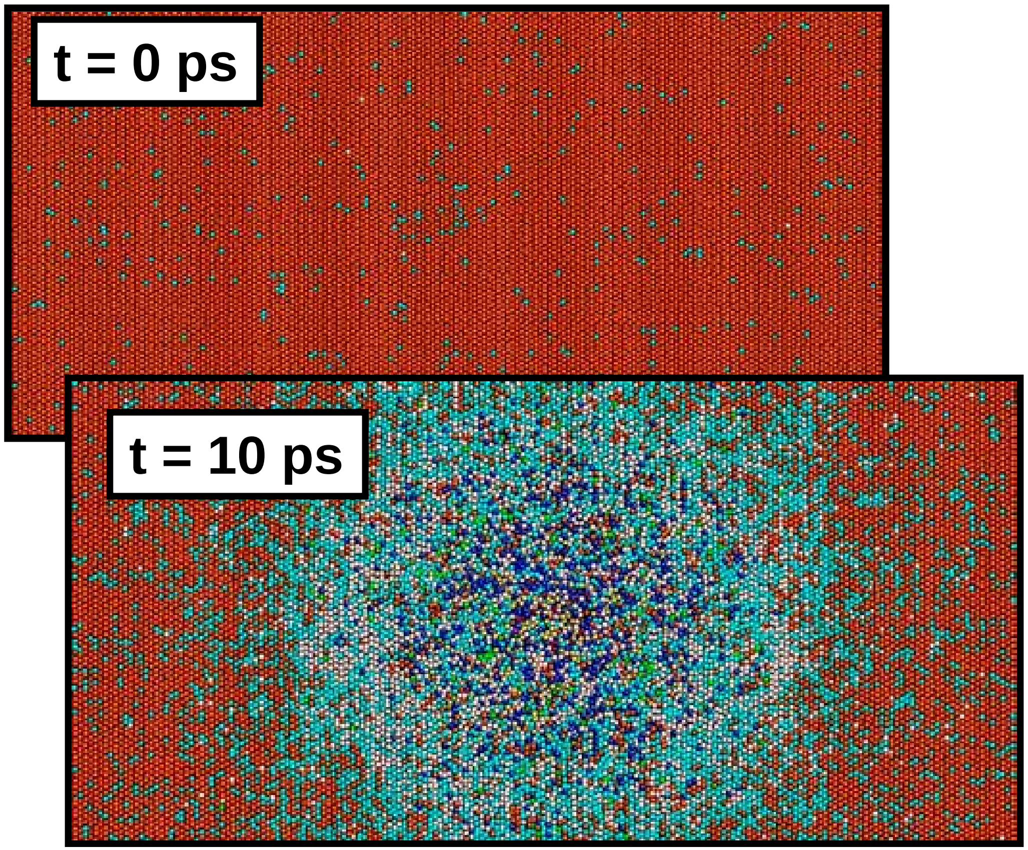
Образование дефектов при нагреве/плавлении ионной подсистемы

(*атомы раскрашены в соответствии с координационным числом*)

α-U

γ-U

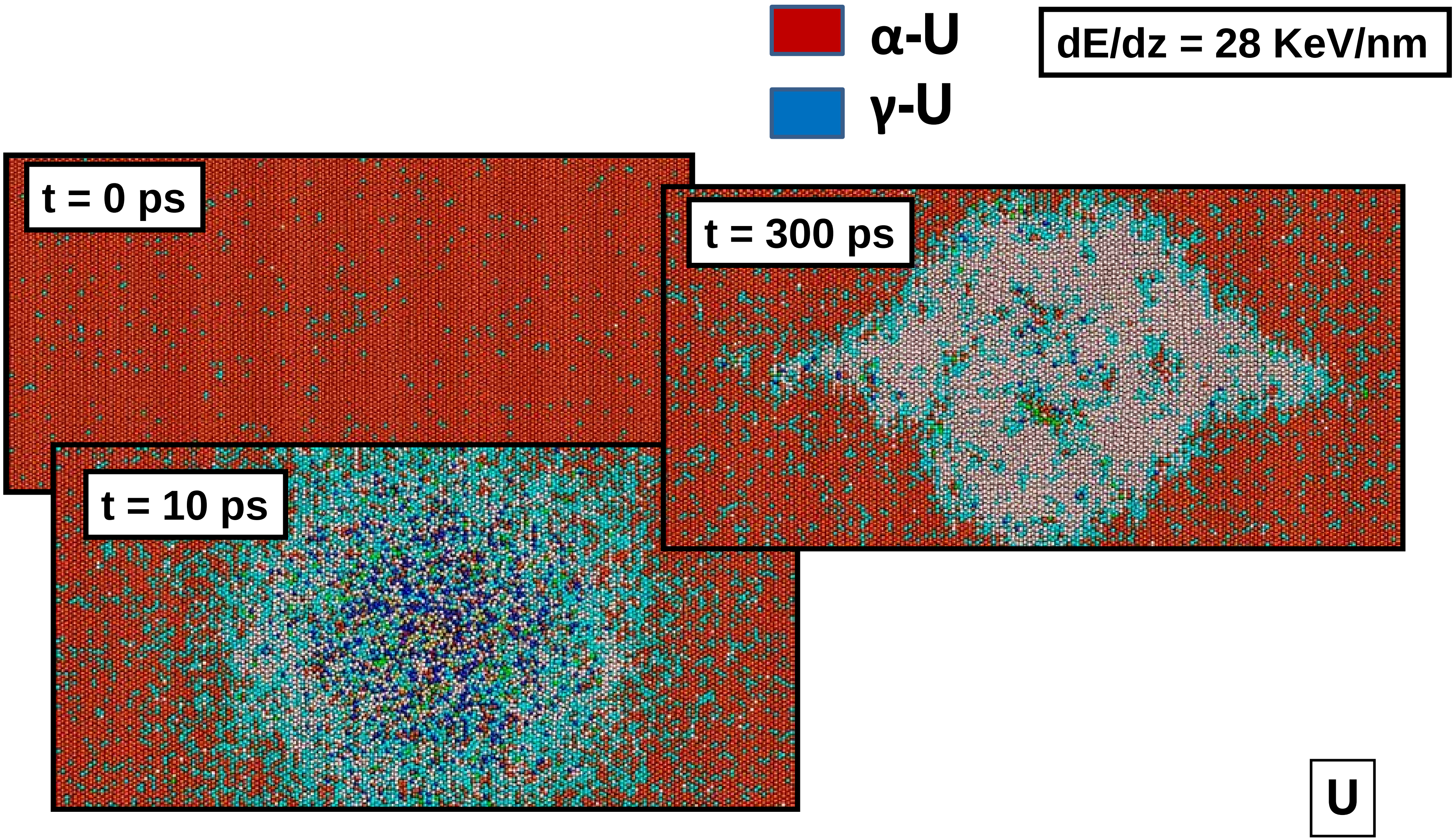
dE/dz = 28 KeV/nm



U

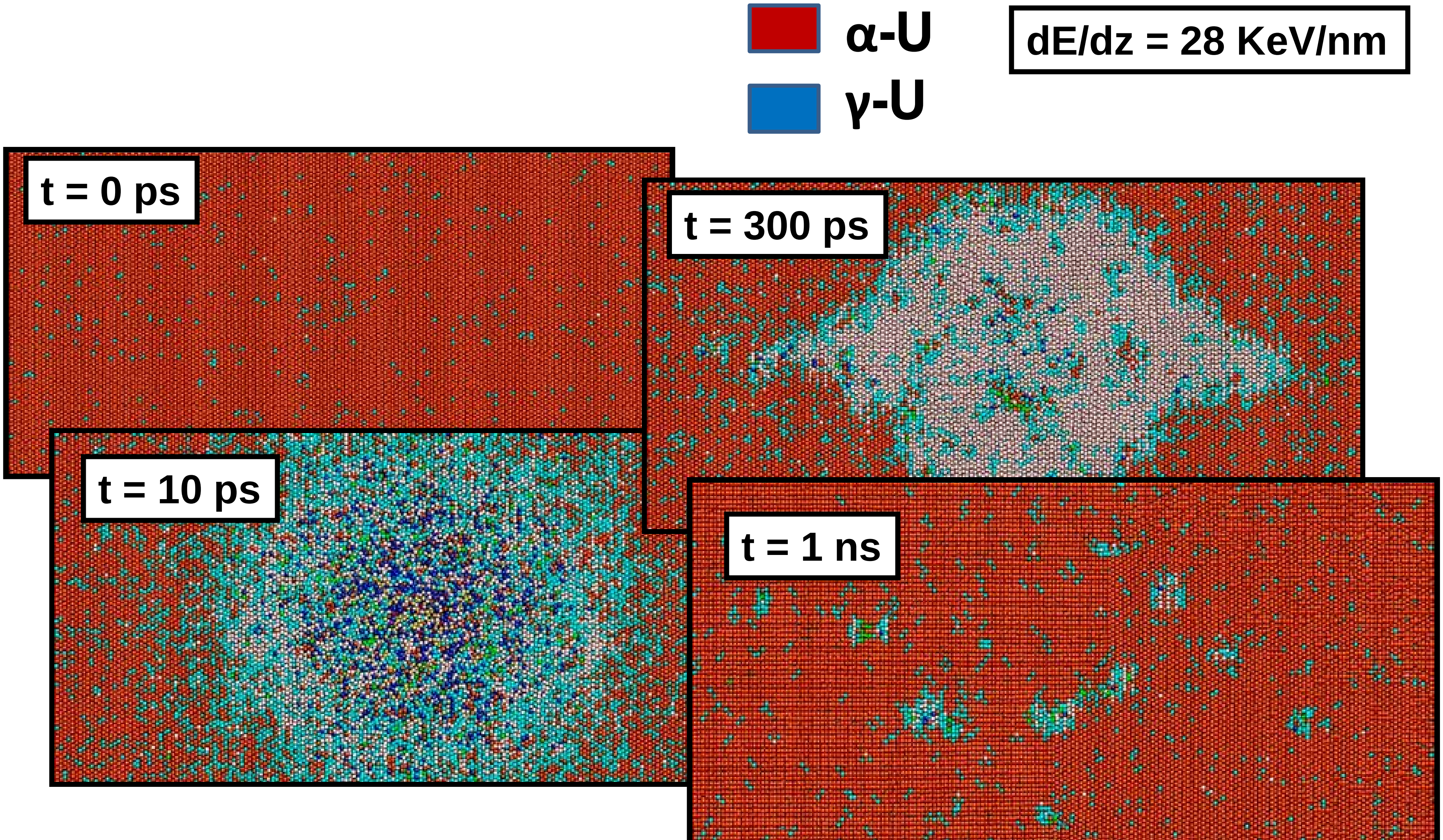
Образование дефектов при нагреве/плавлении ионной подсистемы

(*атомы раскрашены в соответствии с координационным числом*)

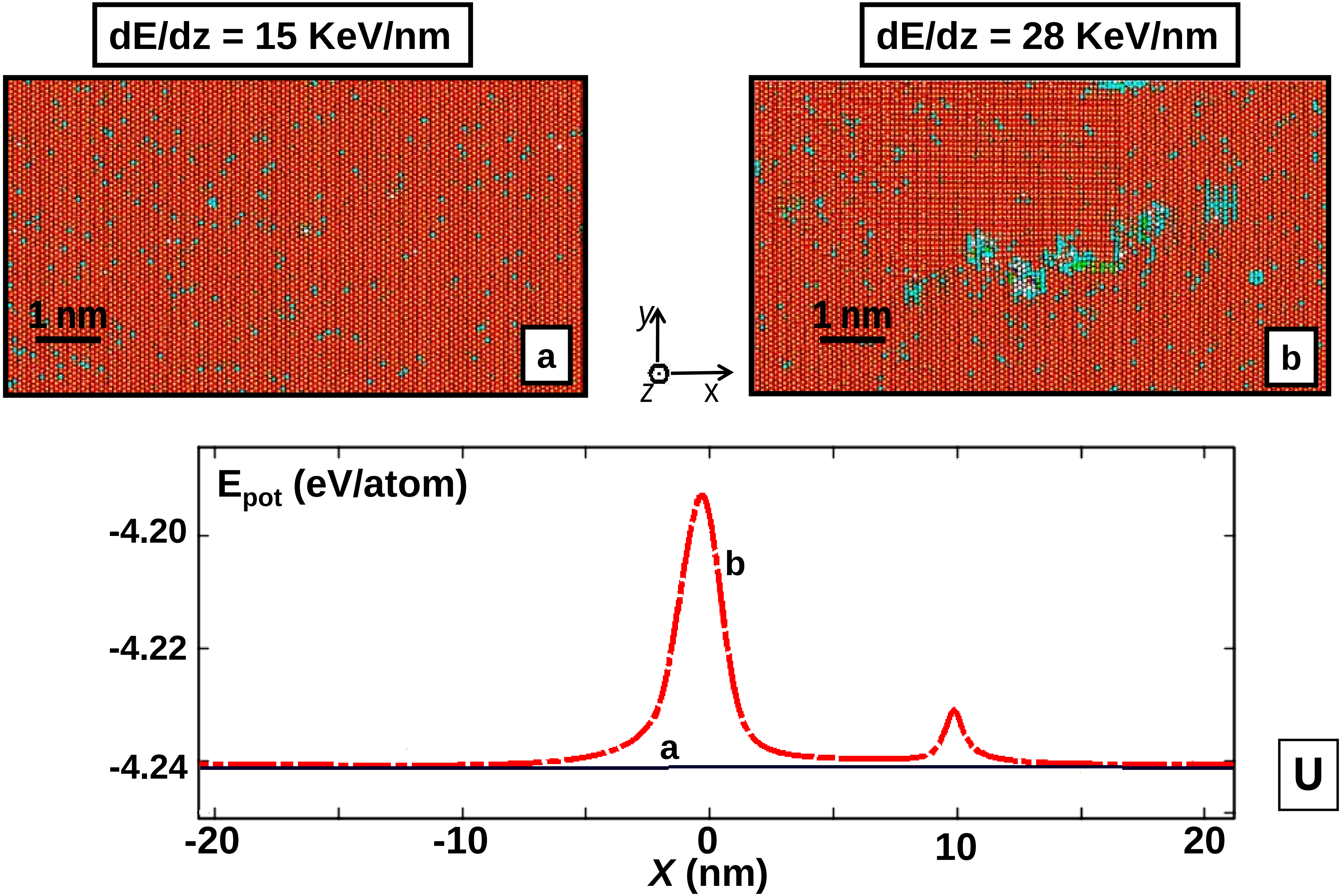


Образование дефектов при нагреве/плавлении ионной подсистемы

(*атомы раскрашены в соответствии с координационным числом*)



Defect formation at heating/melting of ionic subsystem

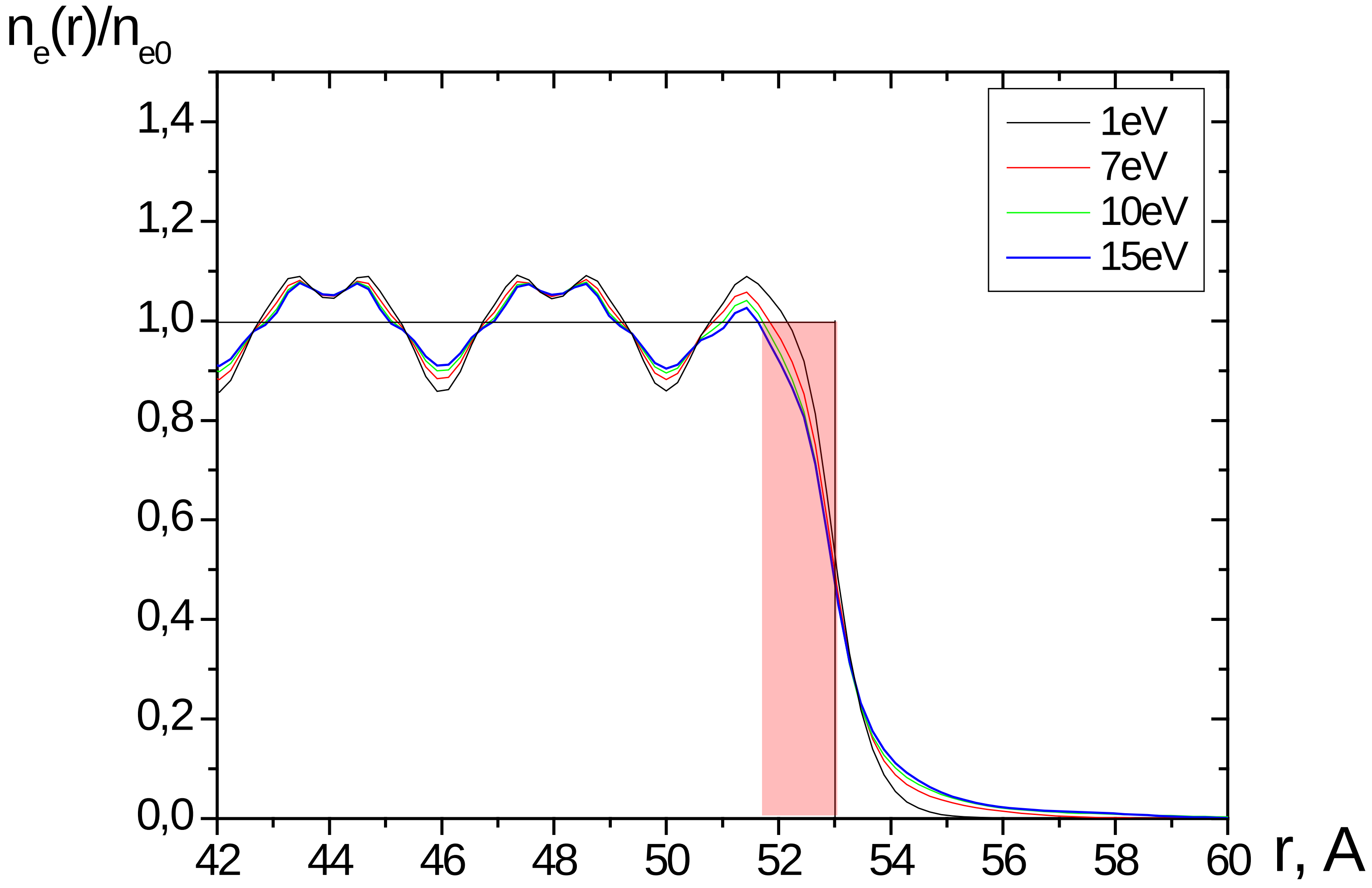


Conclusions

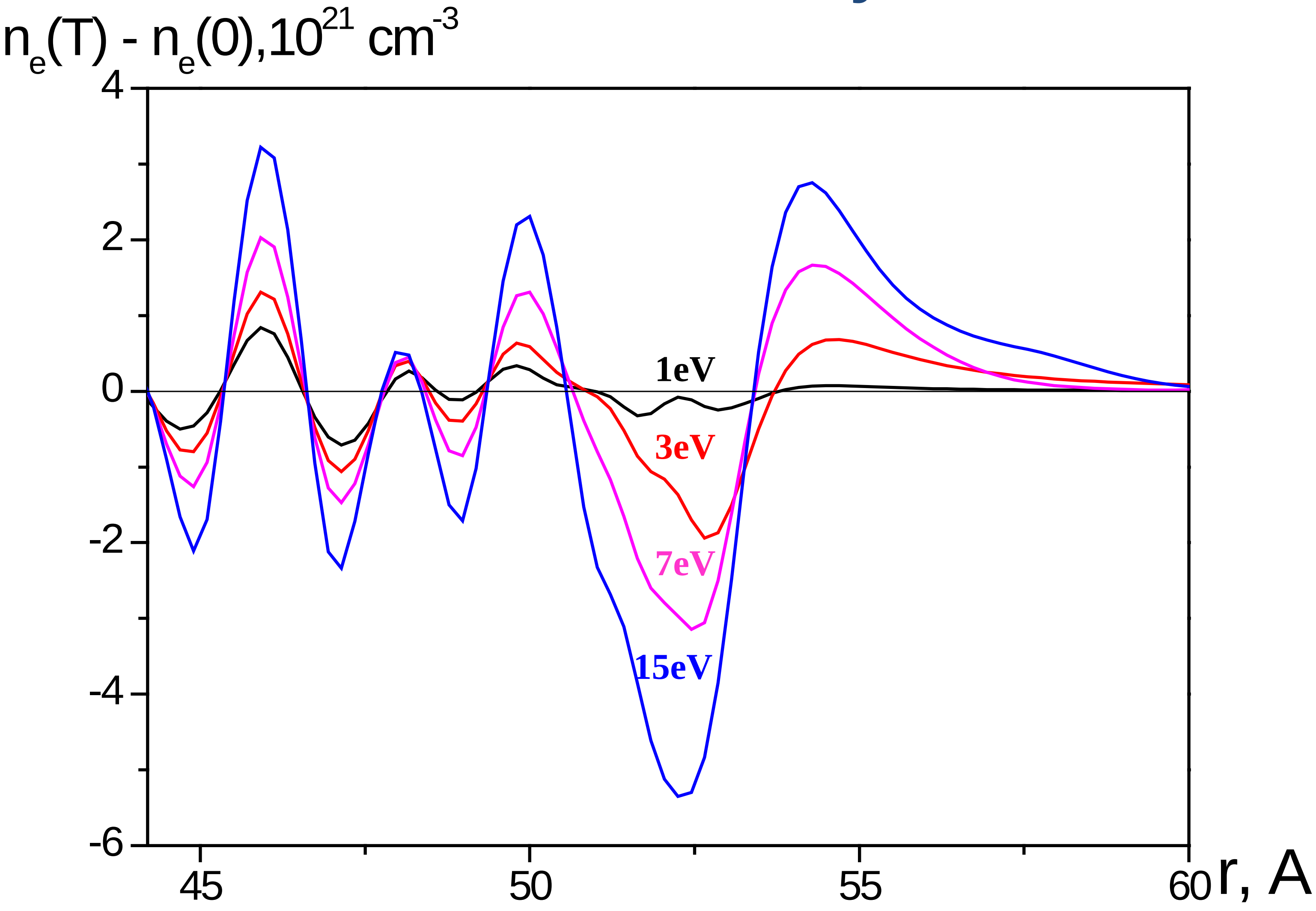
A new approach is developed
to model and simulate
***two-temperature* warm dense matter**
relaxation

The approach is used to study
some examples of
laser ablation and tracks formation

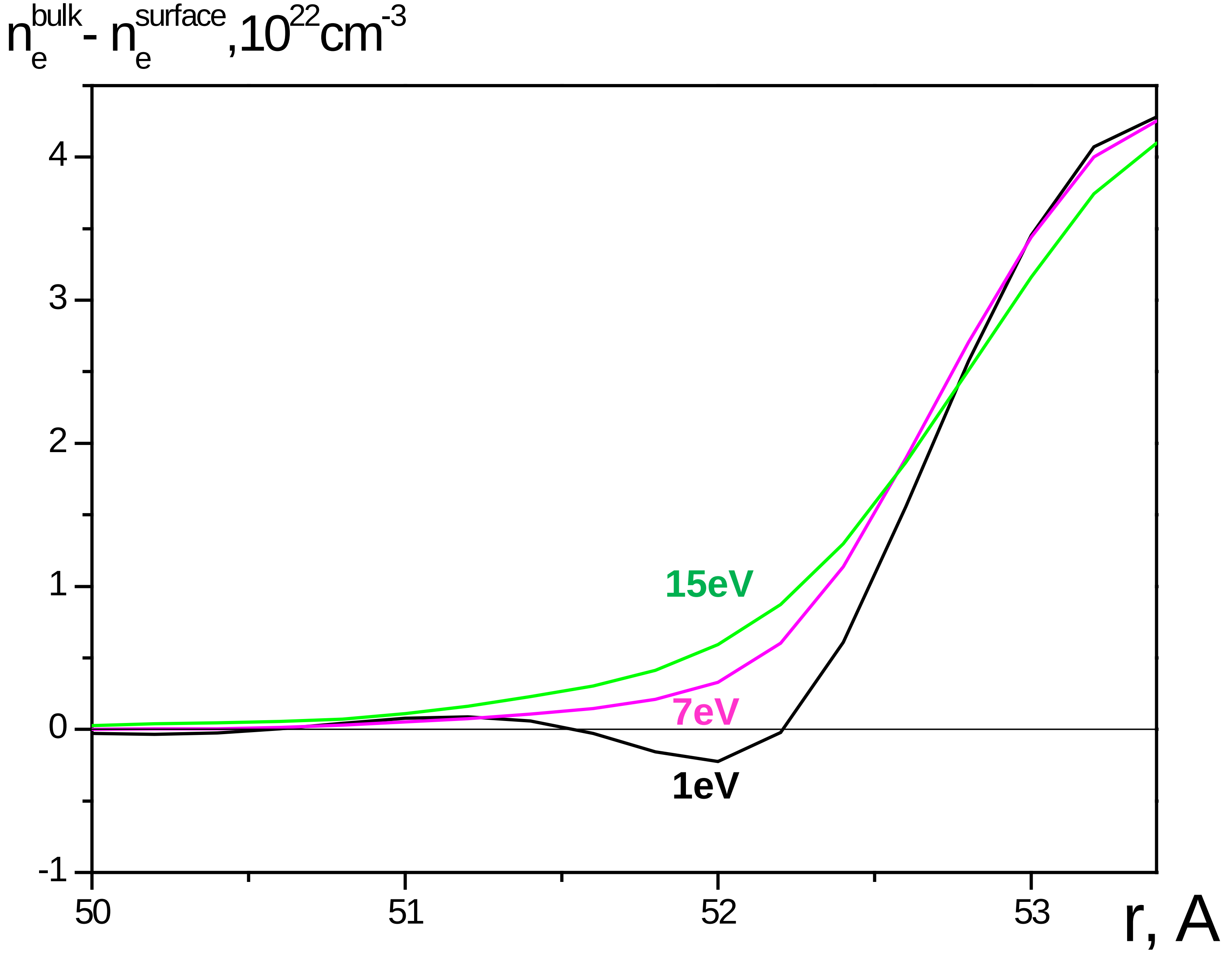
Distribution of electron density in Aluminium.



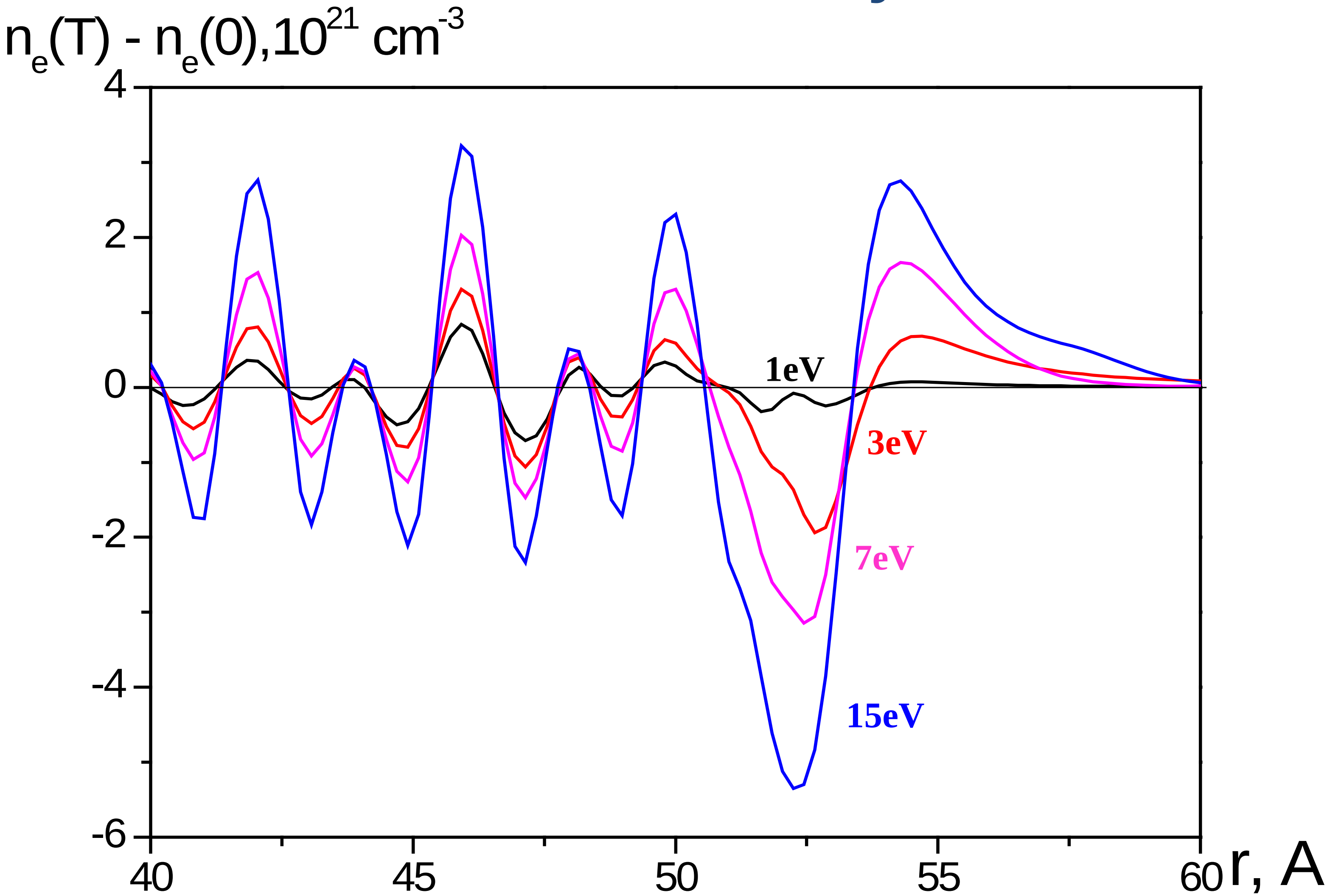
Distribution of electron density in Aluminium.



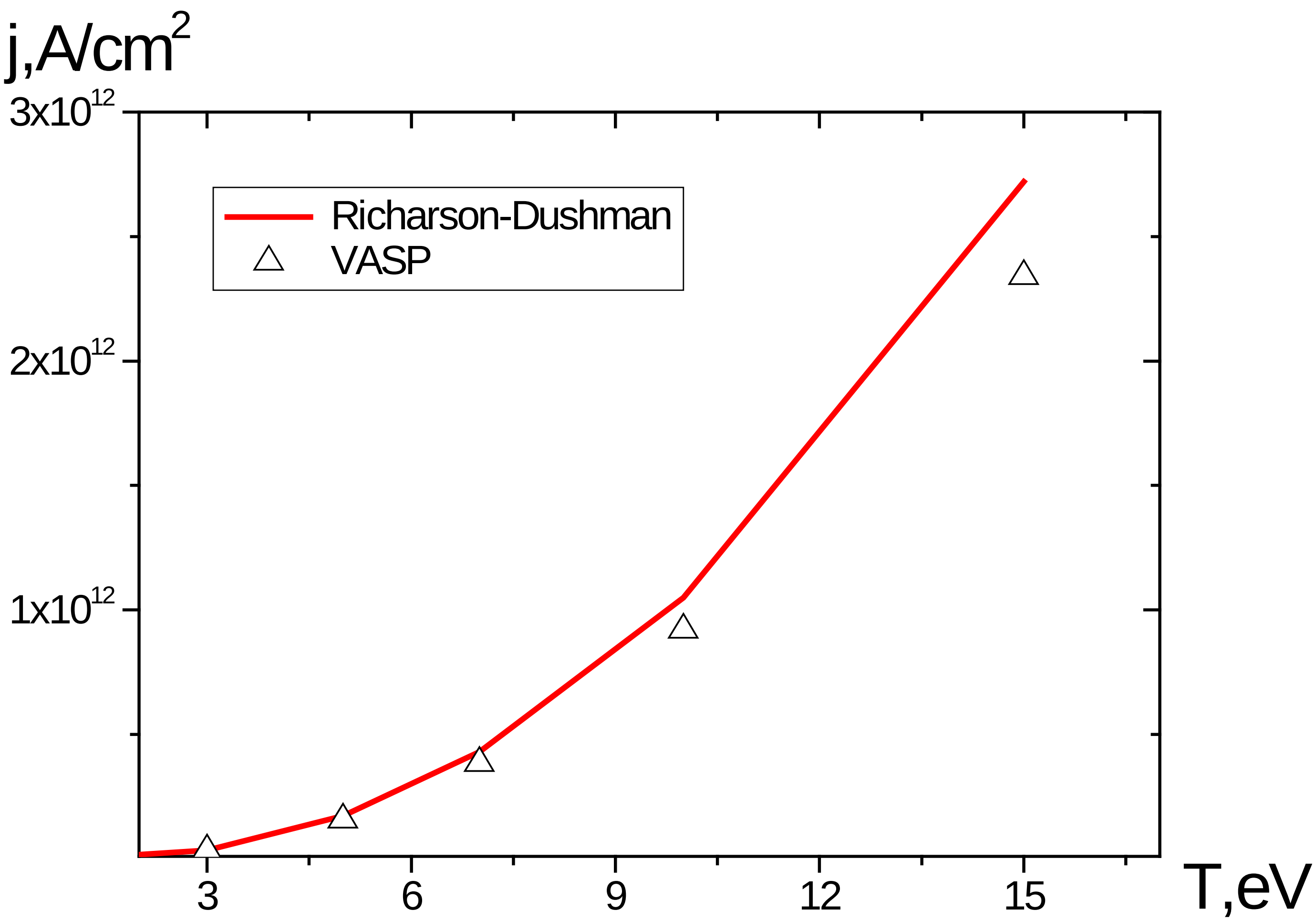
Distribution of electron density in Aluminium.



Distribution of electron density in Aluminium.



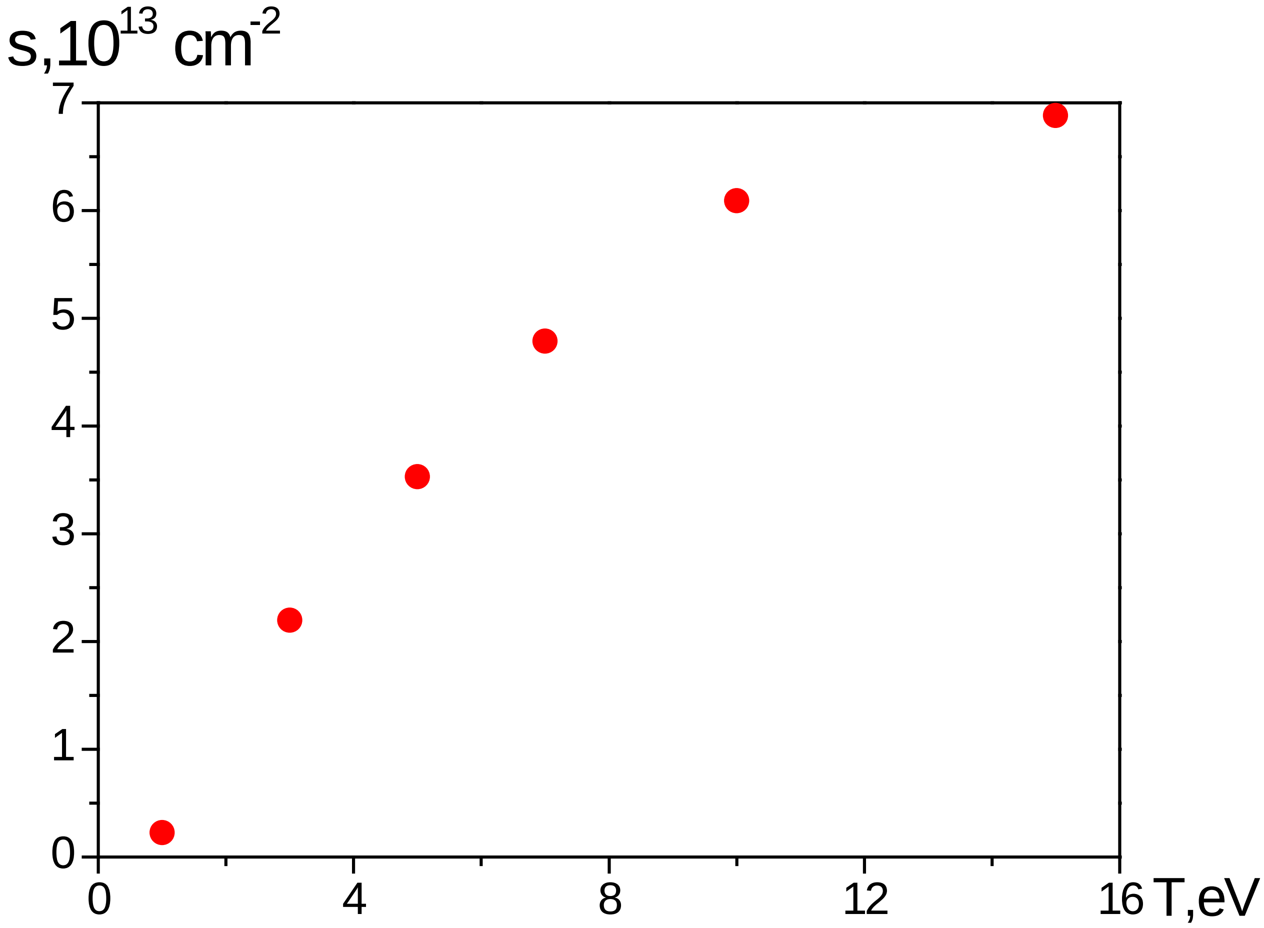
Density of current of thermal emission in Aluminium.



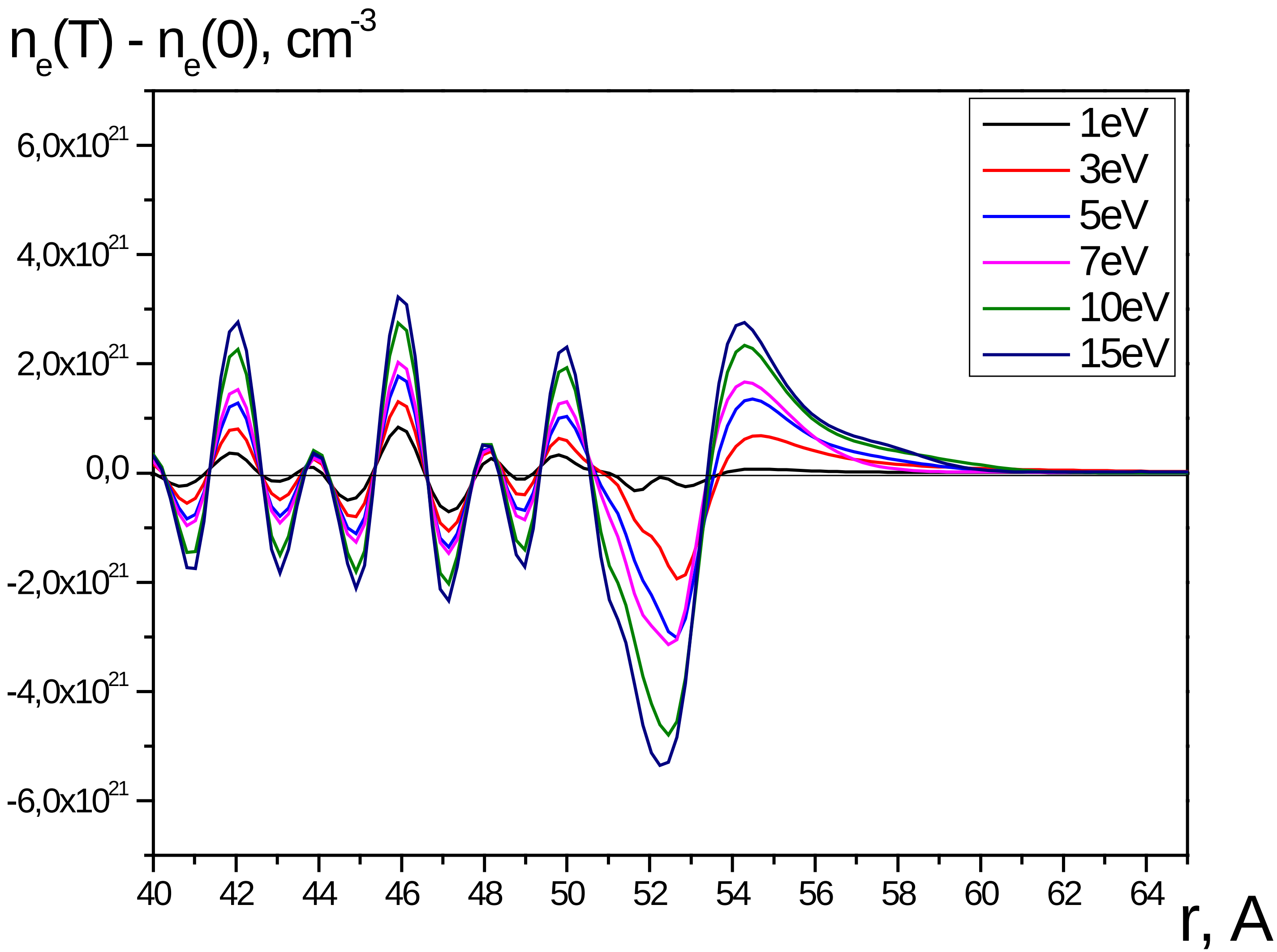
$$j = \frac{4\pi m e}{h^3} \int_{\varepsilon_F + A}^{\infty} \frac{[\varepsilon - (\varepsilon_F + A)]}{1 + \exp((\varepsilon - \varepsilon_F)/kT)} d\varepsilon$$

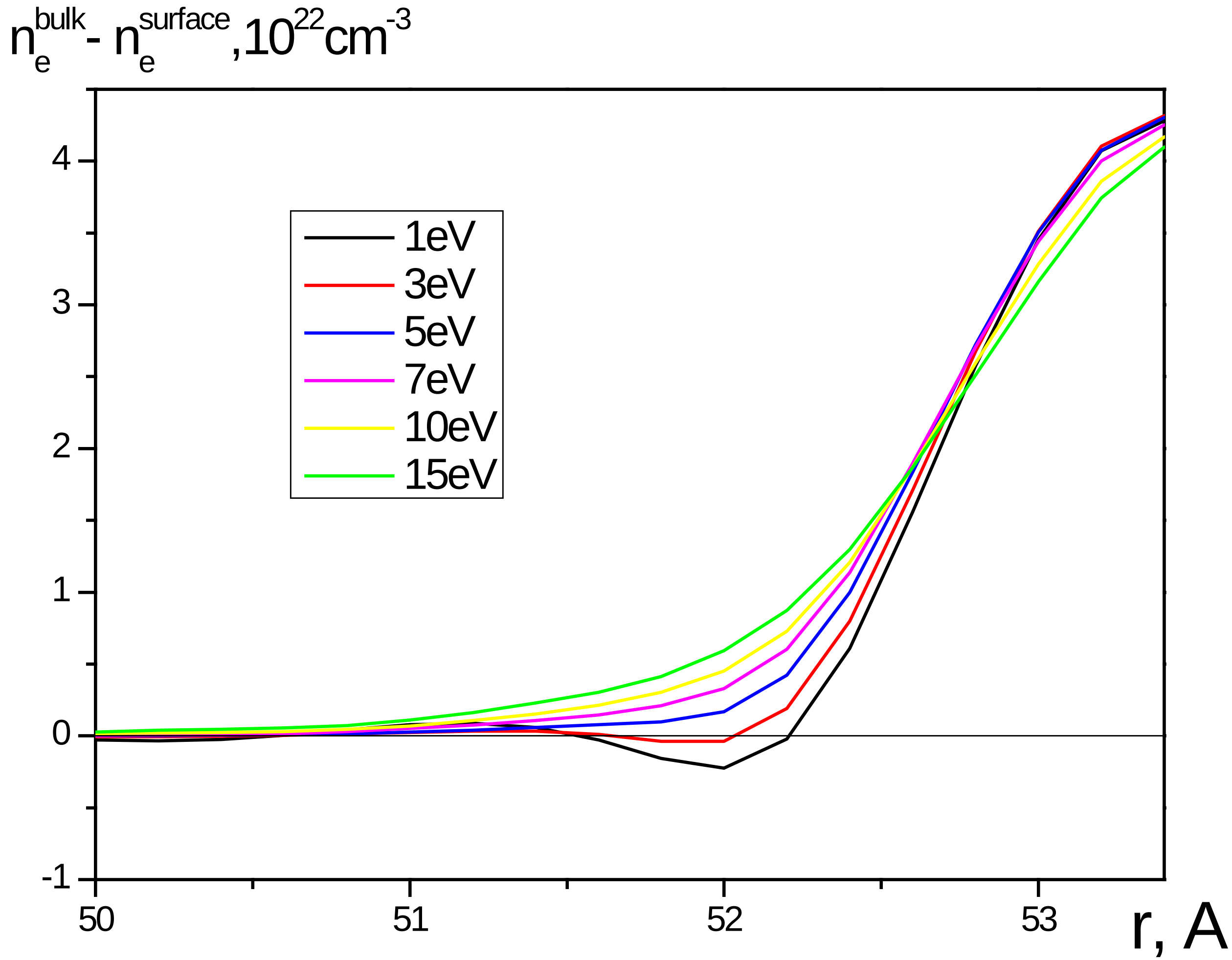
$$j_{R-D} = \frac{4\pi m e}{h^3} (kT)^2 \exp\left(-\frac{A}{kT}\right)$$

Surface density of uncompensated positive charge.

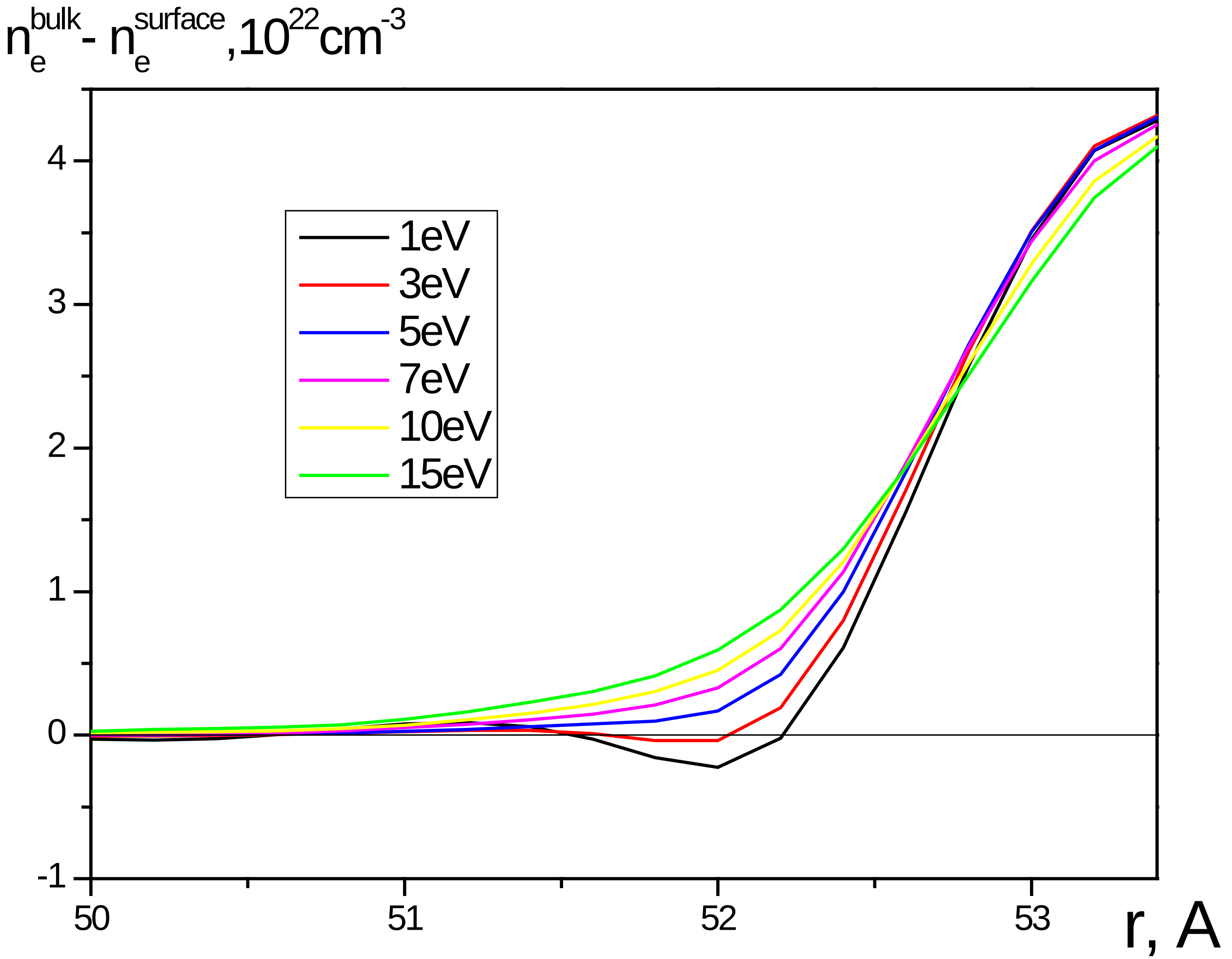


Distribution of electron density in Aluminium.





$n_e^{bulk} - n_e^{surface}$, $10^{22}cm^{-3}$
4
3
2
1
0
-1
1eV
3eV
5eV
7eV
10eV
15eV
50
51
52
53
r, A



$n_e^{bulk} - n_e^{surface}, 10^{22} cm^{-3}$
4
3
2
1
0
-1
1eV
3eV
5eV
7eV
10eV
15eV
50
51
52
53
r, A

Properties of electronic subsystem

Electronic subsystem:

$$\boxed{C_e(T_e)\cdot\rho_e\cdot\frac{\partial T_e}{\partial t}=\nabla(K_e(T_e)\cdot\nabla T_e)-g_p(T_e-T_i)+\nabla Q}$$

$$T_e >> T_i$$

Necessary

$$\boxed{\begin{array}{l} C_e(T_e) \\ K_e(T_e) \\ g_p(T_e-T_i) \end{array}}$$

?

Electronic subsystem:

$$\boxed{C_e(T_e)\cdot\rho_e\cdot\frac{\partial T_e}{\partial t}=\nabla(K_e(T_e)\cdot\nabla T_e)-g_p(T_e-T_i)+\nabla Q}$$

$$T_e >> T_i$$

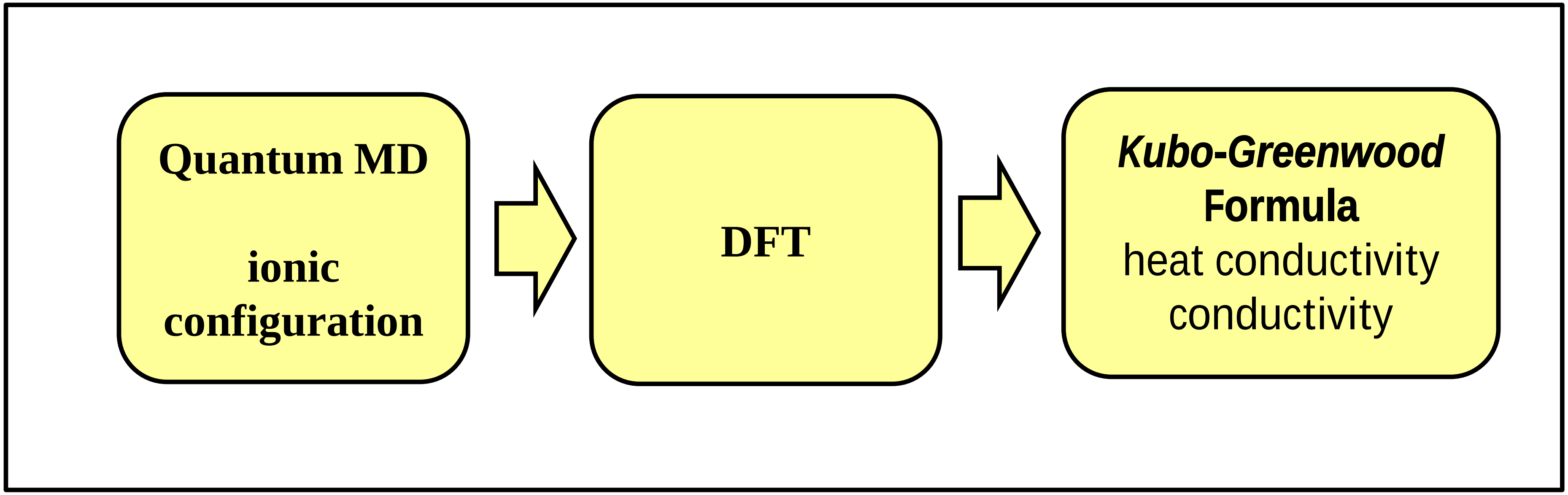
Necessary

$$\boxed{\begin{array}{l} C_e(T_e) \\ K_e(T_e) \\ g_p(T_e-T_i) \end{array}}$$

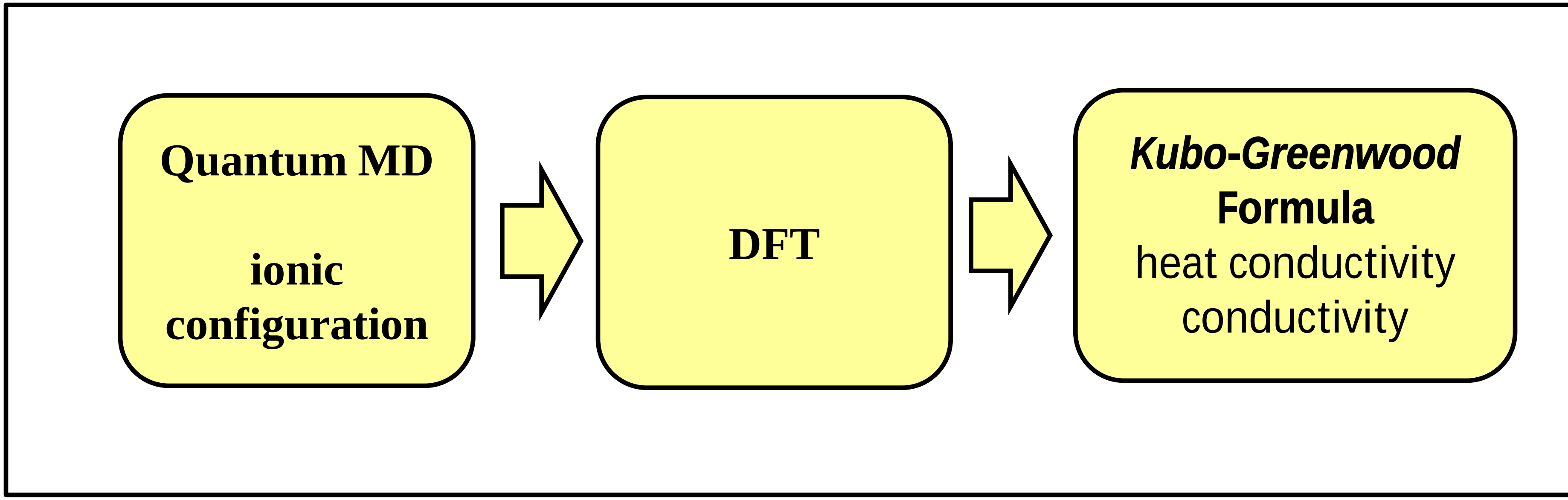
?

DFT calculation

Calculation of electron heat conductivity



Calculation of electron heat conductivity



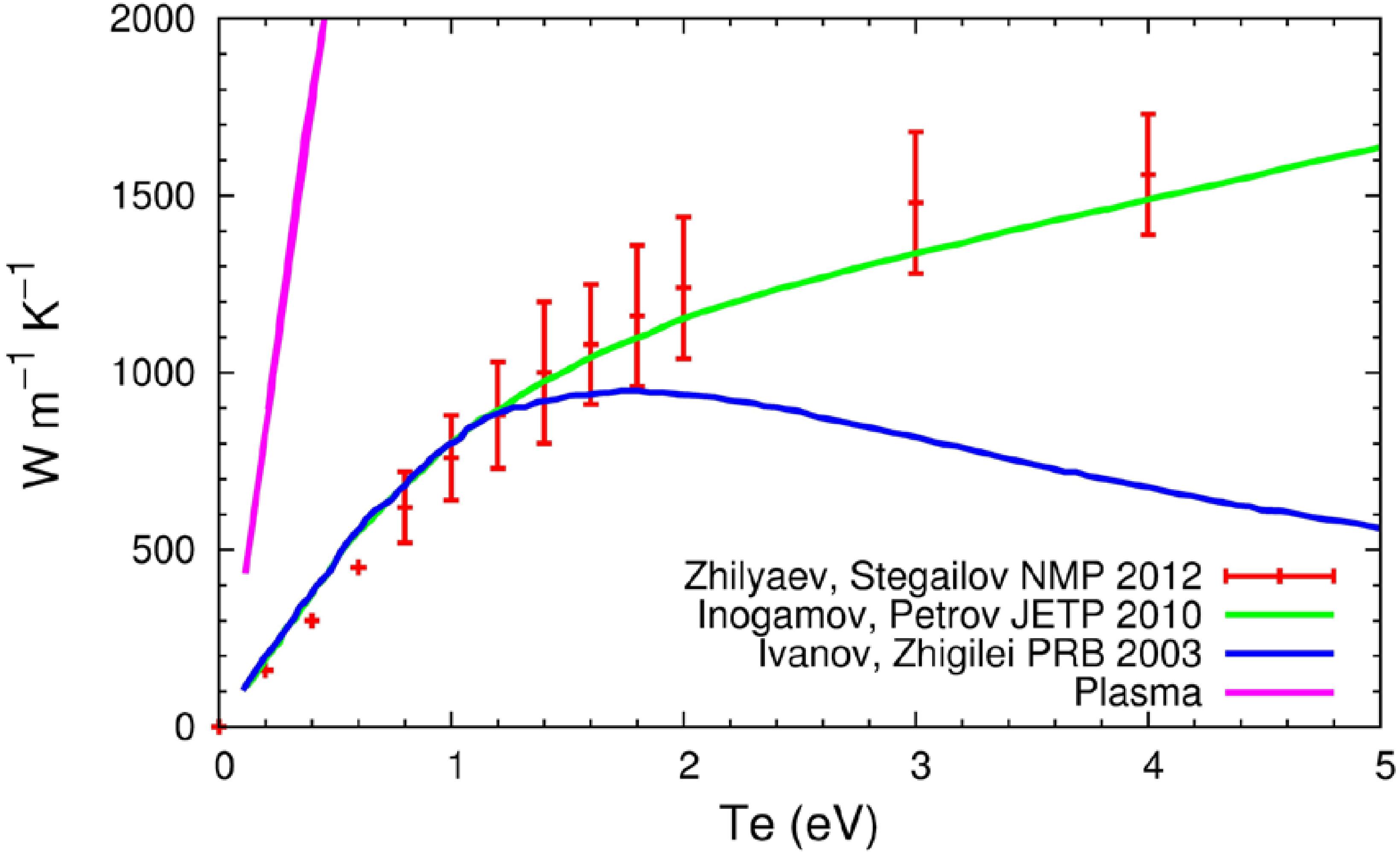
Kubo-Greenwood Formula

$$\mathcal{L}_{ij} = (-1)^{(i+j)} \frac{he^2}{\Omega} \lim_{\varepsilon\to 0} \frac{f(\varepsilon_k') - f(\varepsilon_k)}{\varepsilon}\ \delta(\varepsilon_k' - \varepsilon_k - \varepsilon) \times \sum_{k',\,k} \langle \psi_k | \hat{v} | \psi_{k'} \rangle \langle \psi_{k'} | \hat{v} | \psi_k \rangle\ (\varepsilon_k' - \mu)^{i-1} (\varepsilon_k - \mu)^{j-1}$$

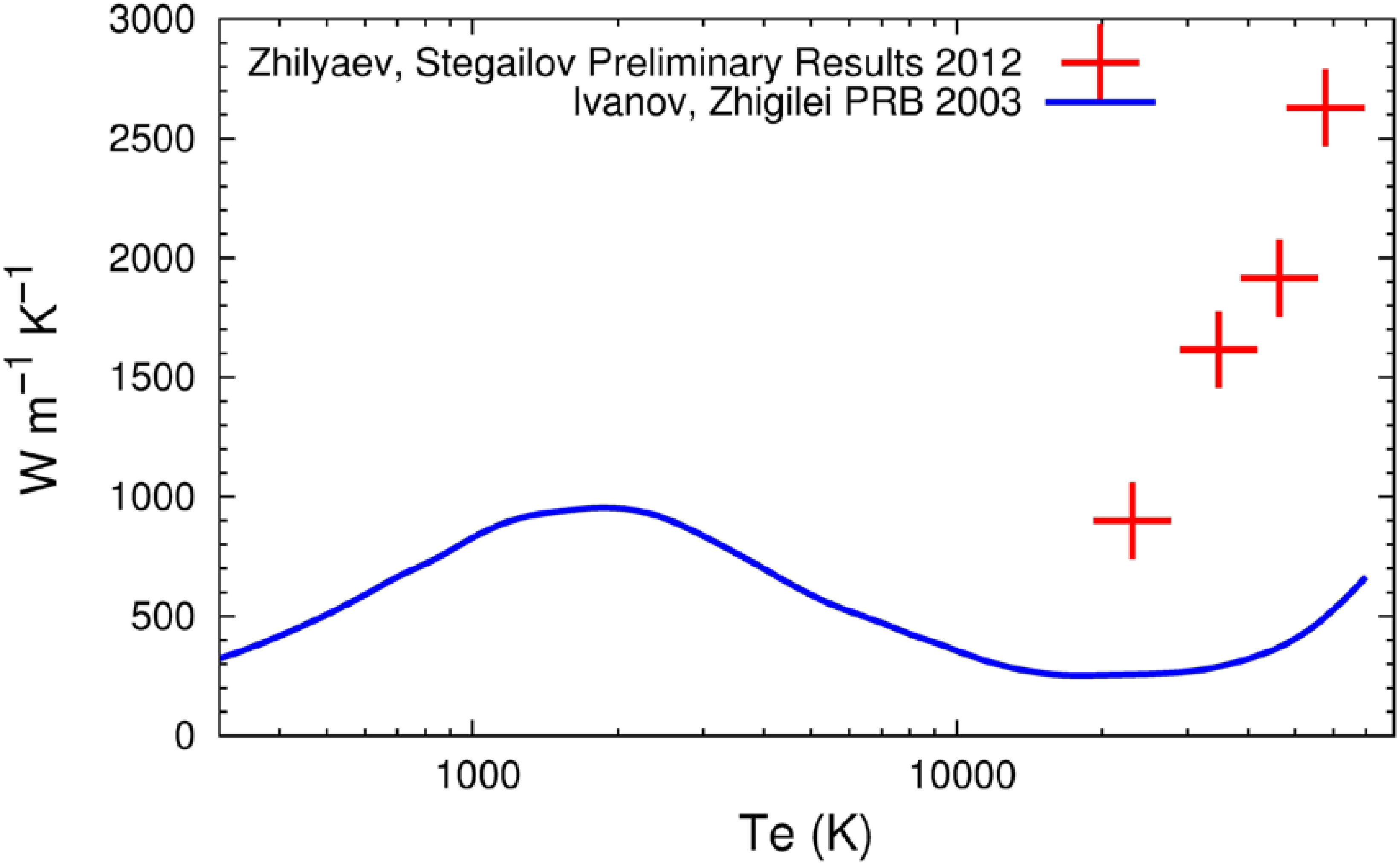
$$K = \frac{1}{eT}\left(\mathcal{L}_{22} - \frac{\mathcal{L}_{12}^2}{\mathcal{L}_{22}}\right);\ K(0) = \lim_{\omega\to 0} K(\omega)$$ ***Static heat conductivity***

$$\sigma = \mathcal{L}_{11};\ \sigma(0) = \lim_{\omega\to 0} \sigma(\omega)$$ ***Static conductivity***

Electron heat conductivity of liquid aluminum in two-temperature state

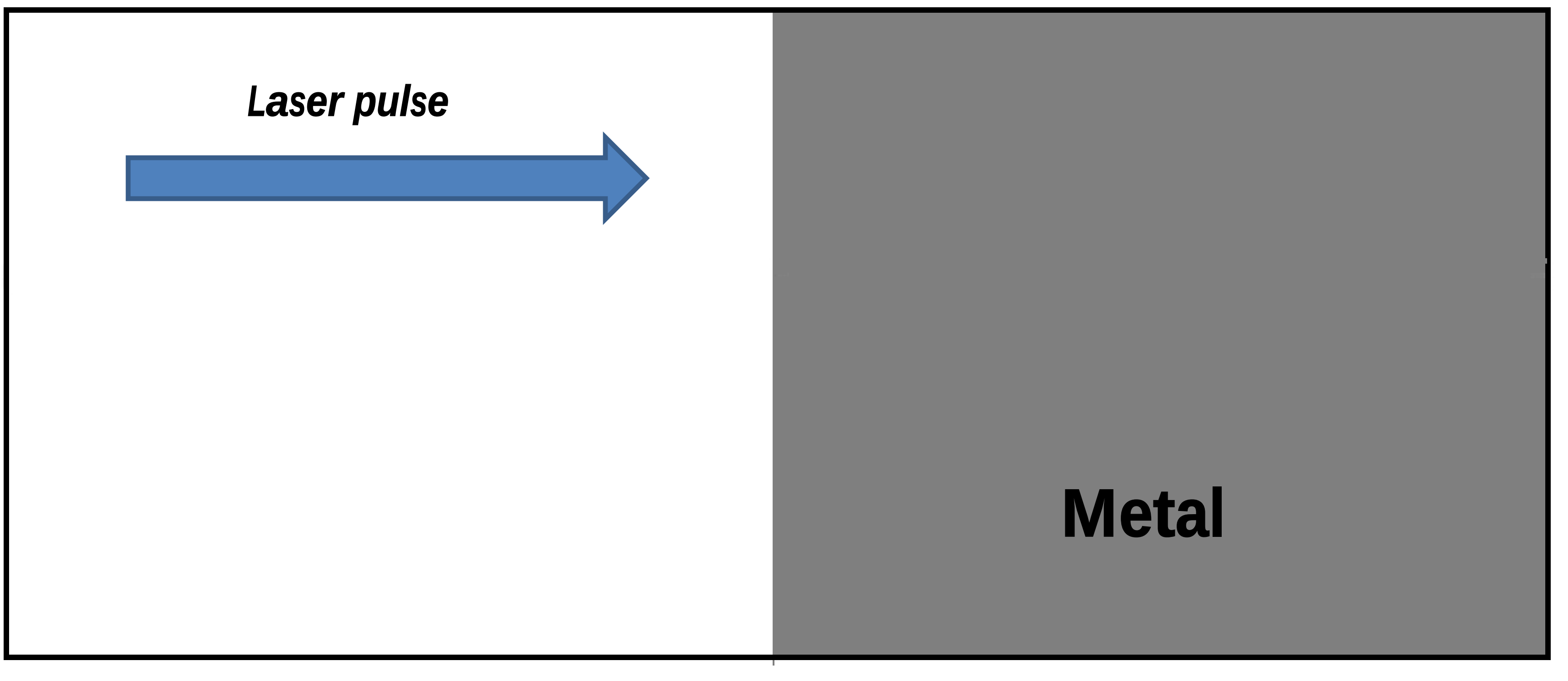


Electron heat conductivity of solid gold in two-temperature state

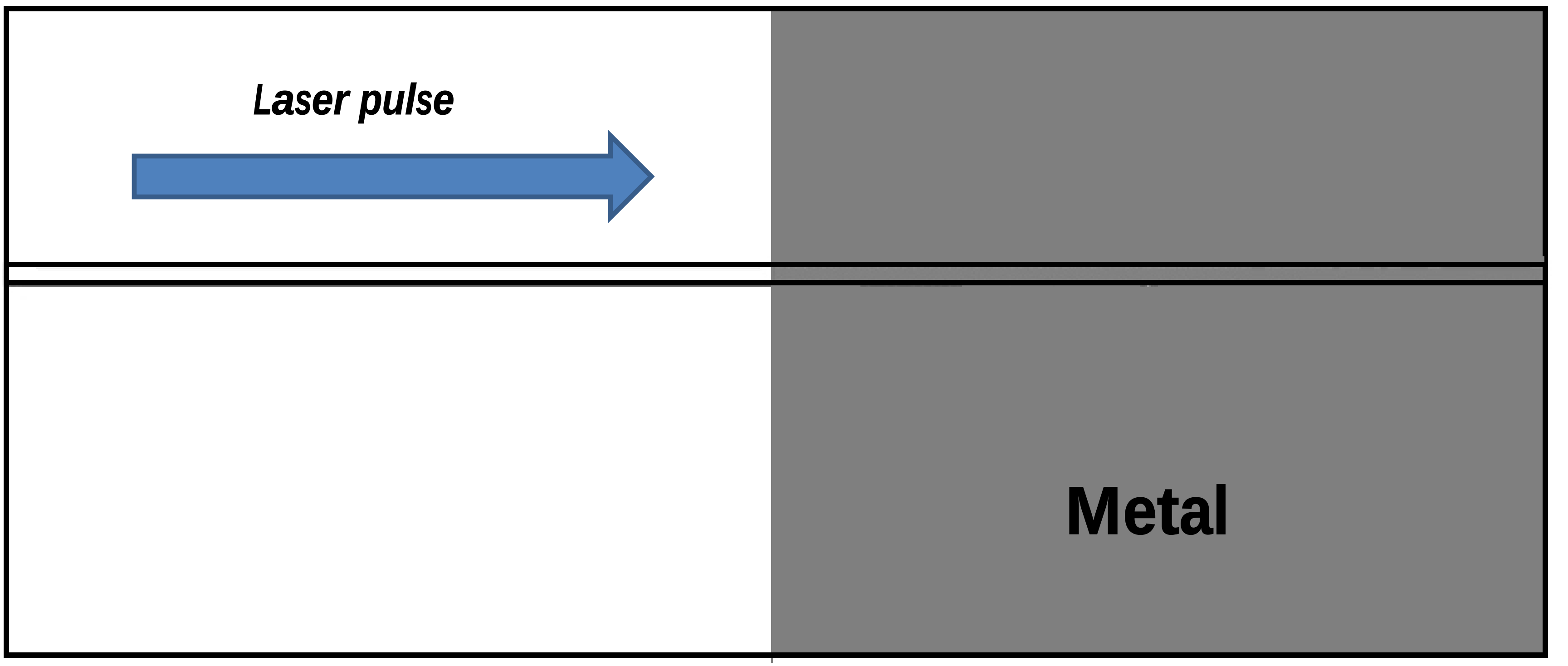


Laser ablation of gold

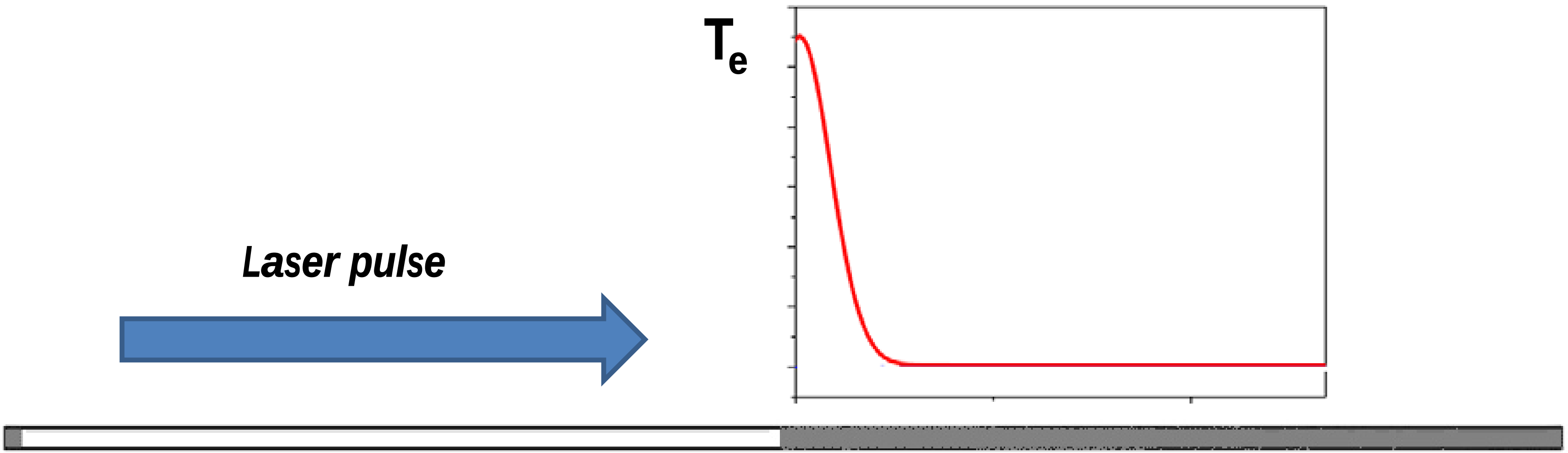
Ion structure in simulation box



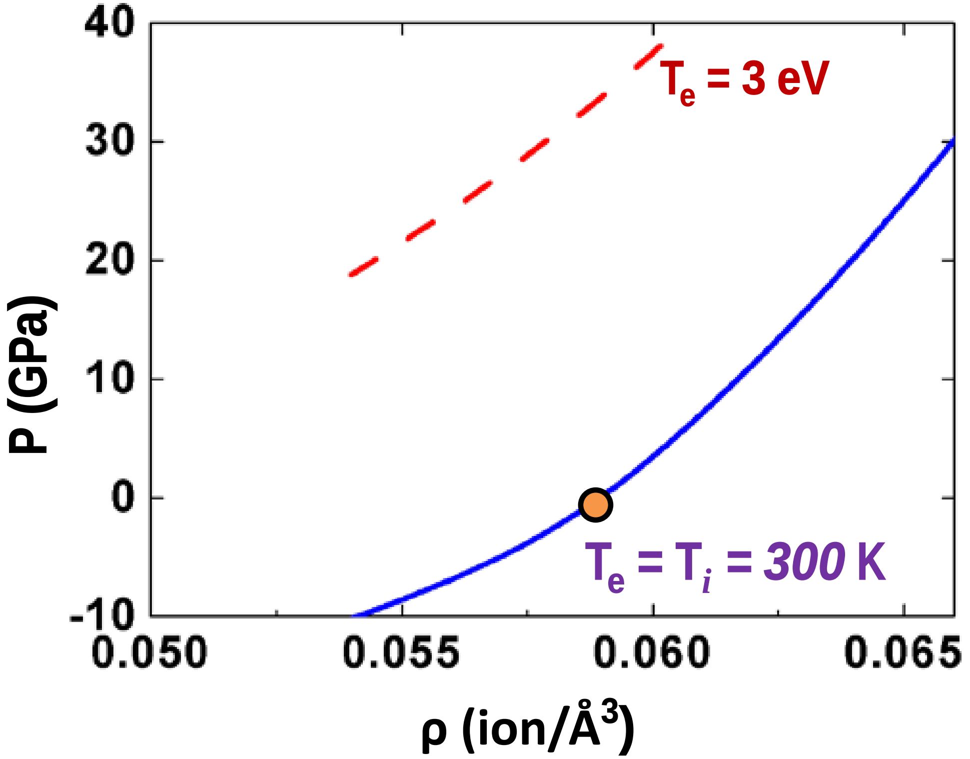
Ion structure in simulation box

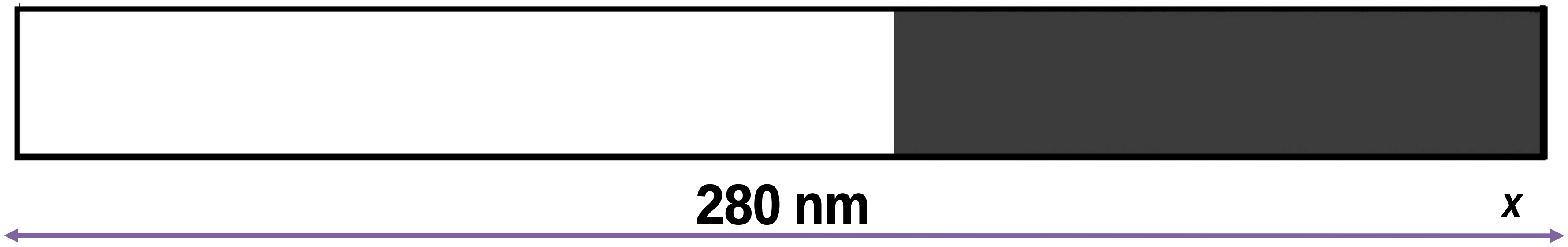


Ion structure in simulation box

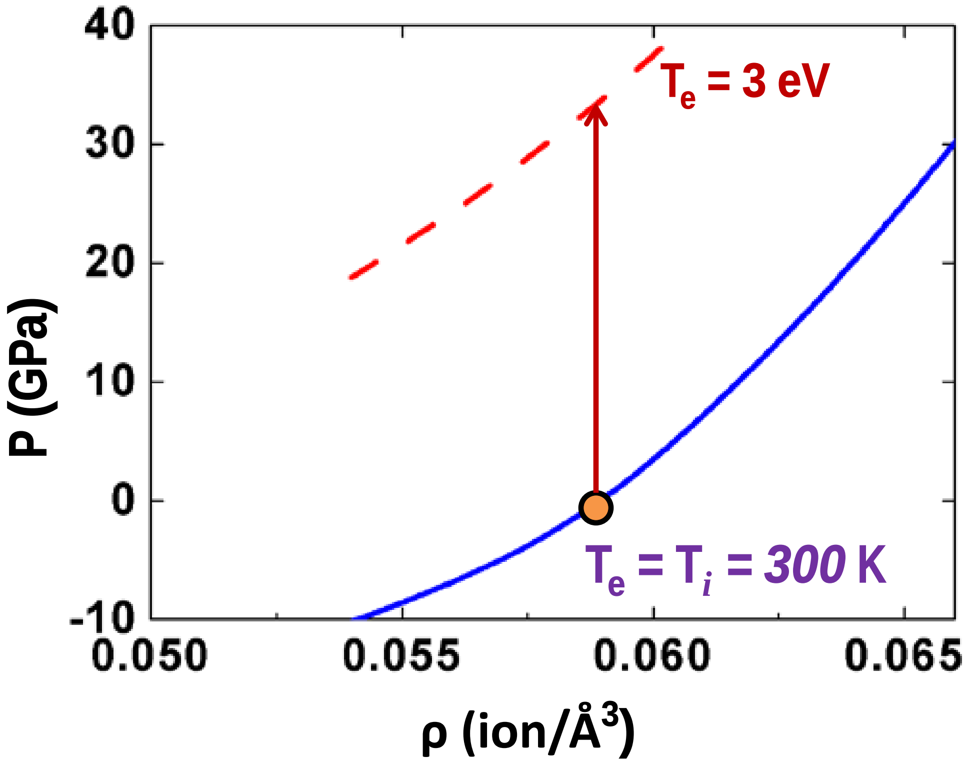


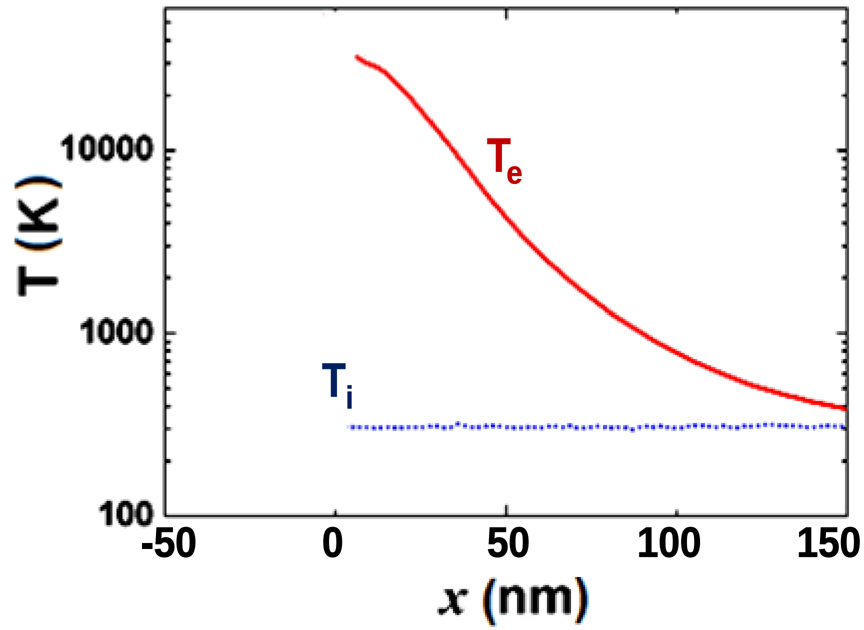
“Short” ablation at sub-ps laser pulse duration

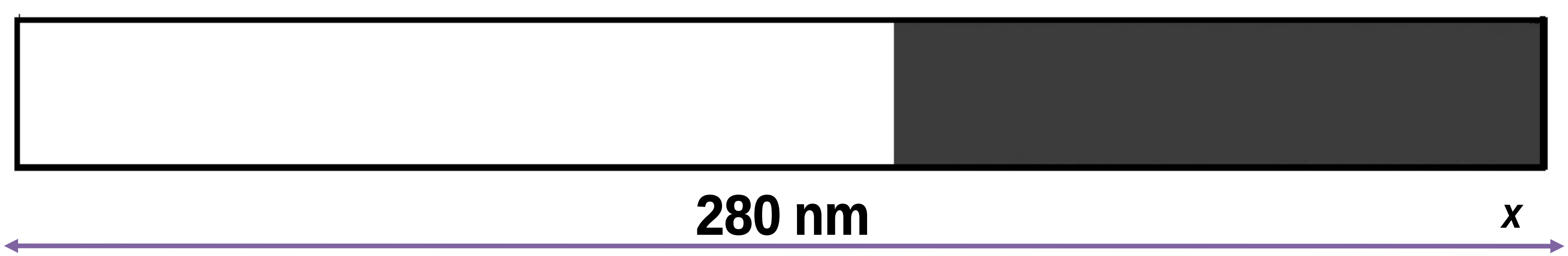




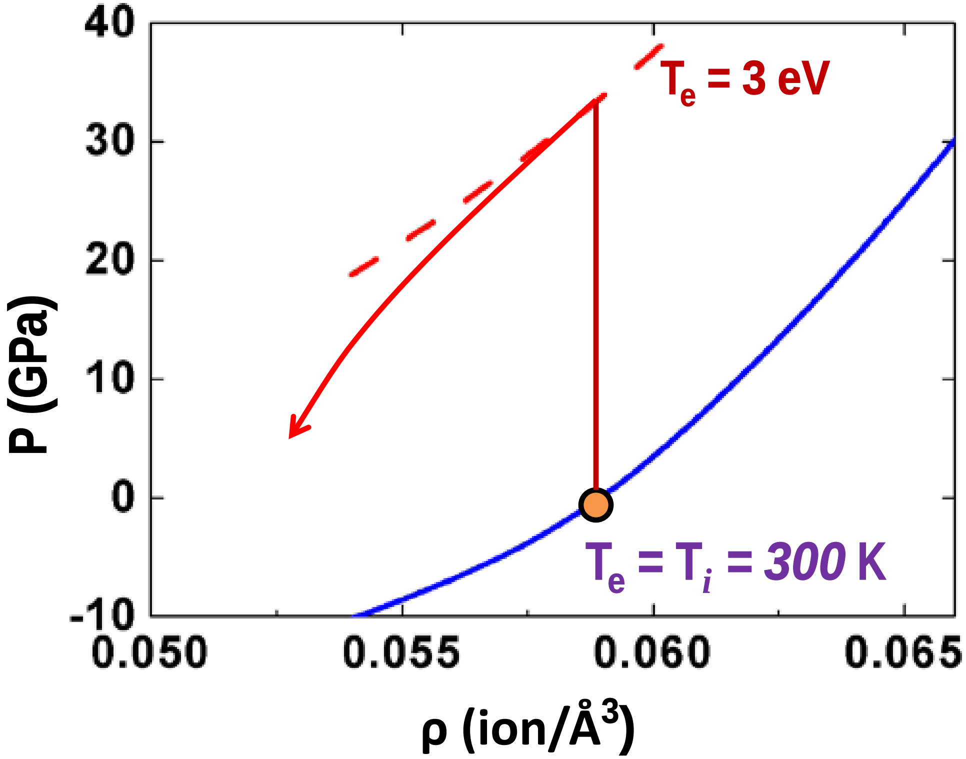
“Short” ablation at sub-ps laser pulse duration

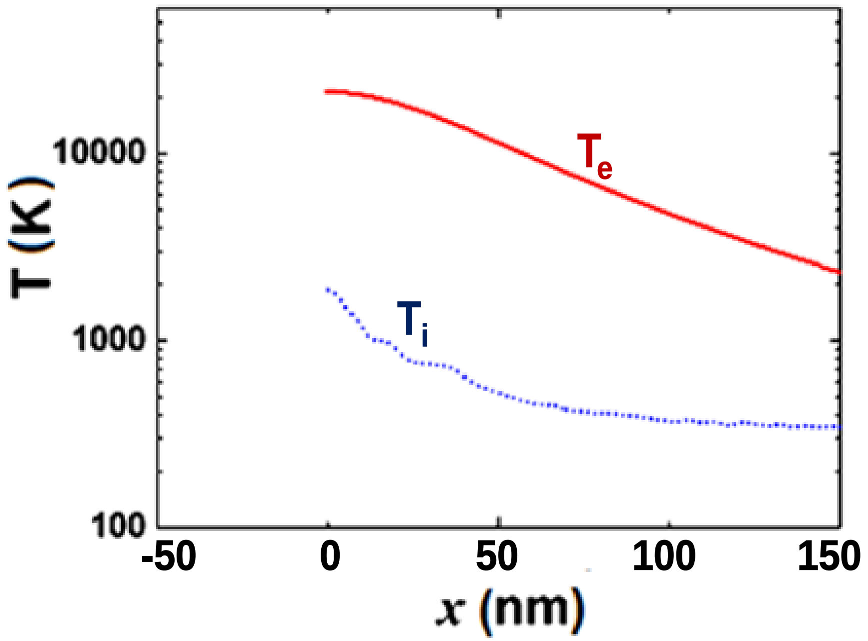


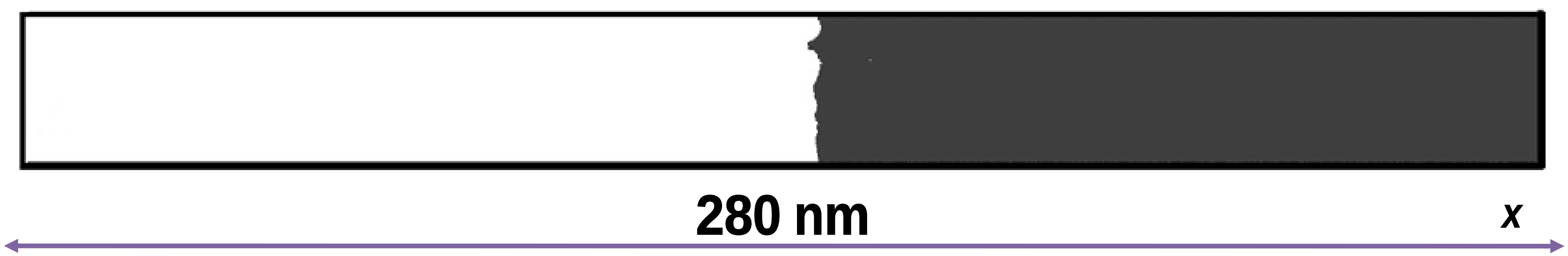




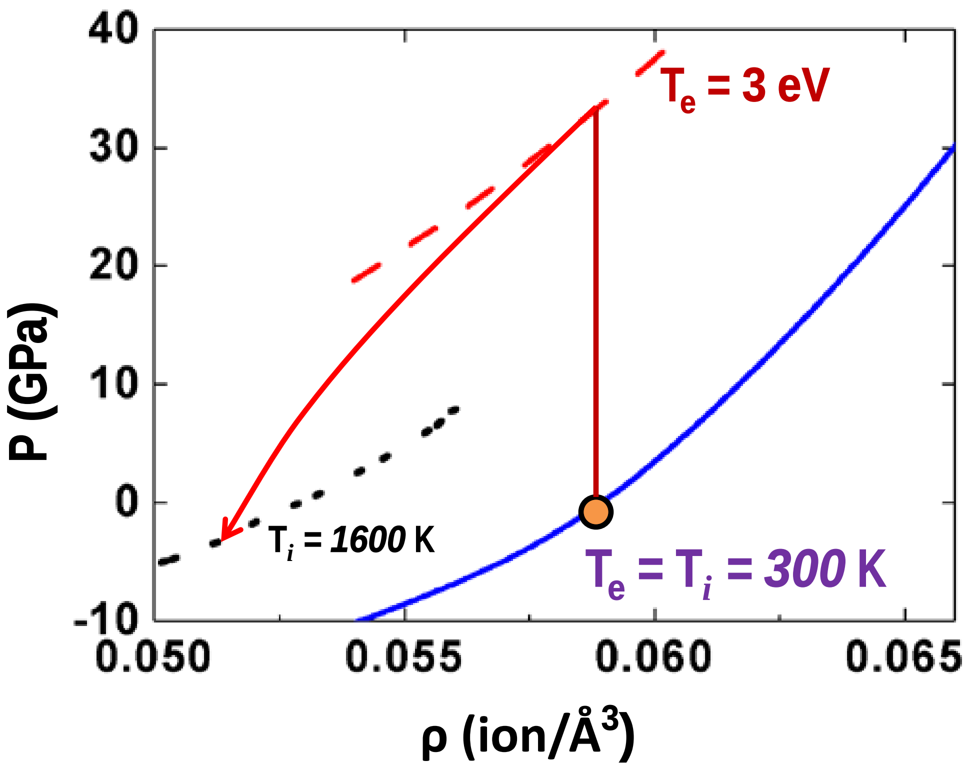
“Short” ablation at sub-ps laser pulse duration

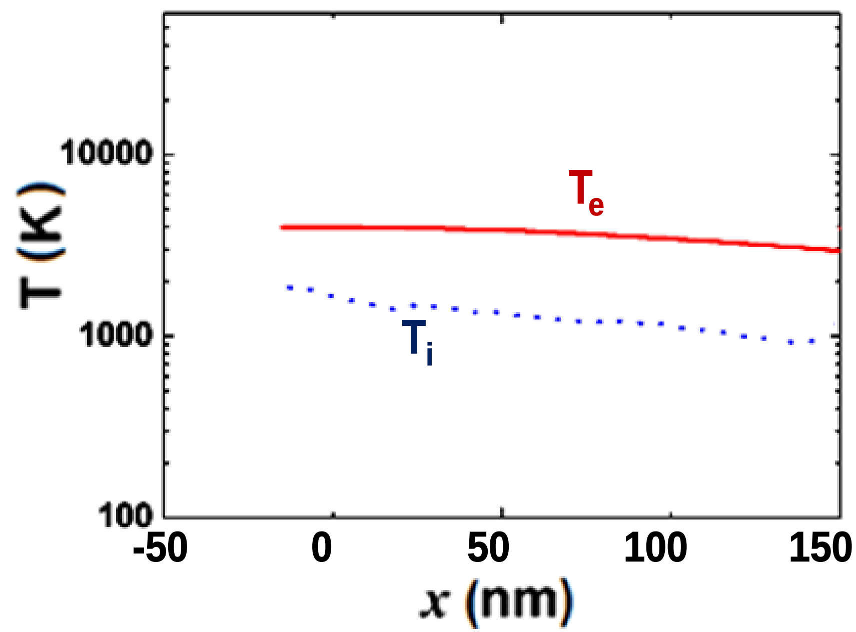






“Short” ablation at sub-ps laser pulse duration





"Short" ablation at sub-ps laser pulse duration