Scientific-coordination session
"Non-Ideal Plasma Research"

# Atomistic simulation of laser ablation of metals


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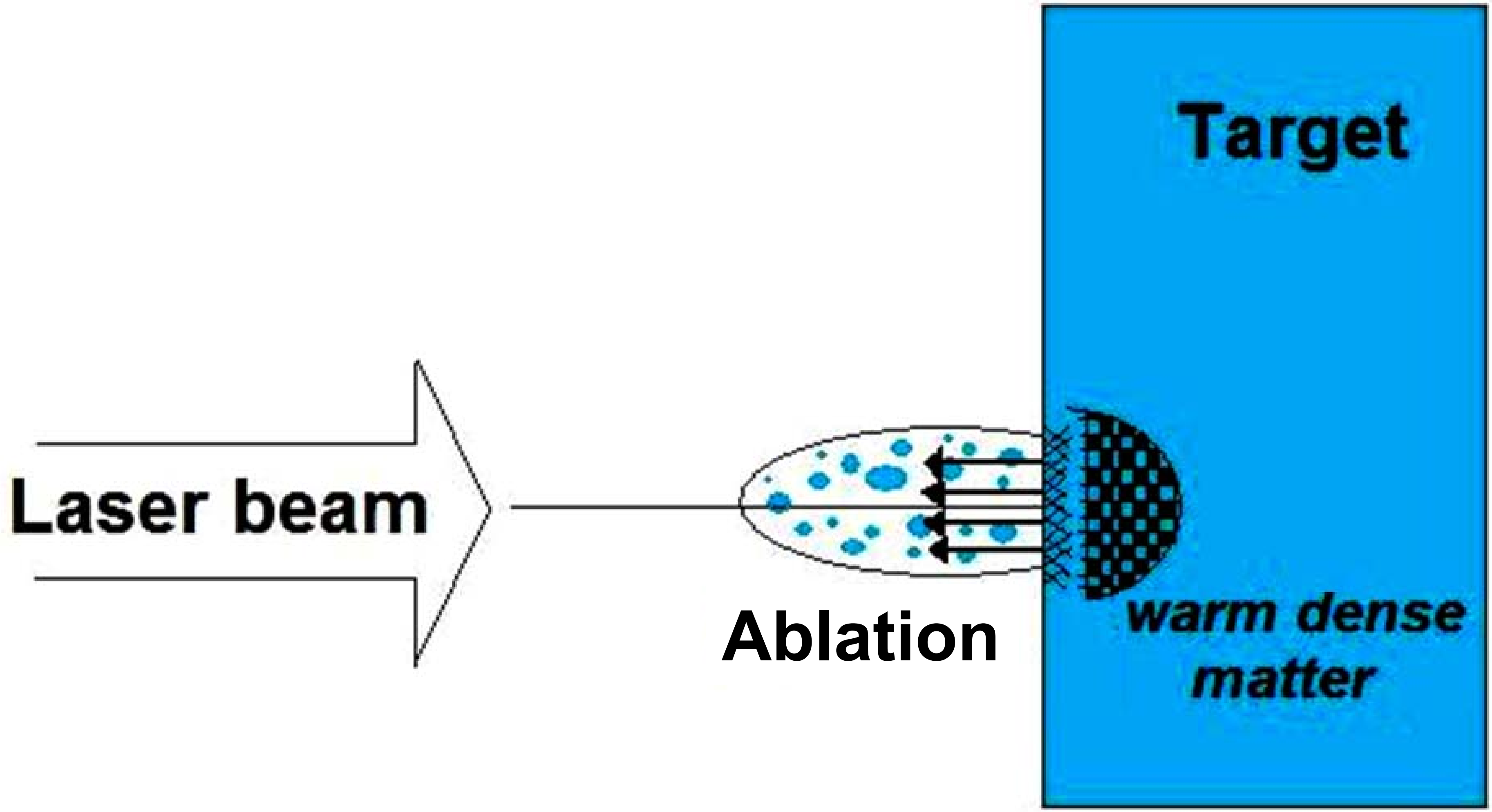
Joint Institute for High Temperatures RAS, Moscow
Moscow Institute of Physics and Technology

# Laser ablation – complex phenomenon



# Laser ablation – complex phenomenon

***Ion subsystem***

**condensed phase properties**

***Electronic subsystem***

**Two-temperature model**

EOS

Metastable states

Phase transitions

**Atomistic description!**

Heat capacity

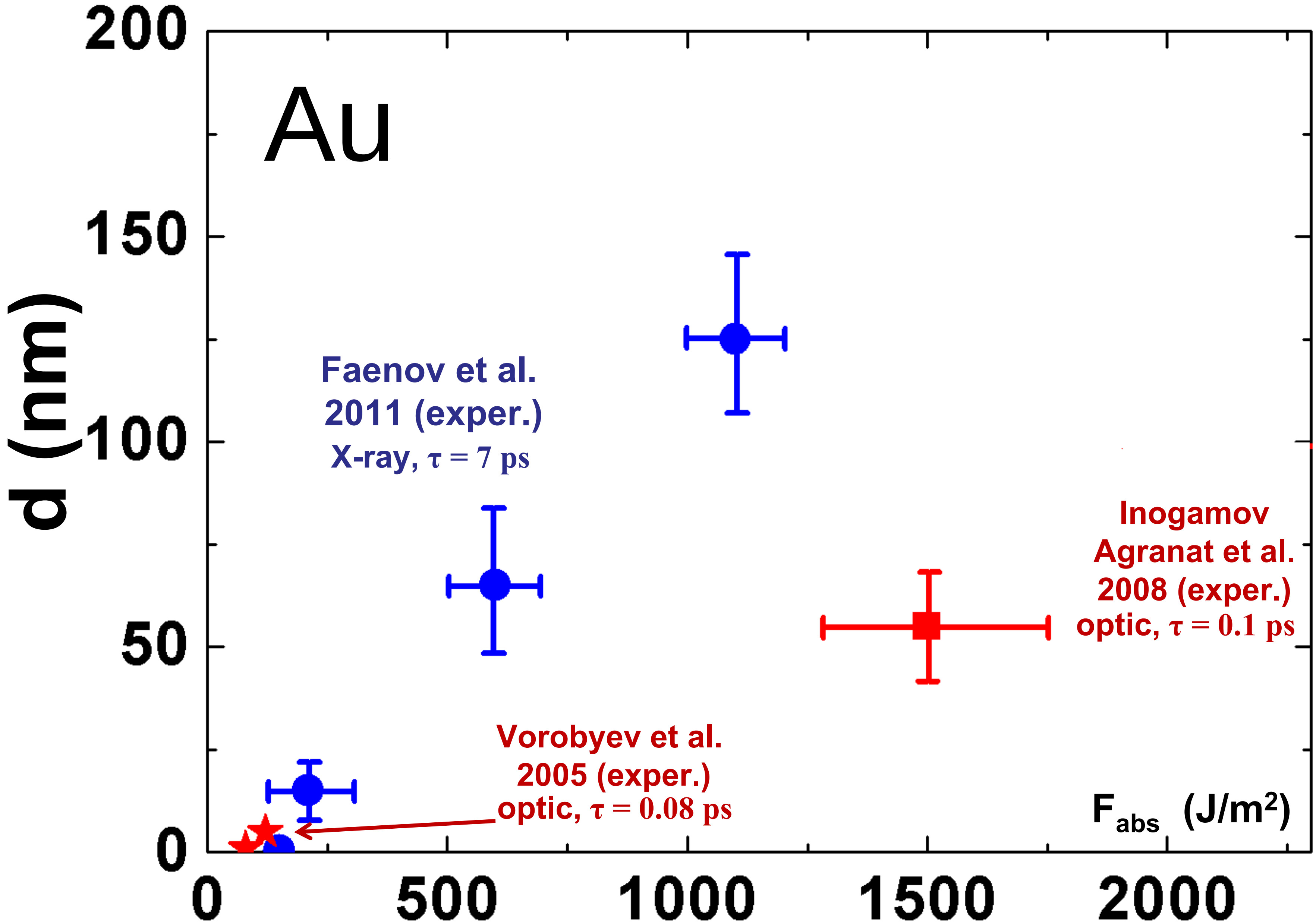
Heat conductivity

Electron-ion relaxation

Interatomic forces

Electronic pressure

# Dependence of crater depth on absorbed fluence



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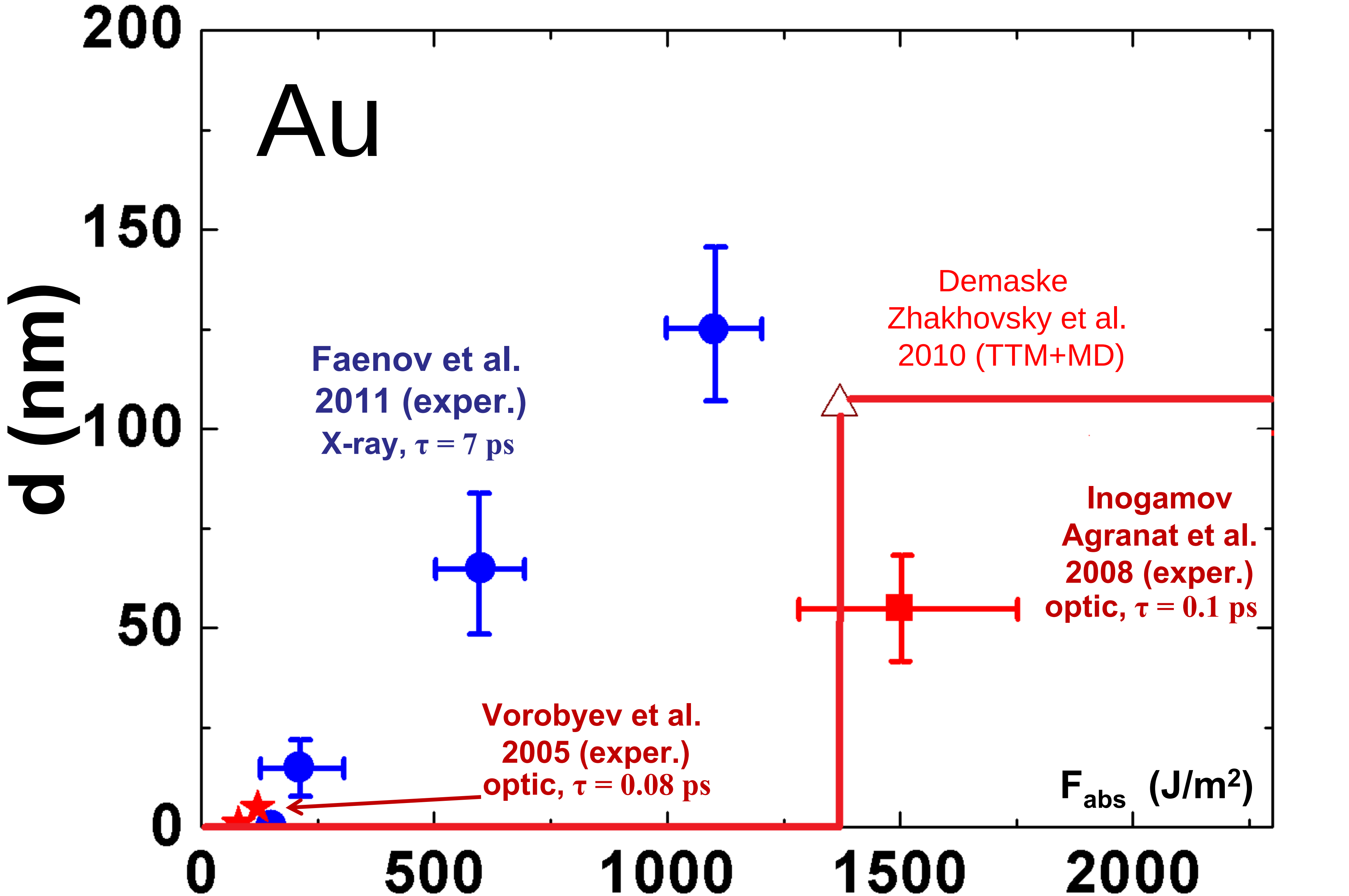
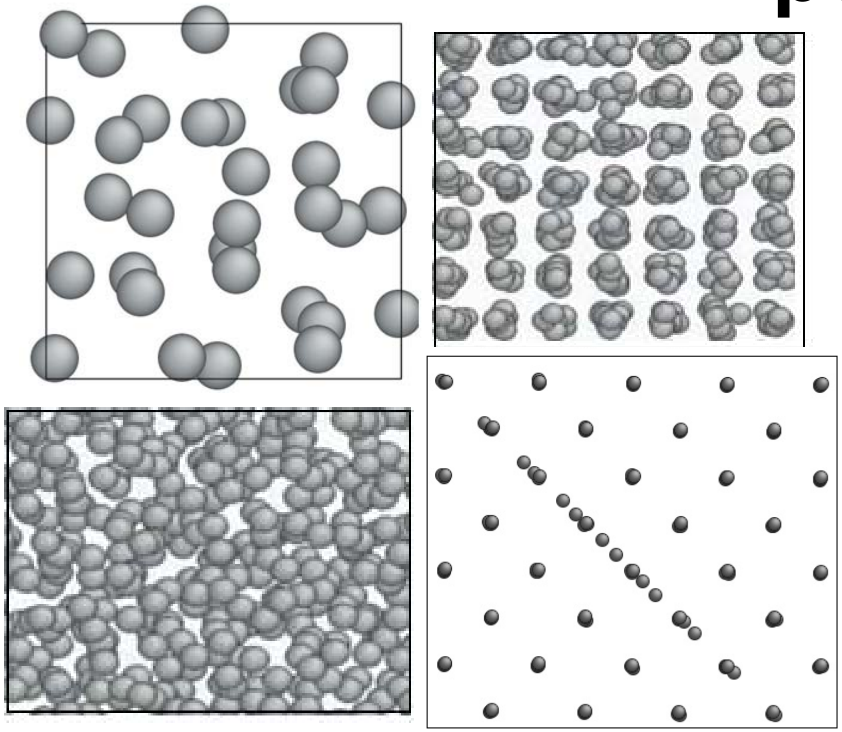


# Development of electronic-temperature-dependent potential for gold

# Force matching method for development of potential



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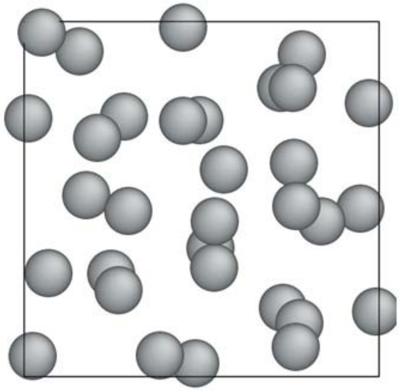
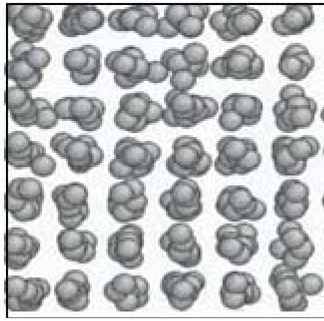
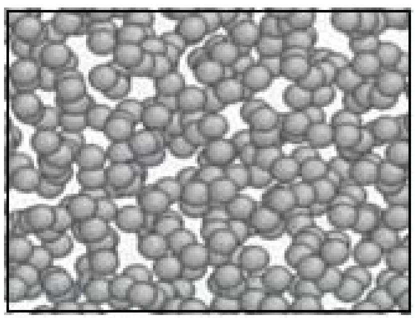
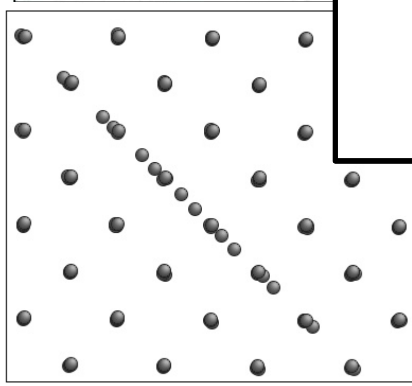




$$E = \frac{\langle \varphi_n | \mathbf{H} | \varphi_n \rangle}{\langle \varphi_n | \varphi_n \rangle}$$

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





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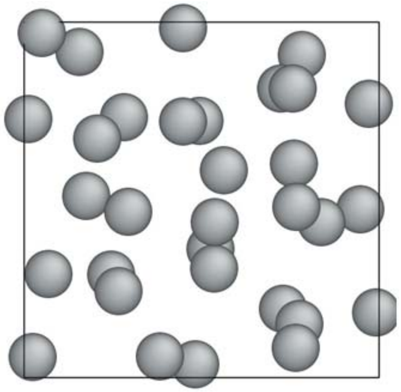

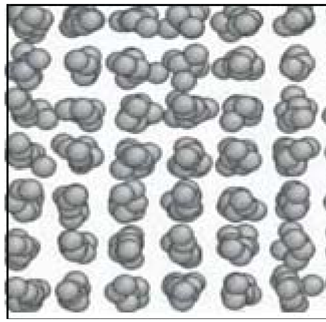

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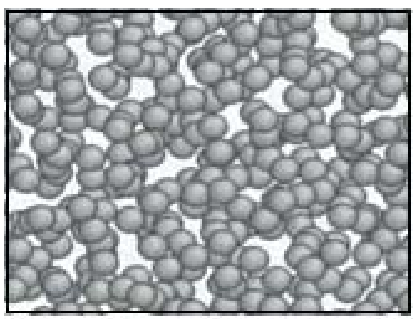

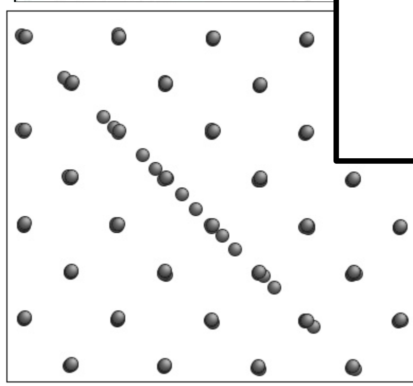

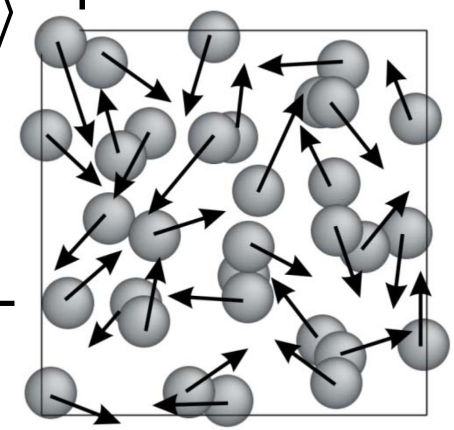

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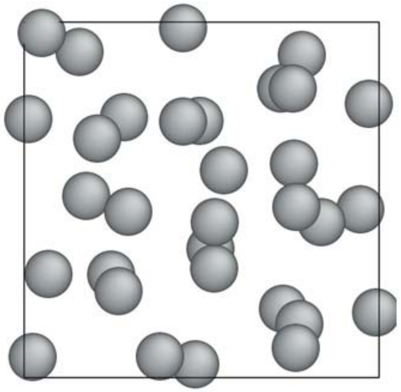

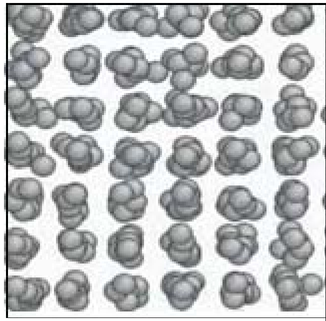

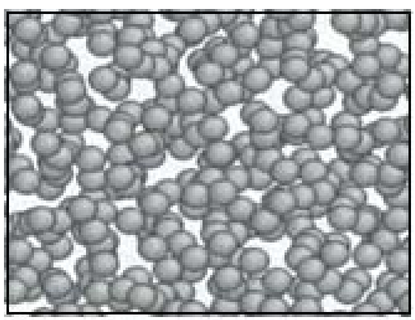

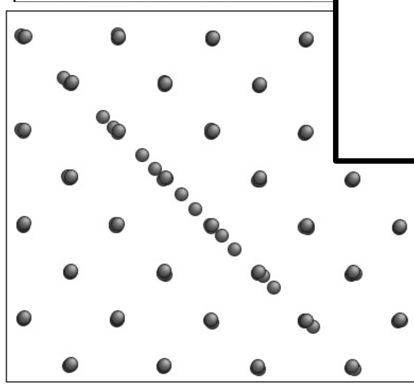

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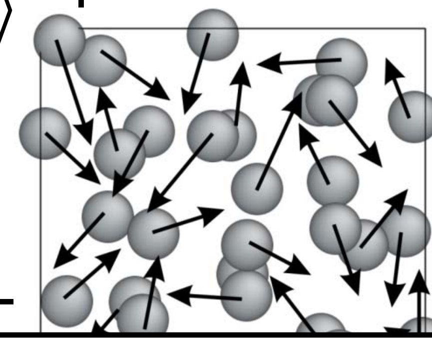
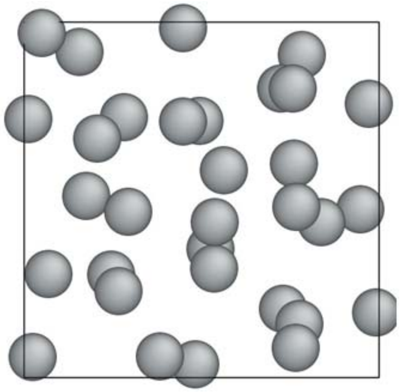
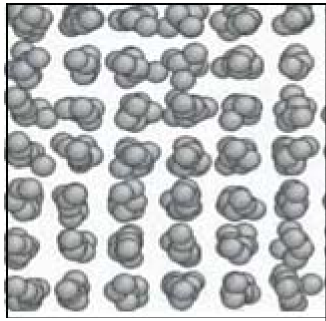


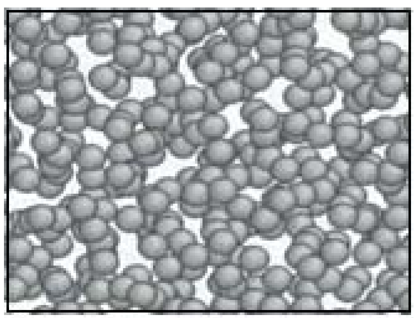
$$U = \sum_{i,\, j<i} \phi_{ij}(r_{ij}) + \sum_i F(\rho_i), \qquad \text{(EAM)}$$

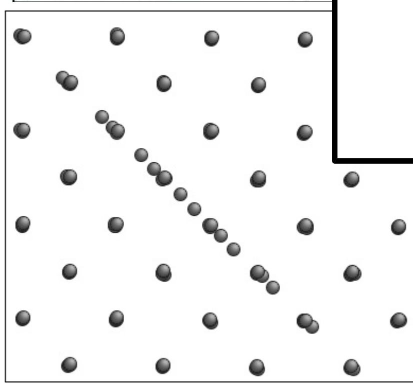
$$\rho_i = \sum_{j \neq i} \rho(r_{ij})$$

# Force matching method for development of potential









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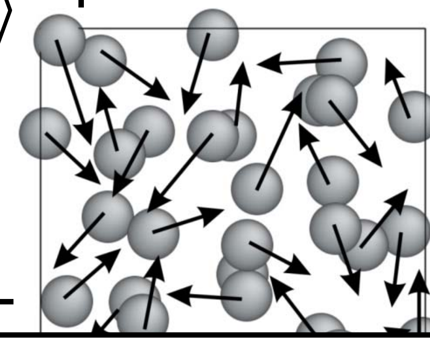


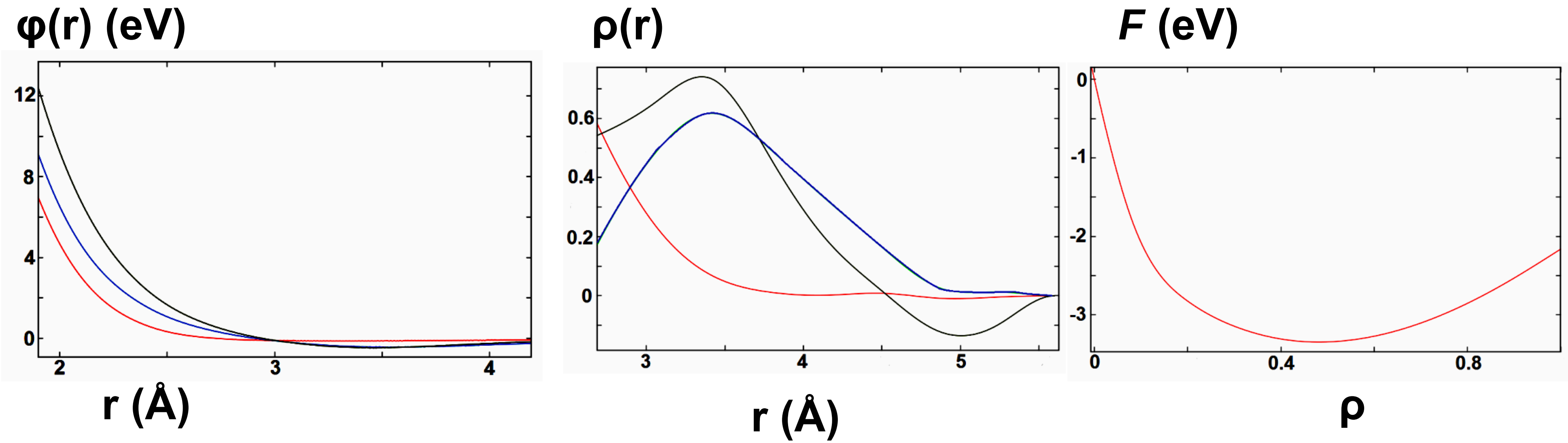
$$U = \sum_{i, j<i} \phi_{ij}(r_{ij}) + \sum_{i} F(\rho_i),$$

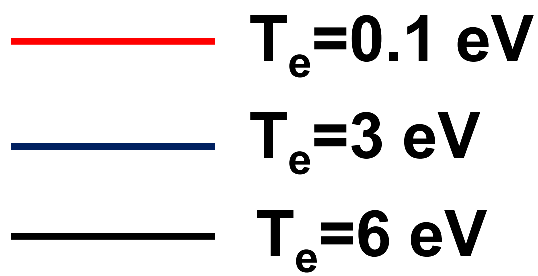
$$\rho_i = \sum_{j \neq i} \rho(r_{ij})$$

(EAM)

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

# ETD-potential for gold

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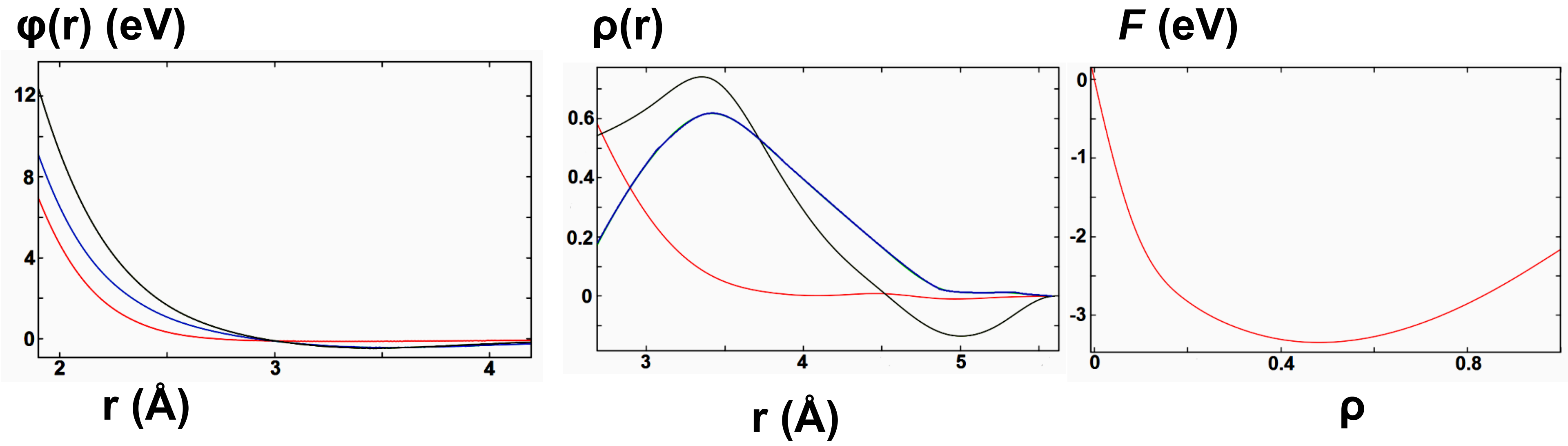



$T_e$=0.1 eV

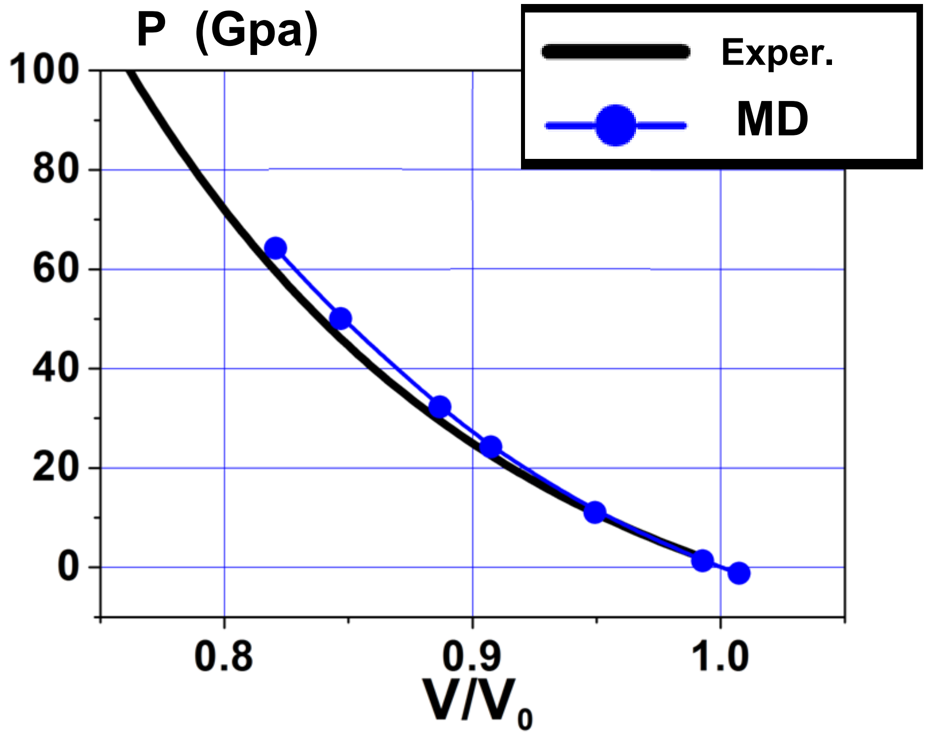
$T_e$=3 eV

$T_e$=6 eV

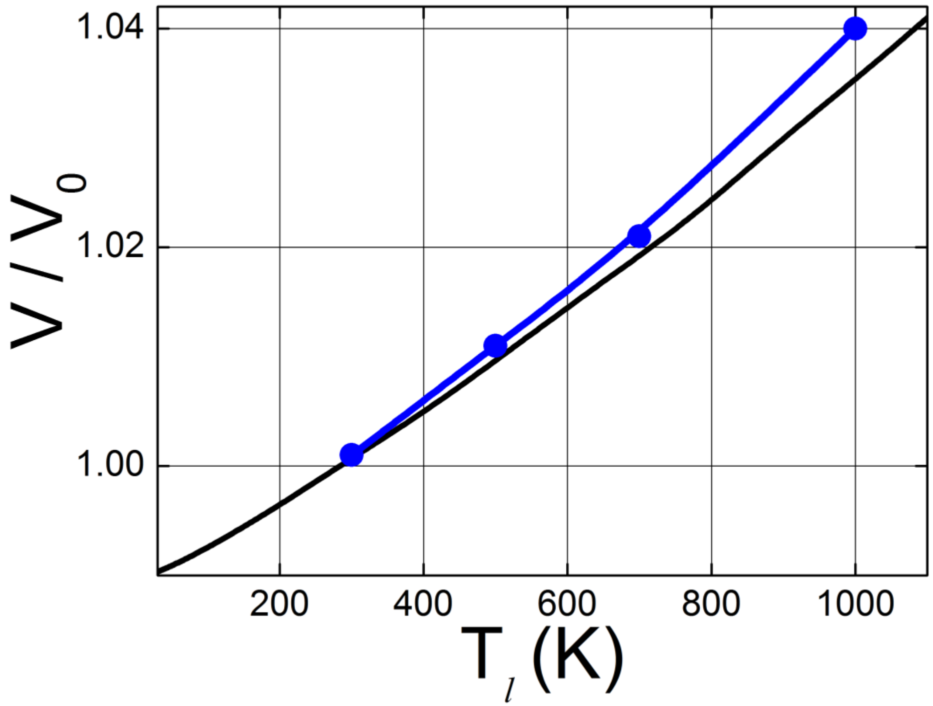
$$E = \sum_{j}^{N} \left( \varphi_0(r_j) + \varphi_1(r_j) \cdot T_e + \varphi_2(r_j) \cdot T_e^{\,2} \right) + $$

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

# Verification of ETD-potential at $T_e = 0.05$ eV



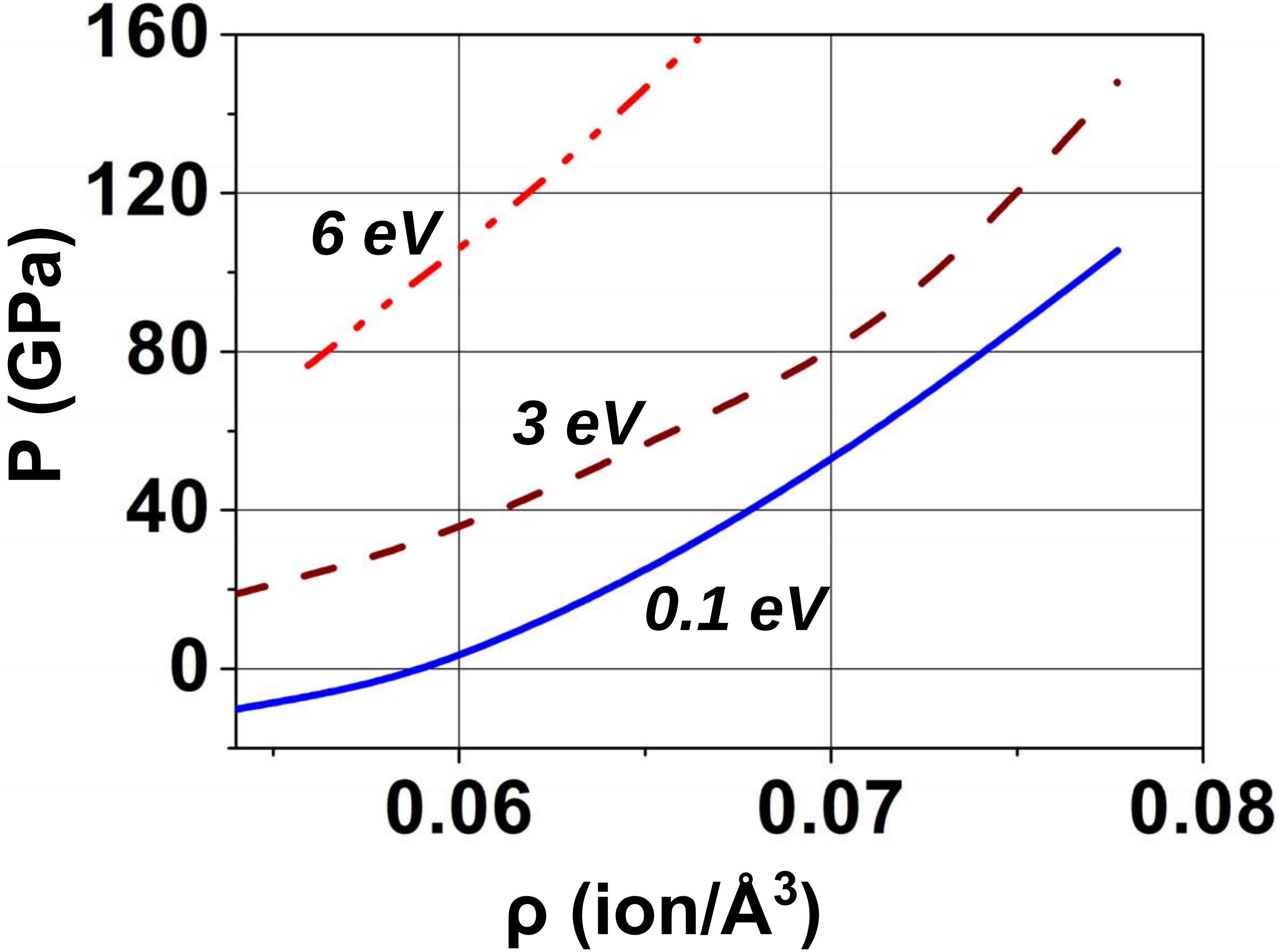

*Cold curve Au*




*Thermal expansion Au*

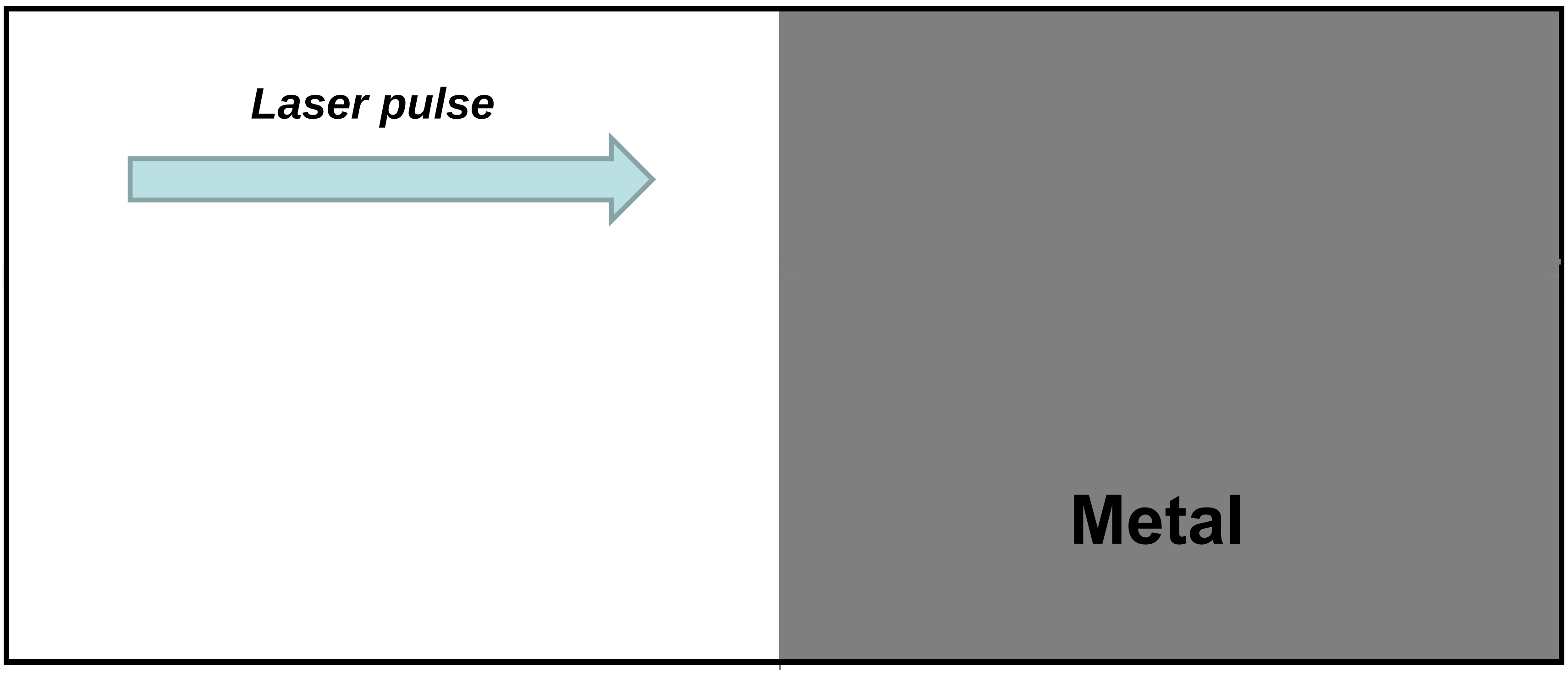
| | $V_0$ , $Å^3$ | $E_c$ , eV | $C_{11}$ , GPa | $C_{12}$ , GPa | $T_{melt}$ , K |
|---|---|---|---|---|---|
| **experiment** | 10.22 | 3.8 | 202 | 170 | 1338 |
| **MD** | 10.23 | 4.1 | 225 | 180 | 1210 |

# Gold isotherms at $T_i$ = *300* K and various temperatures $T_e$



# Atomistic simulation of laser ablation of gold

# Ion structure in simulation box



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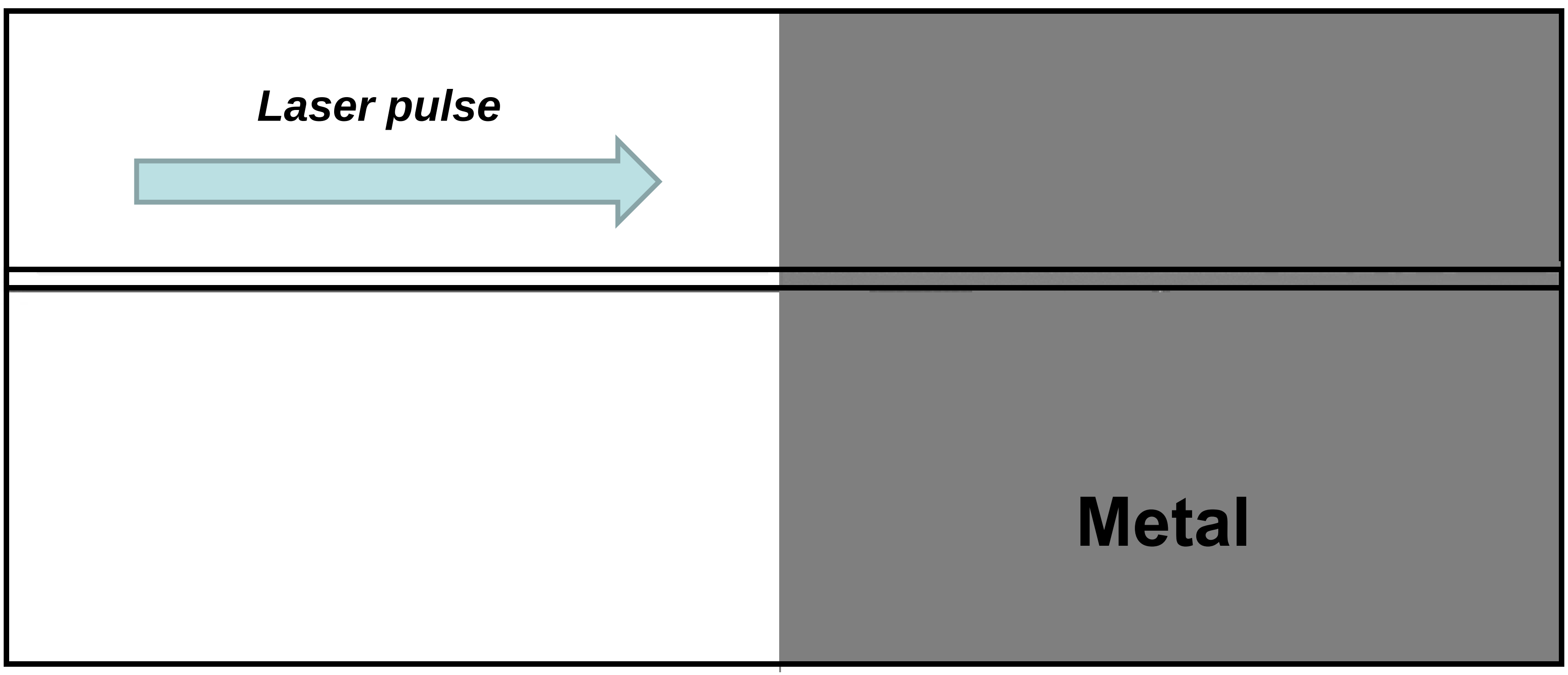

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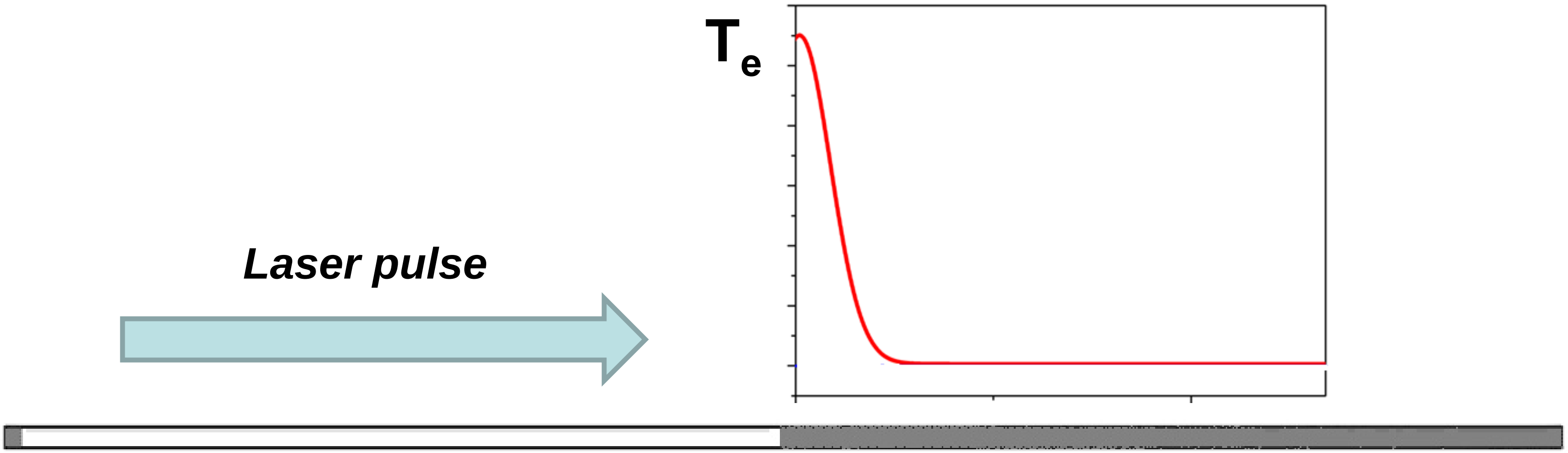
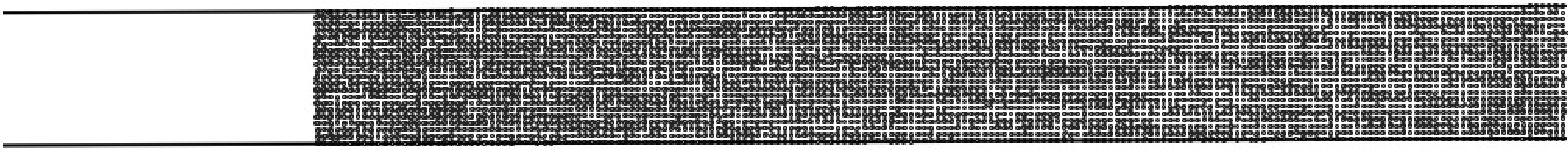
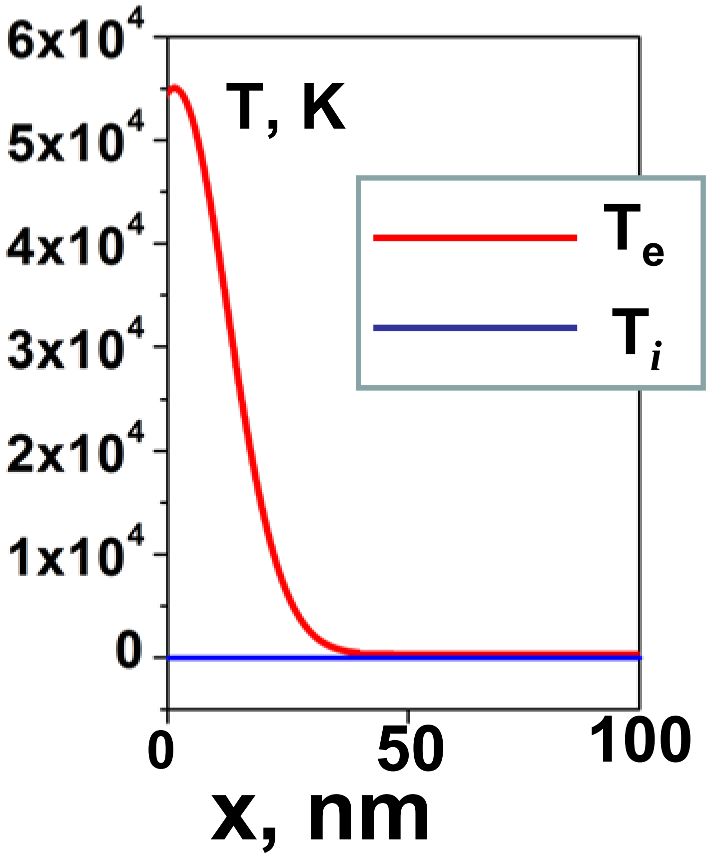


# Model






$G_p = 2.1 \cdot 10^{16}$ [W/(K•m³)]

$C_e(T_e)$ [Lin, Zhigilei (2008)]

$k_e(T_e)$ [Ivanov, Zhigilei (2003)]

$$C_e \frac{\partial T_e}{\partial t} = \nabla\left(\kappa_e \nabla T_e\right) - G_p\left(T_e - T_i\right) - \frac{I(t)\exp(-x/l)}{l}$$

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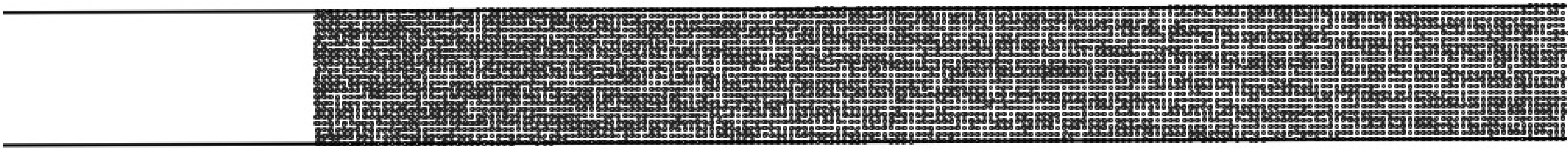

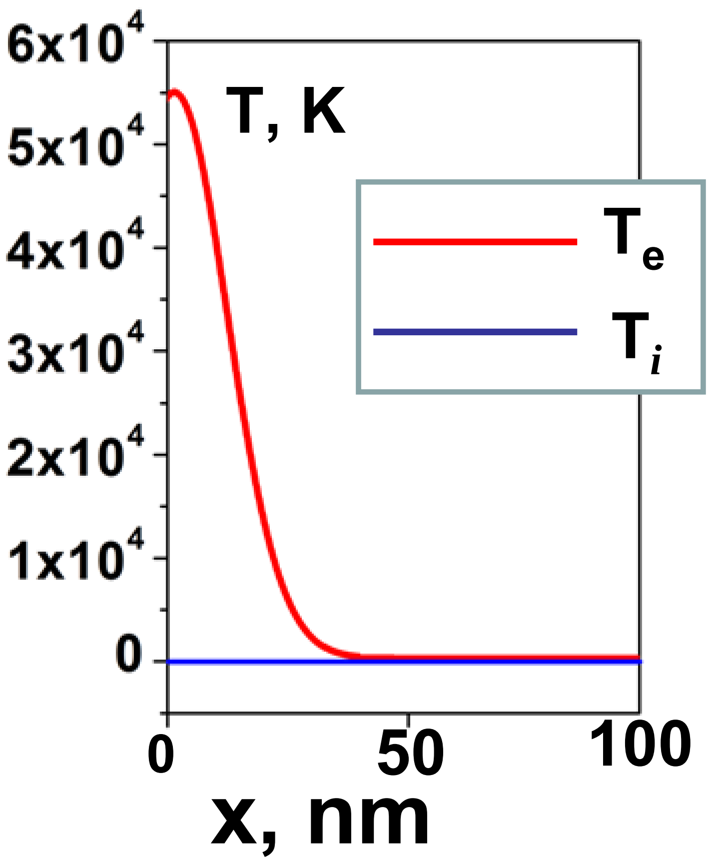


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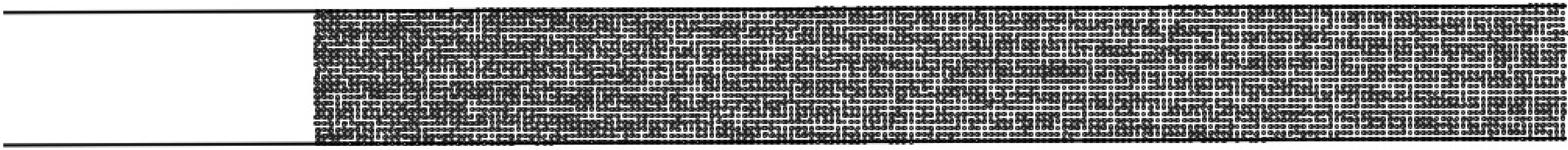
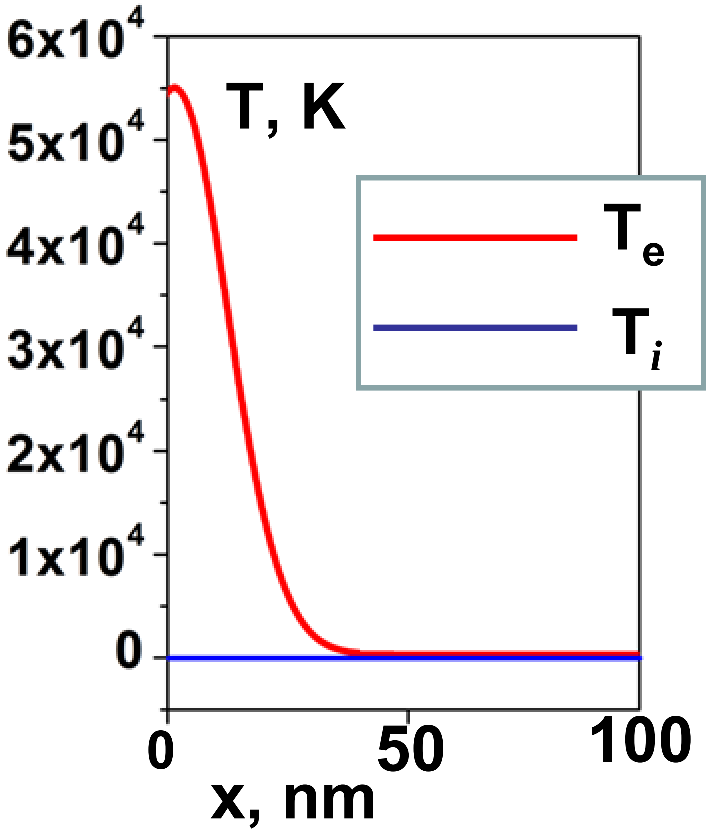
$$m\frac{d\vec{v}_i}{dt} = \vec{F}_i\left(T_e\right) - \beta\vec{v}_i + \vec{\xi}\left(T_e\right) - \frac{\nabla P_{e\ \mathrm{free}}}{\rho_{ion}}$$

$$\xi \sim T_e^{1/2} \quad \tau = \frac{m}{\beta} \quad \tau \sim 20\ \mathrm{ps}$$

MD simulation +
ETD-potential +
Langevin thermostat

# Model



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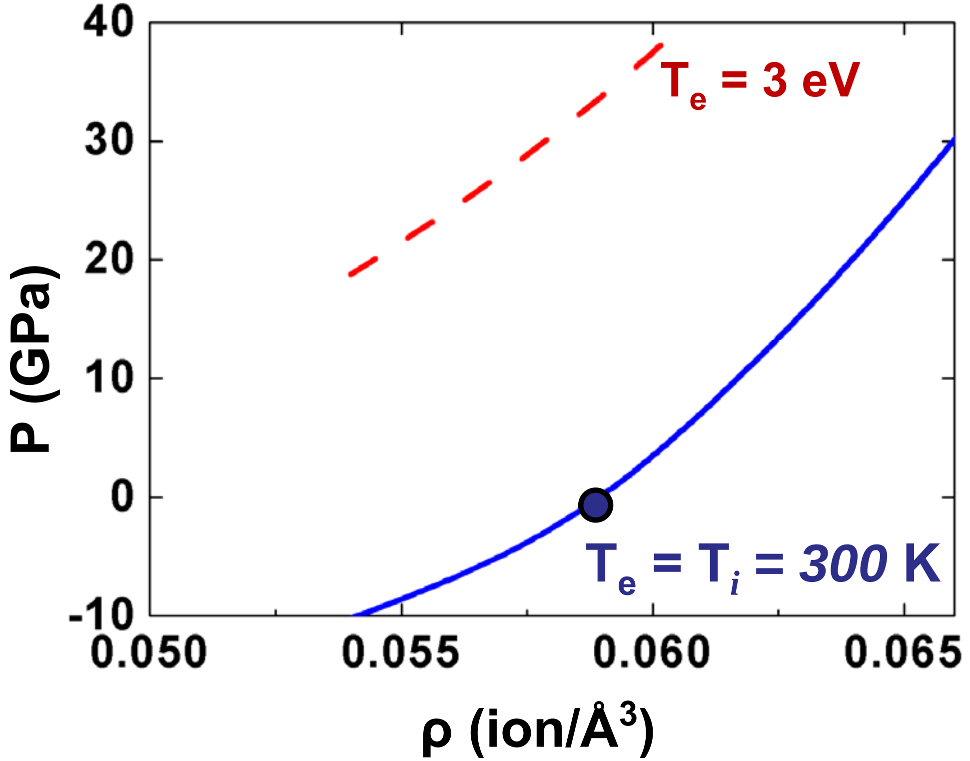
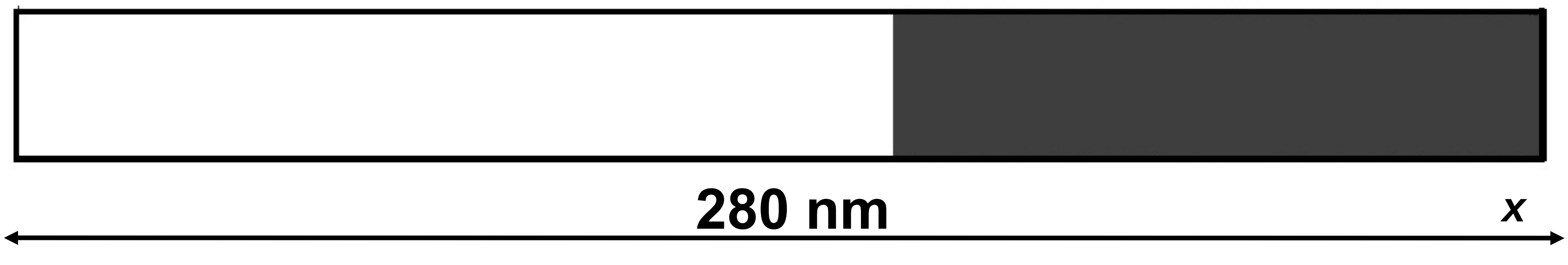
MD simulation +
ETD-potential +
Langevin thermostat

# “Short” ablation at sub-ps laser pulse duration



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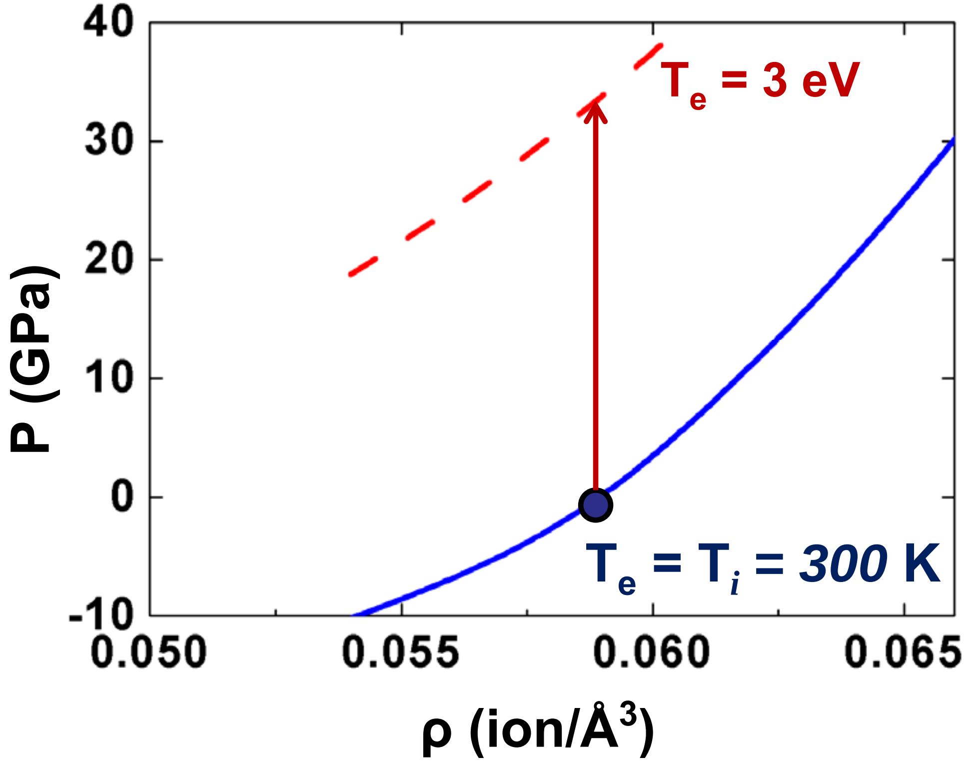


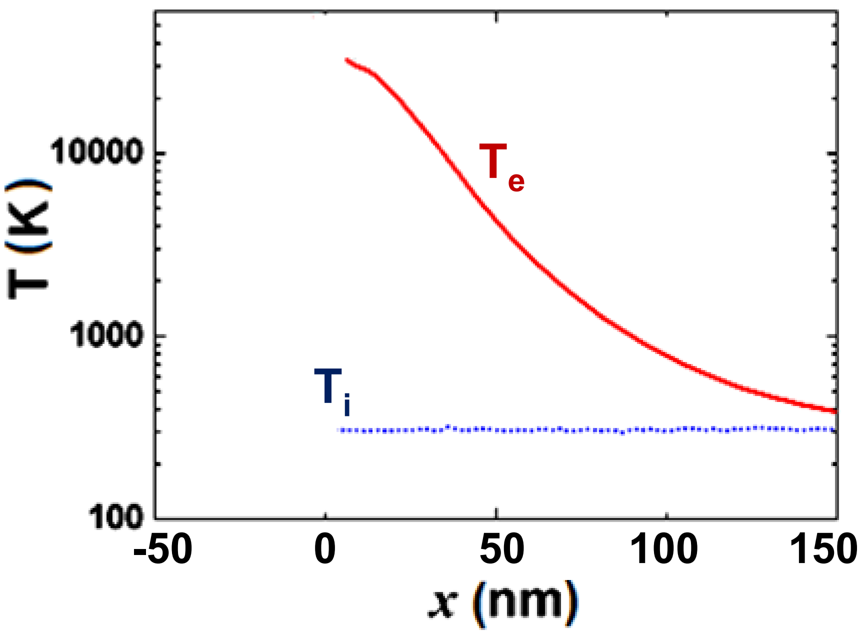


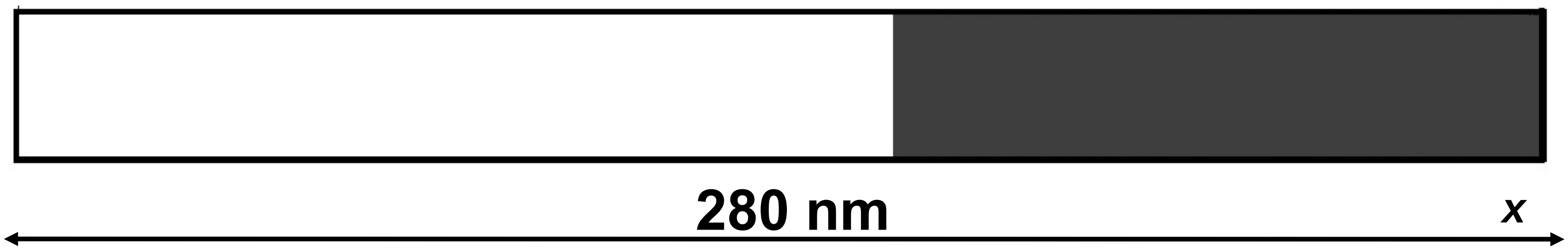

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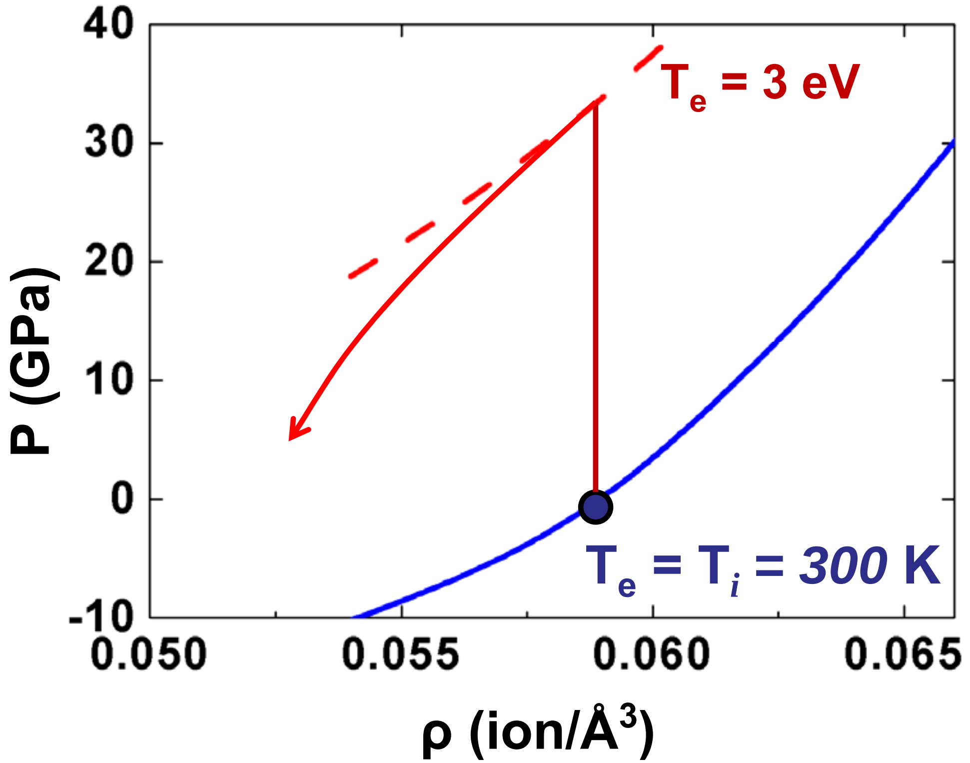


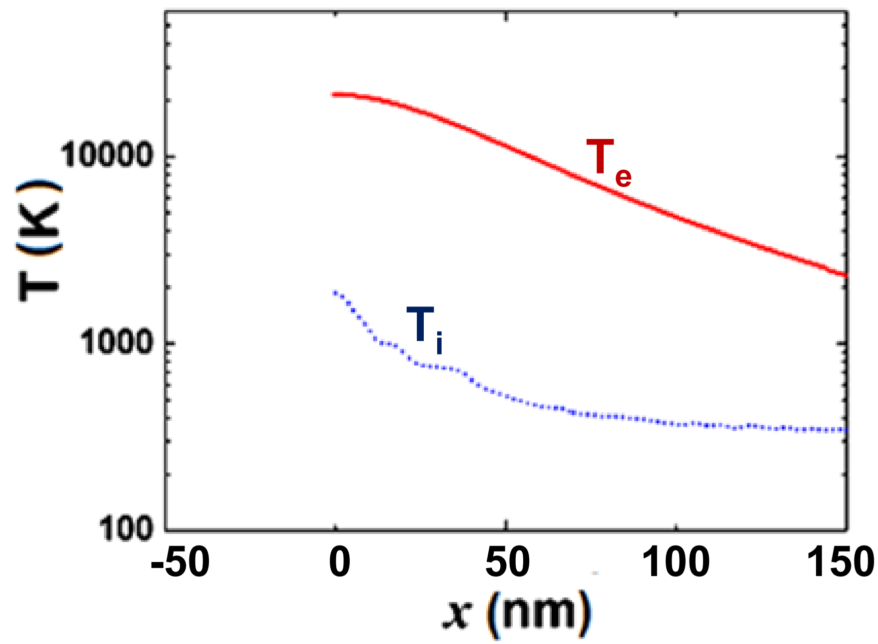


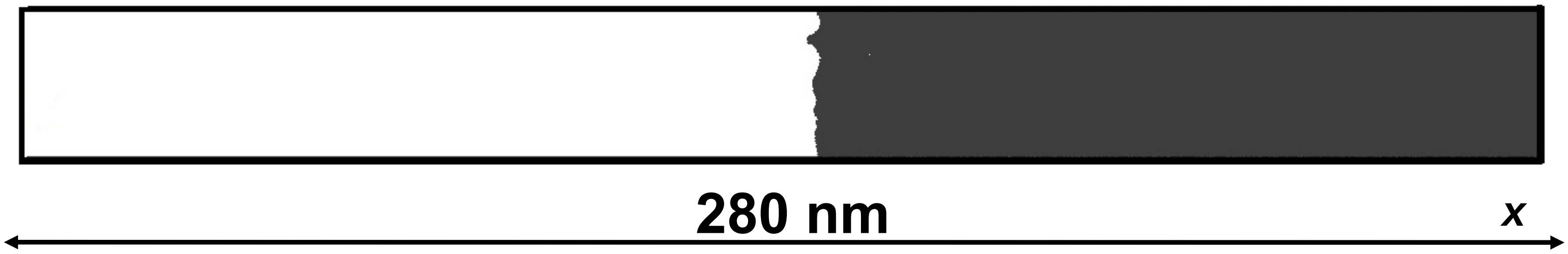

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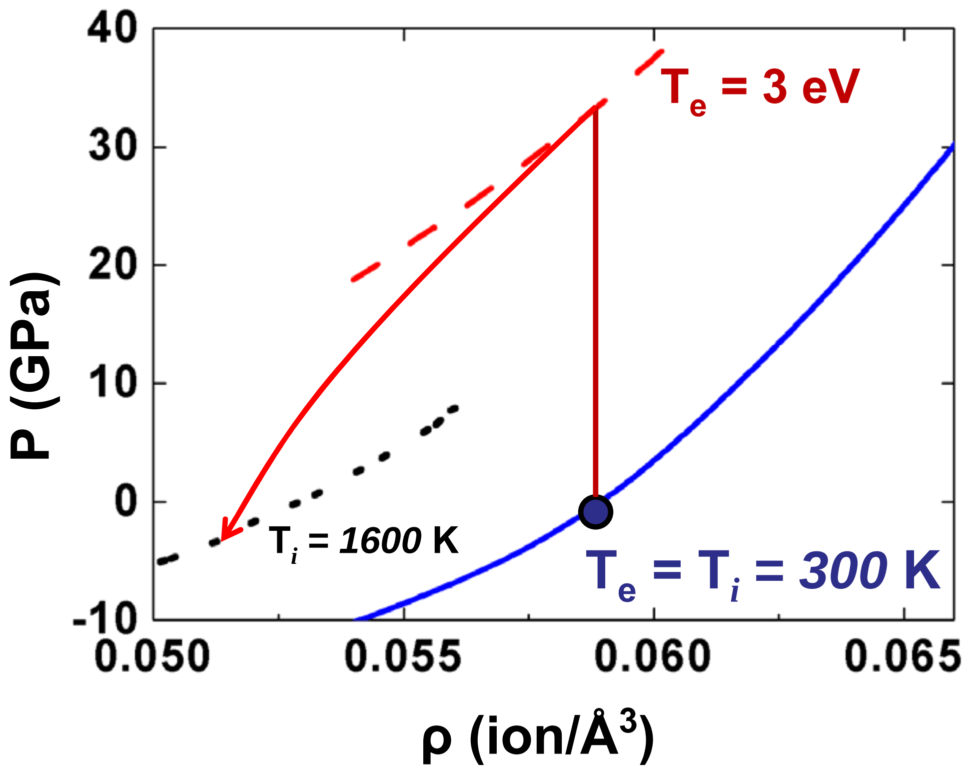


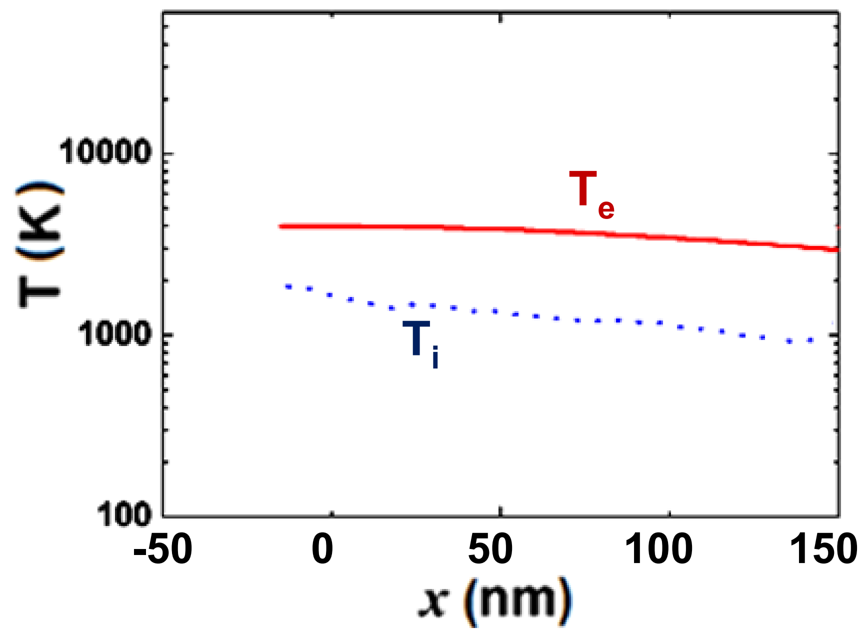


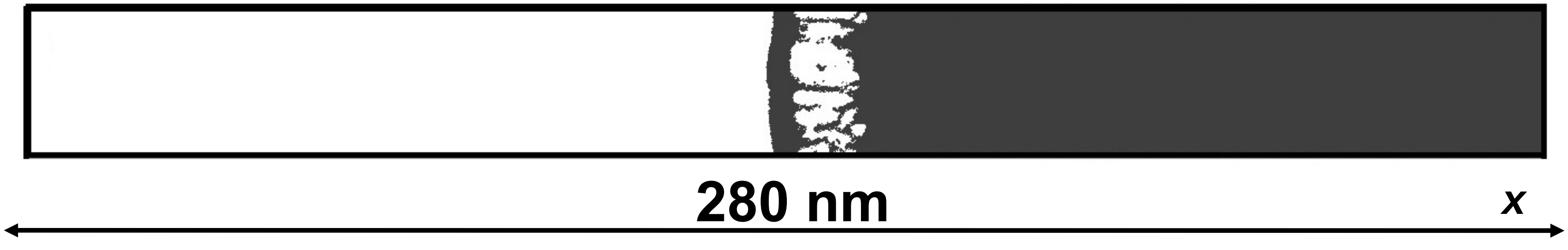

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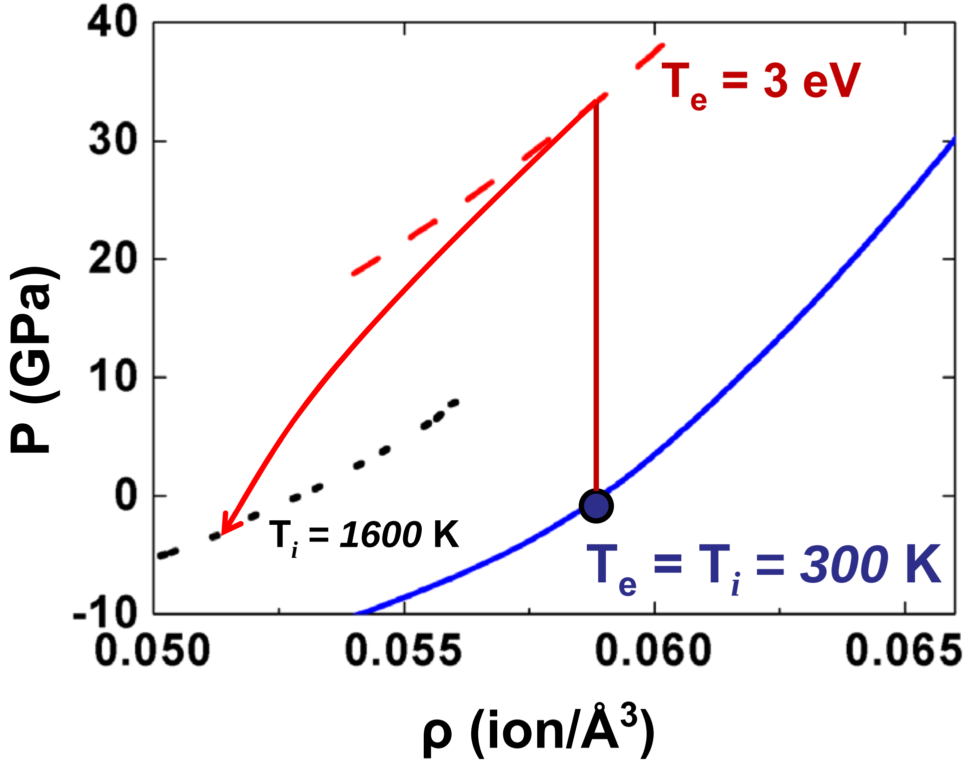
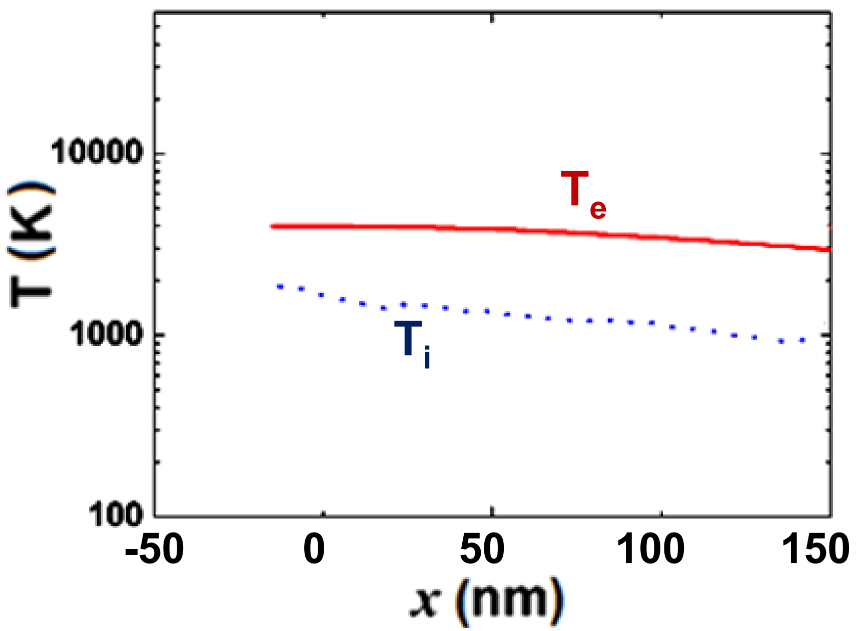
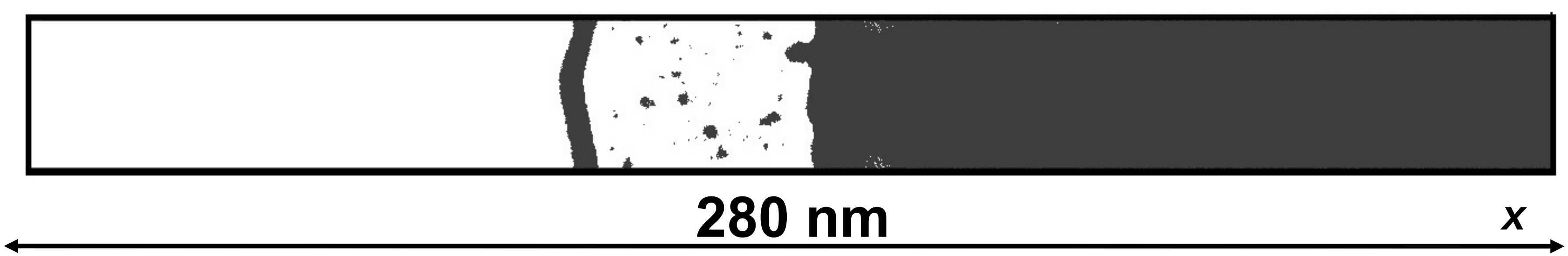
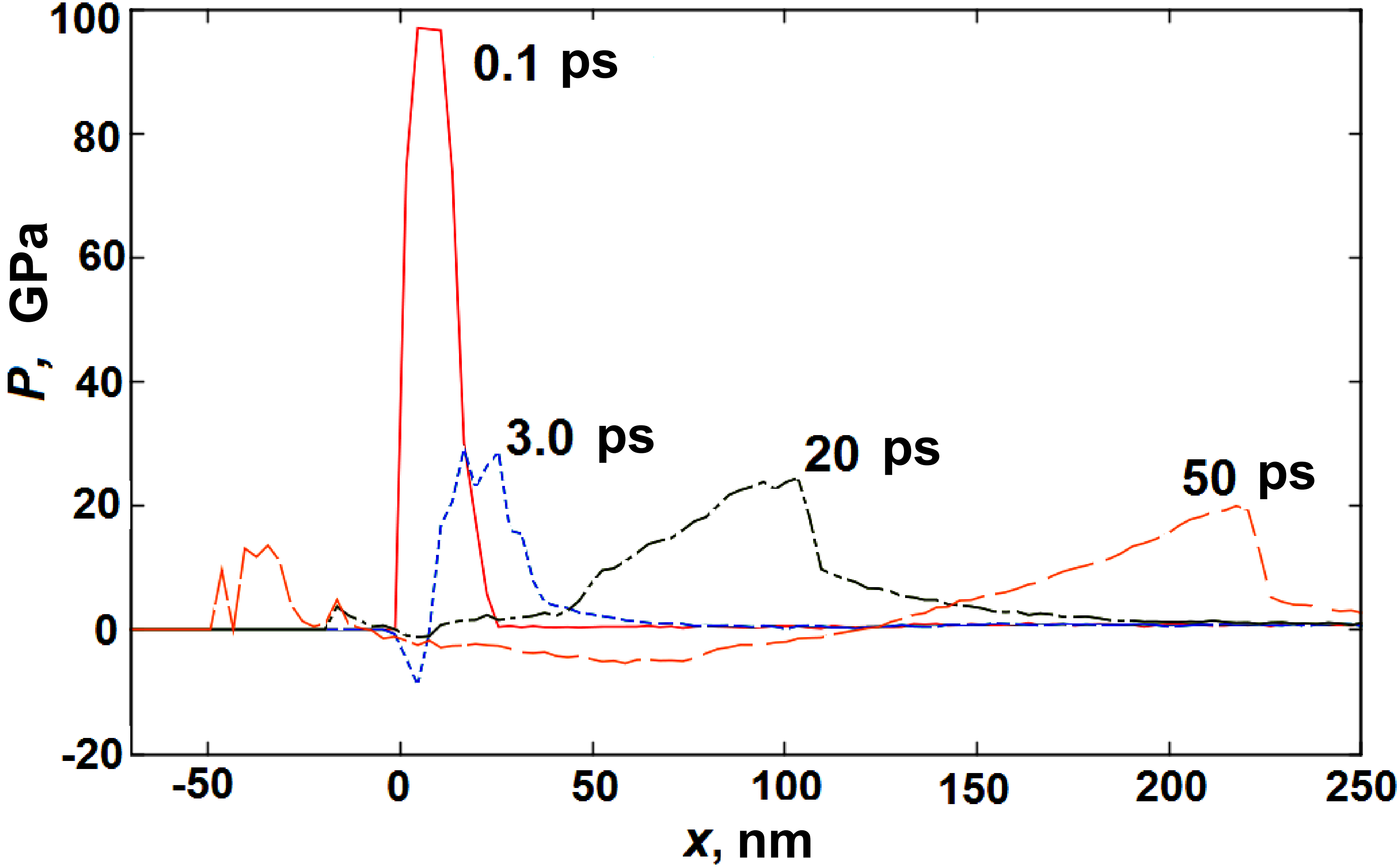


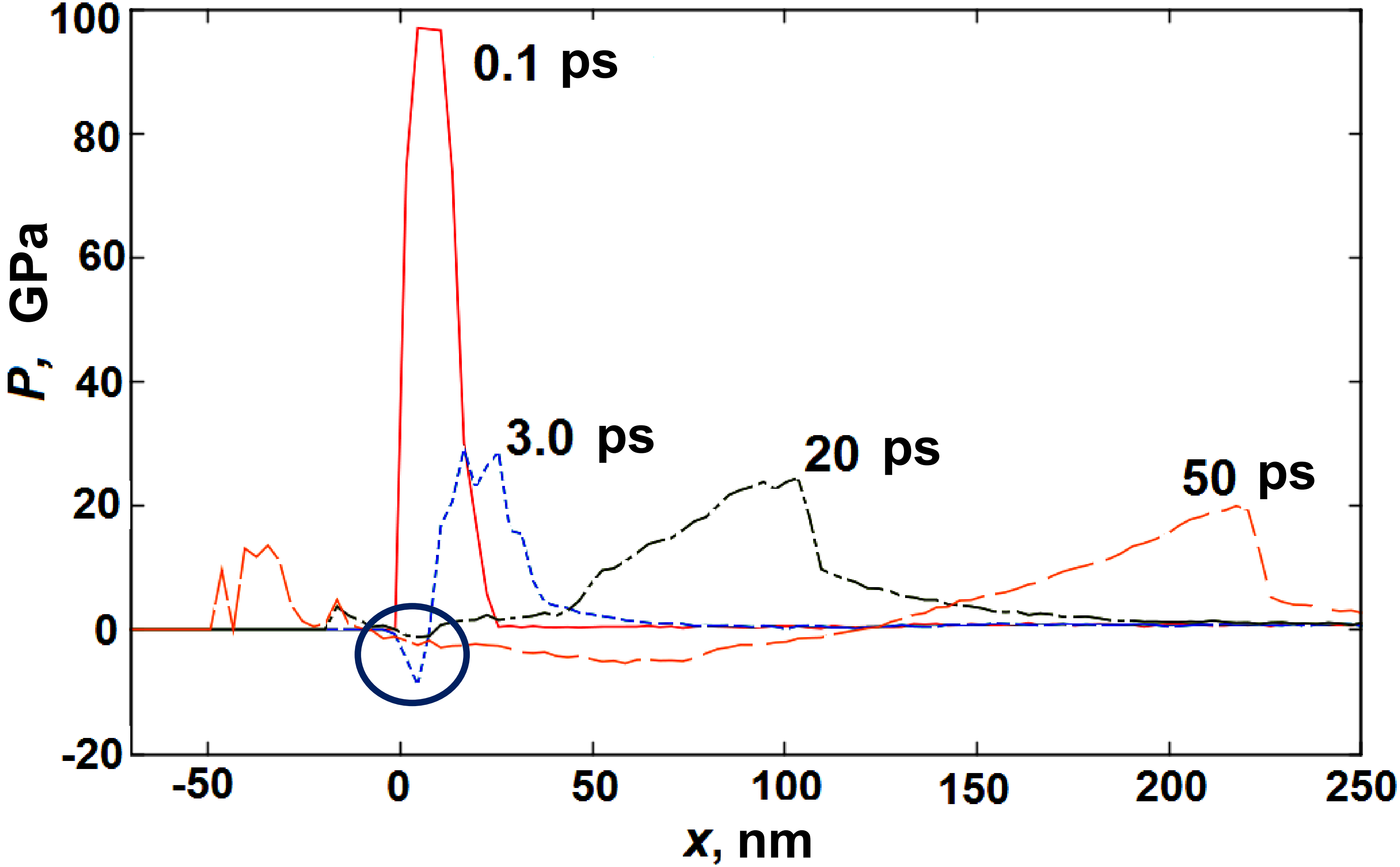



# ablation mechanism



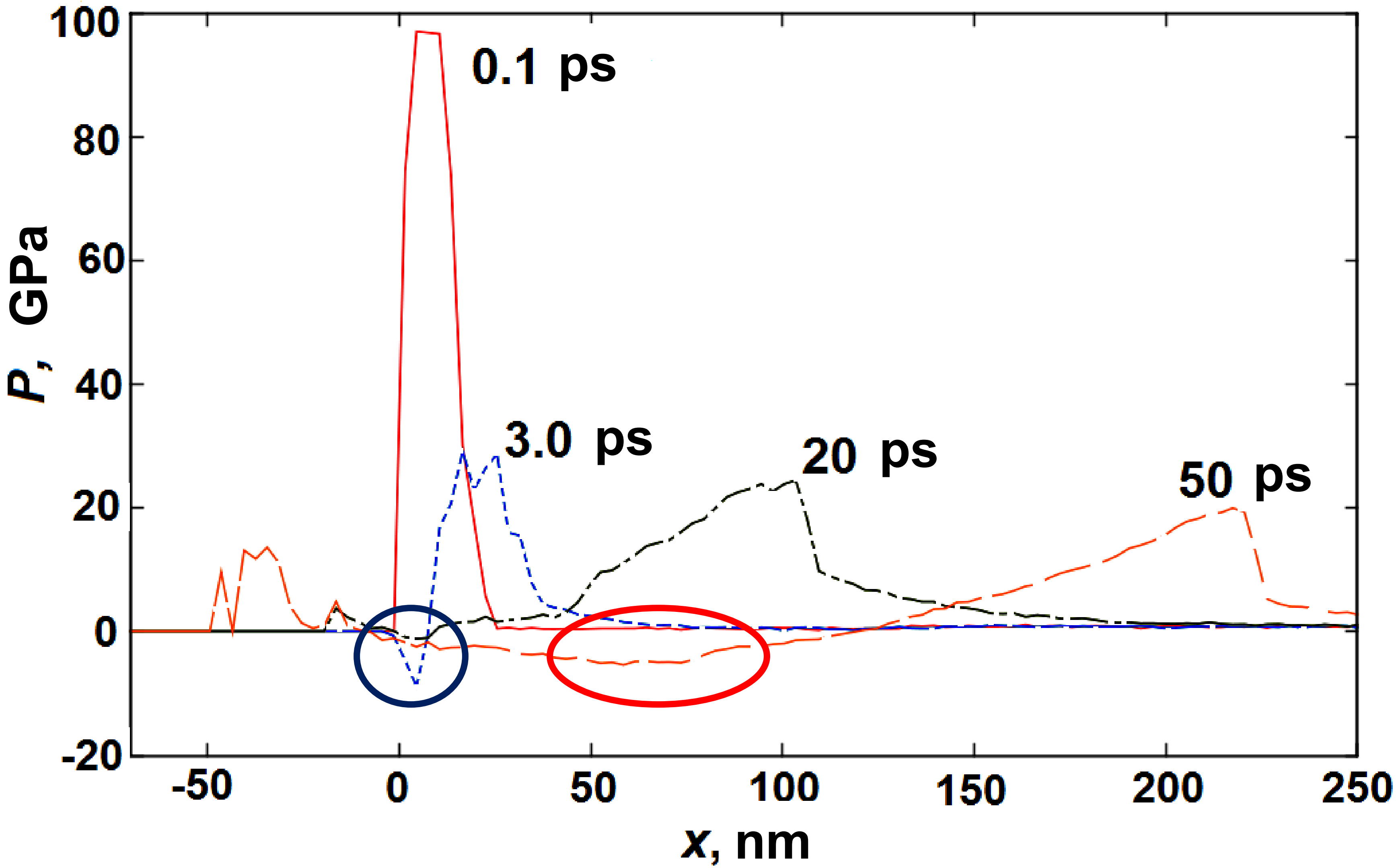

F = 1600 J/m²

# "short" ablation mechanism




F = 1600 J/m$^2$

# "short" and "long" ablation mechanisms




F = 1600 J/m$^2$

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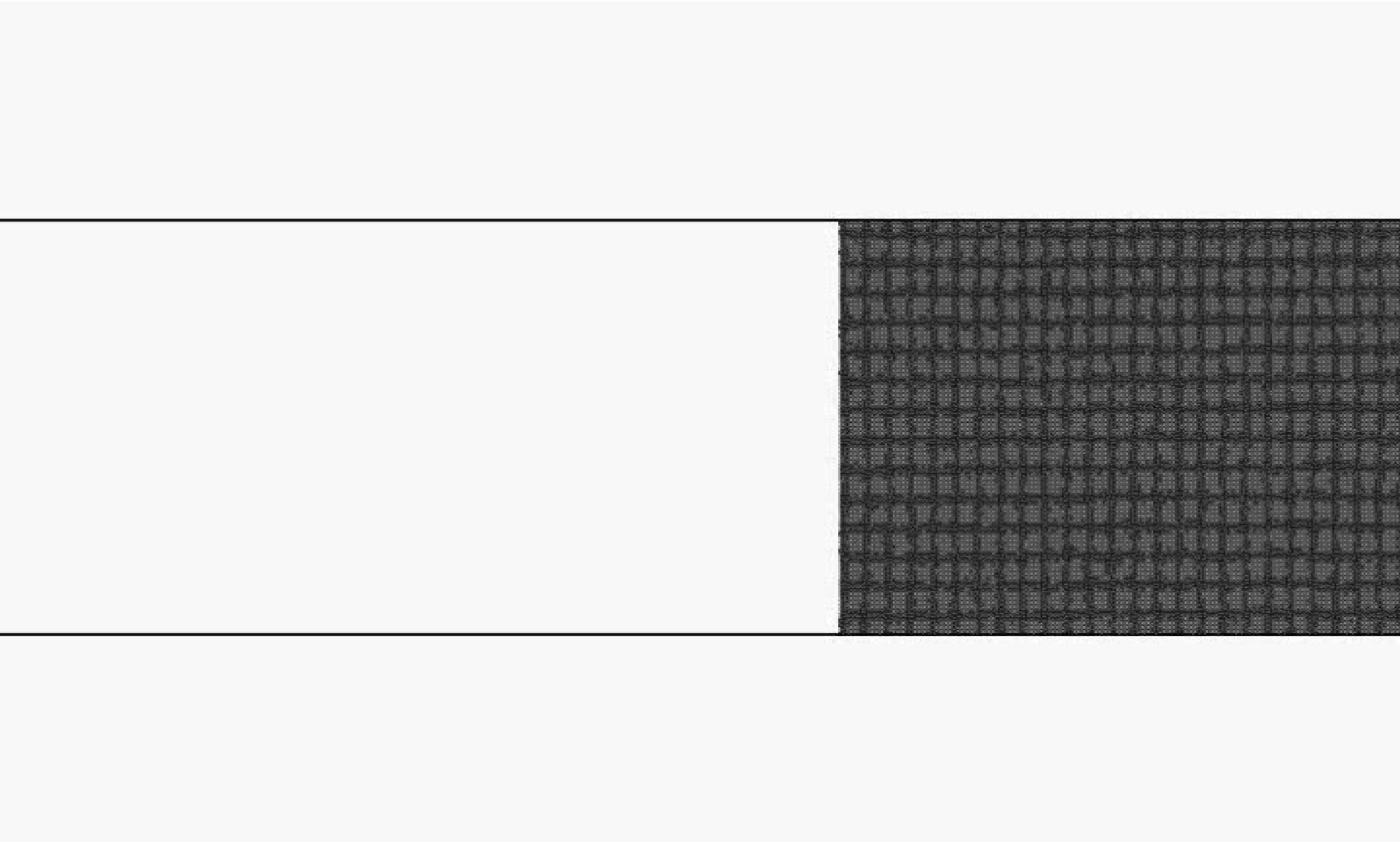
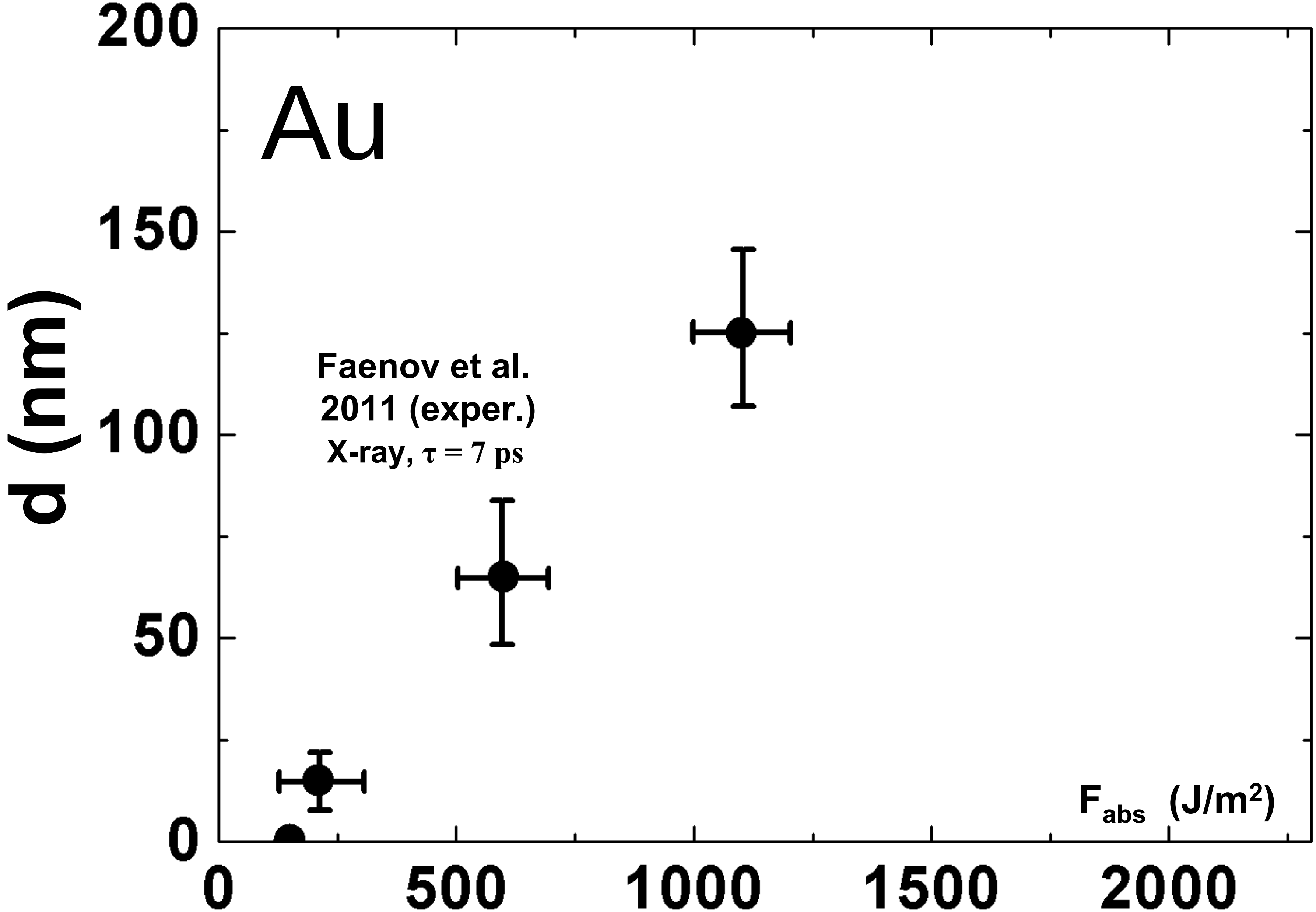


F = 1600 J/m$^2$

# Dependence of crater depth on absorbed fluence



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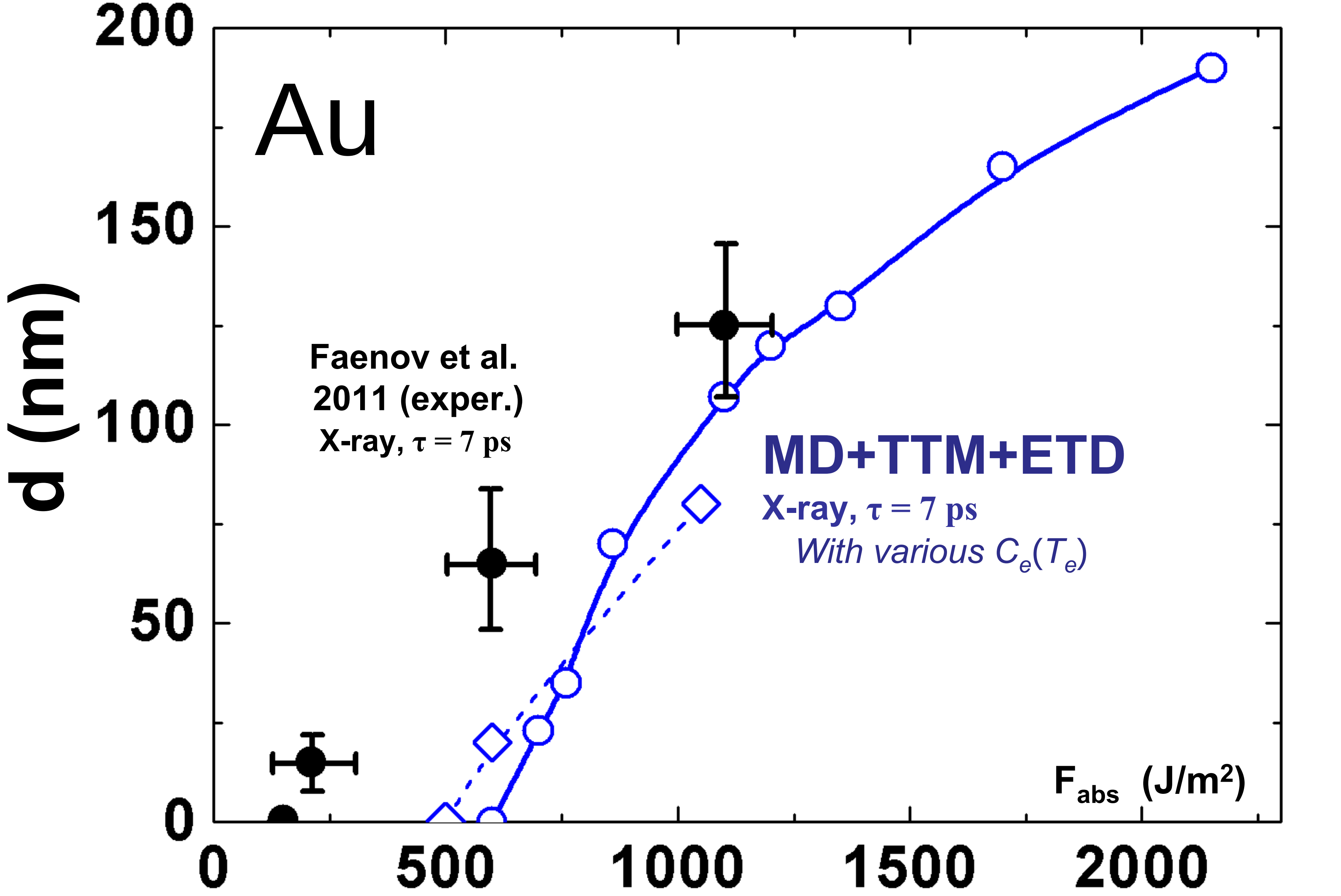

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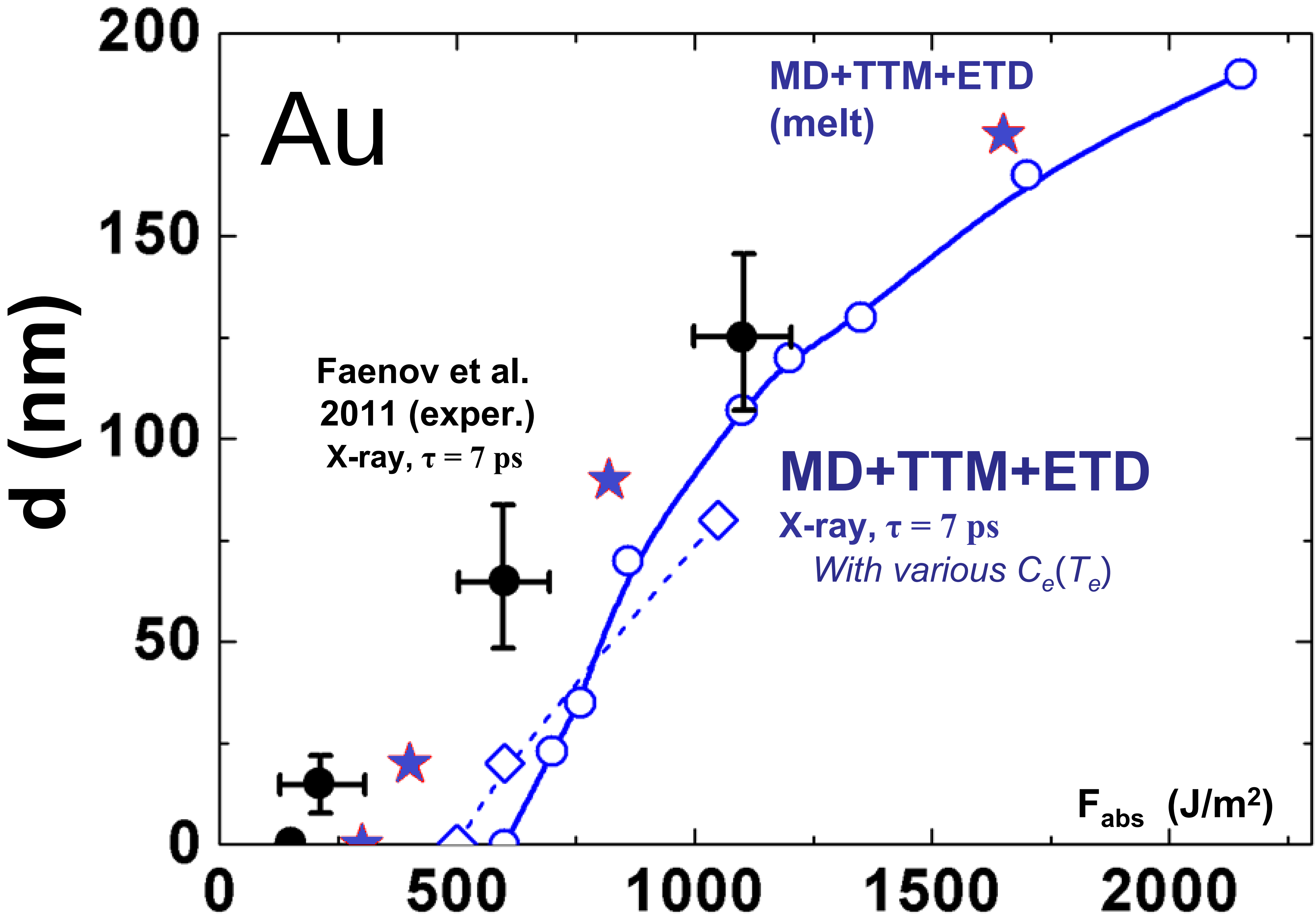
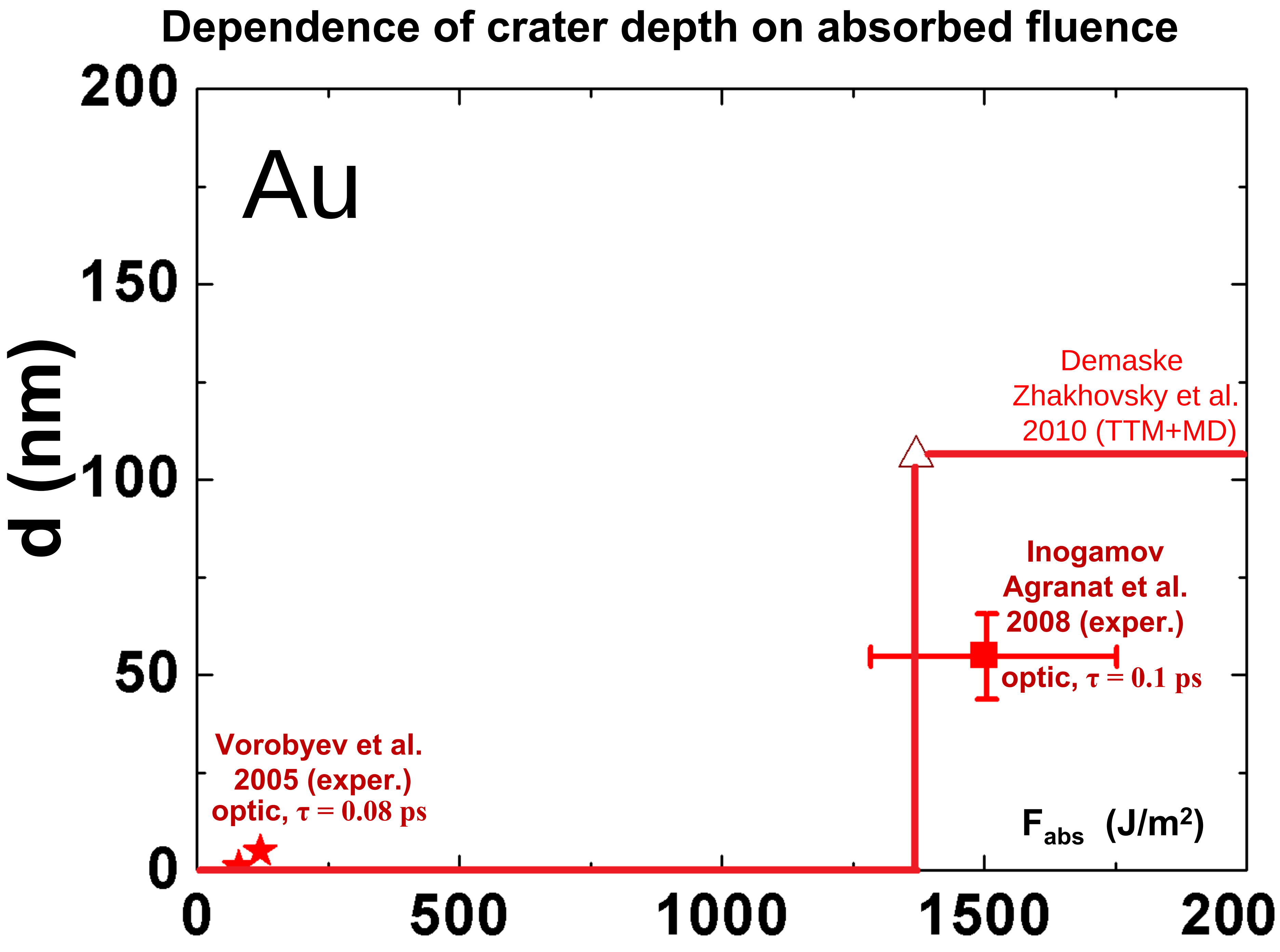
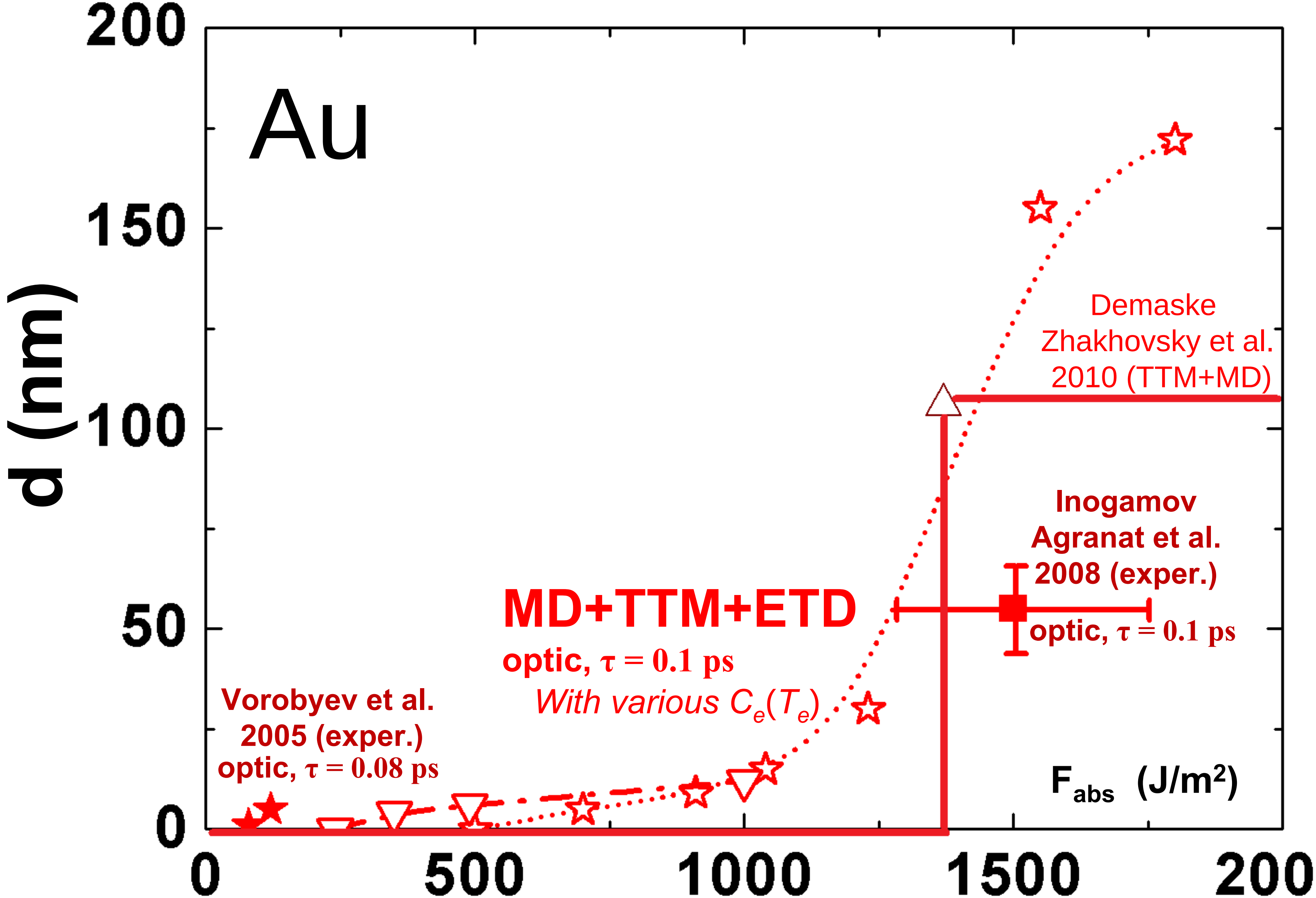




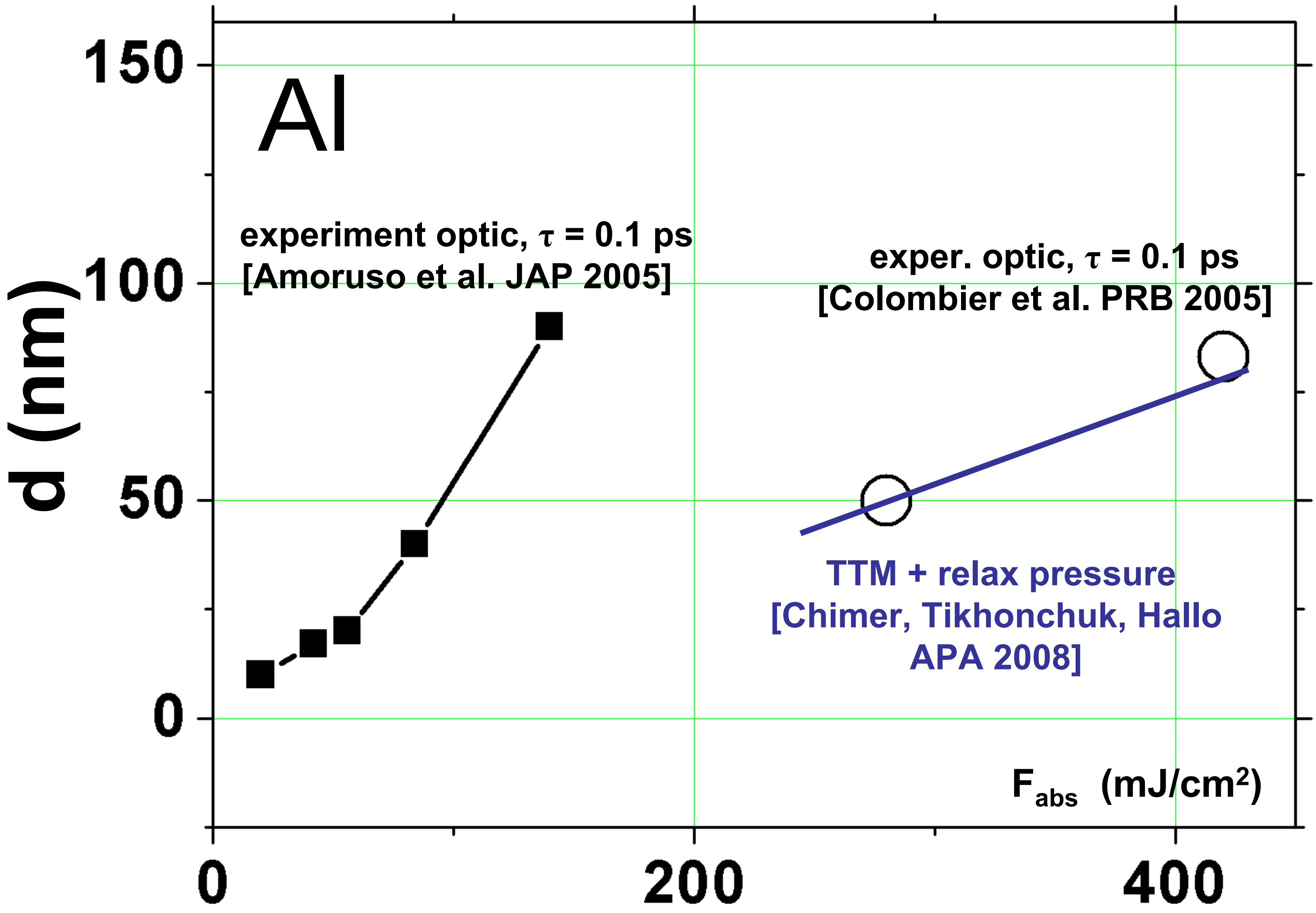
Dependence of crater depth on absorbed fluence
Au
d (nm)
200
150
100
50
0
0
500
1000
1500
200
Demaske
Zhakhovsky et al.
2010 (TTM+MD)
Inogamov
Agranat et al.
2008 (exper.)
optic, τ = 0.1 ps
Vorobyev et al.
2005 (exper.)
optic, τ = 0.08 ps
F abs (J/m2)

Dependence of crater depth on absorbed fluence

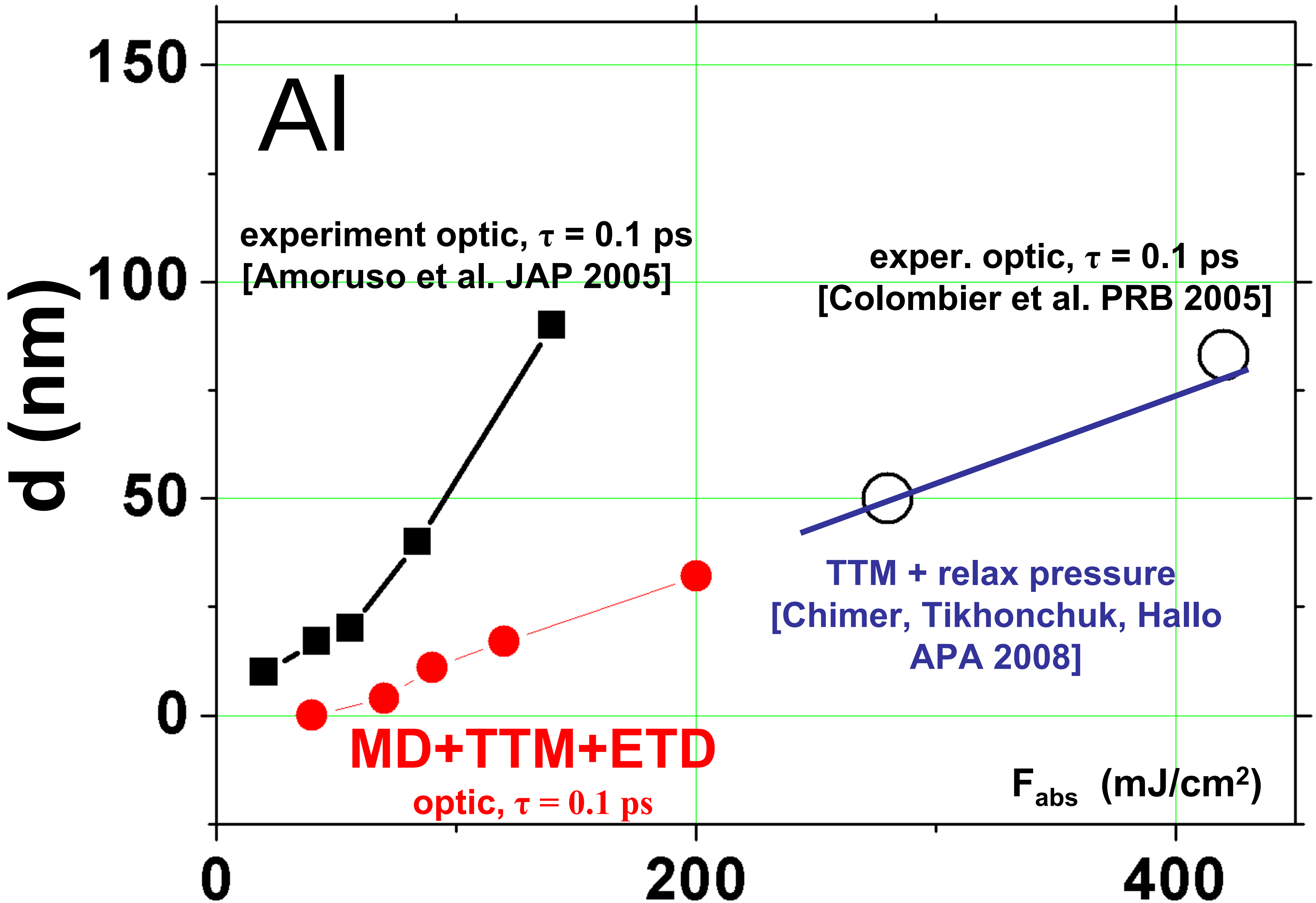


# Atomistic simulation of laser ablation of aluminum

# Dependence of crater depth on absorbed fluence



# Dependence of crater depth on absorbed fluence



# Conclusions

1. The electronic-temperature-dependent EAM potential for gold has been developed that allows a consistent description of the electronic pressure effects in atomistic simulations together with the TTM approach.

2. The threshold fluence of ablation has been calculated for different fs-ps pulse types with good agreement with experimental data for optical and X-ray ablation.

3. The resulting electron-driven ‘short’ ablation is shown to be the ablation mechanism at low fluences and sub-ps pulses.

S.V.Starikov, V.V.Stegailov, G.E.Norman et al. //
JETP Letters, Vol. 93. Issue 11, pp.719-725 (2011)

G.E.Norman, S.V.Starikov, V.V.Stegailov //
JETP, Vol. 141. Issue 4, pp.1-15 (2012) (in print)

2716 J. Opt. Soc. Am. B/Vol. 14, No. 10/October 1997

# Ablation of metals by ultrashort laser pulses

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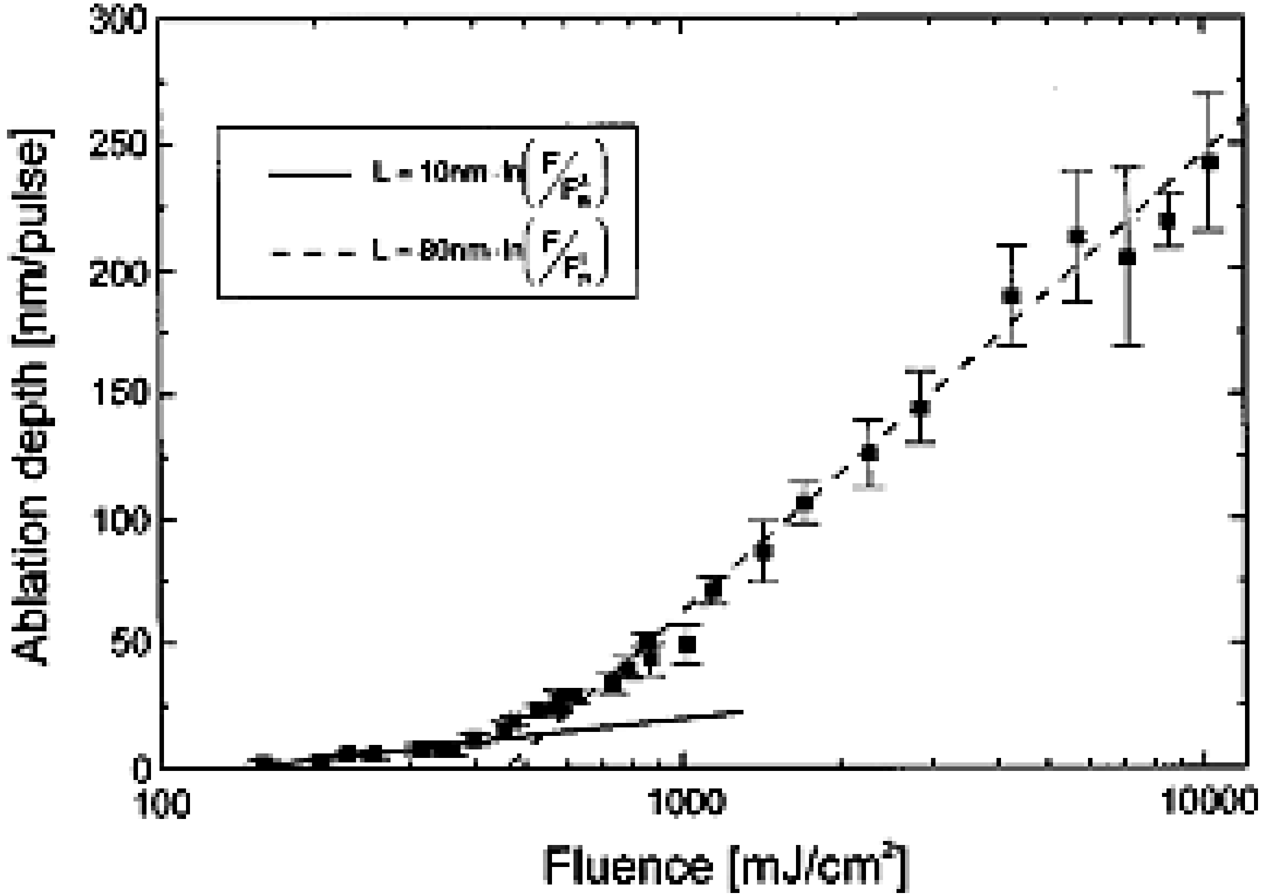
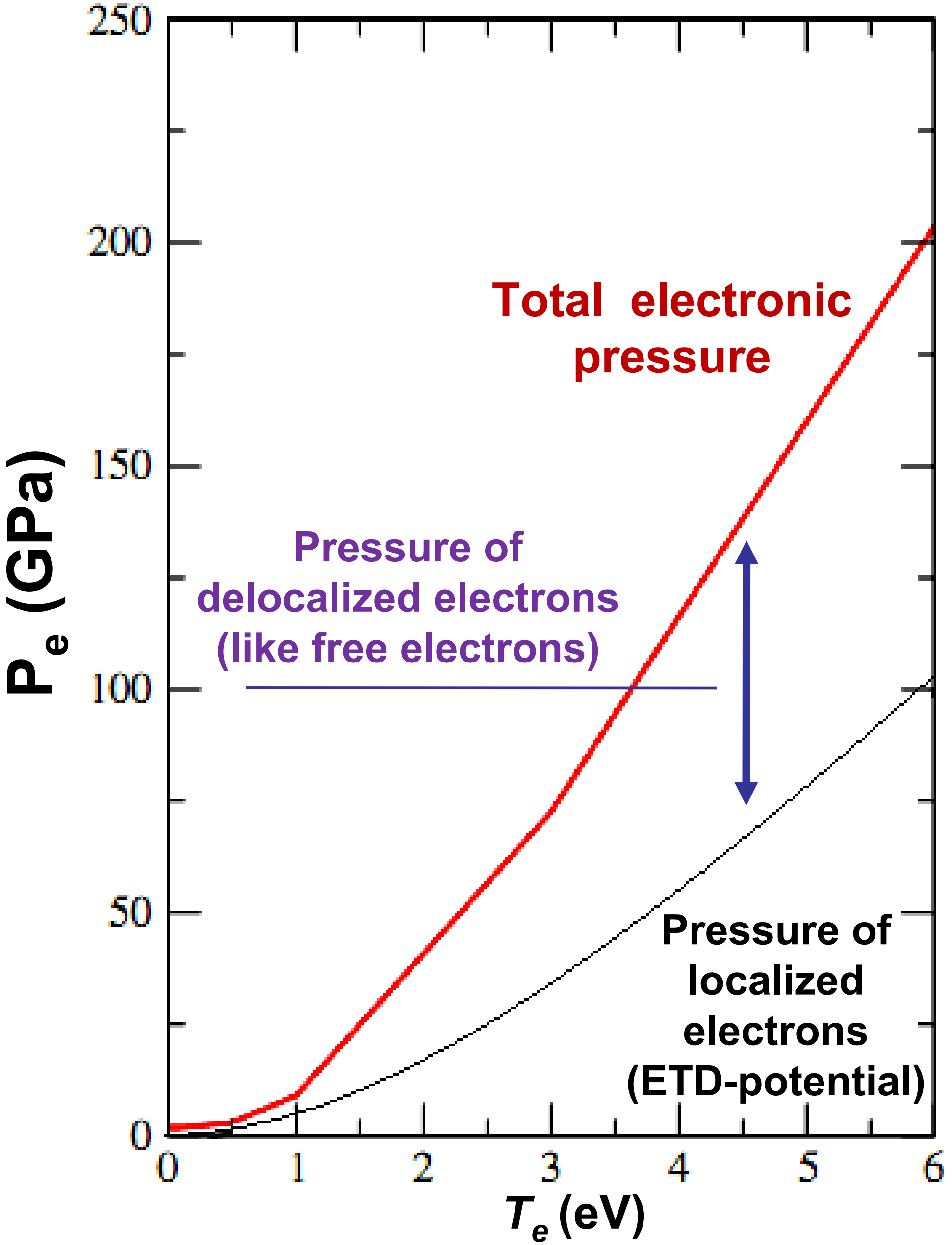


# Electronic pressure in gold




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

Like gas of free electrons

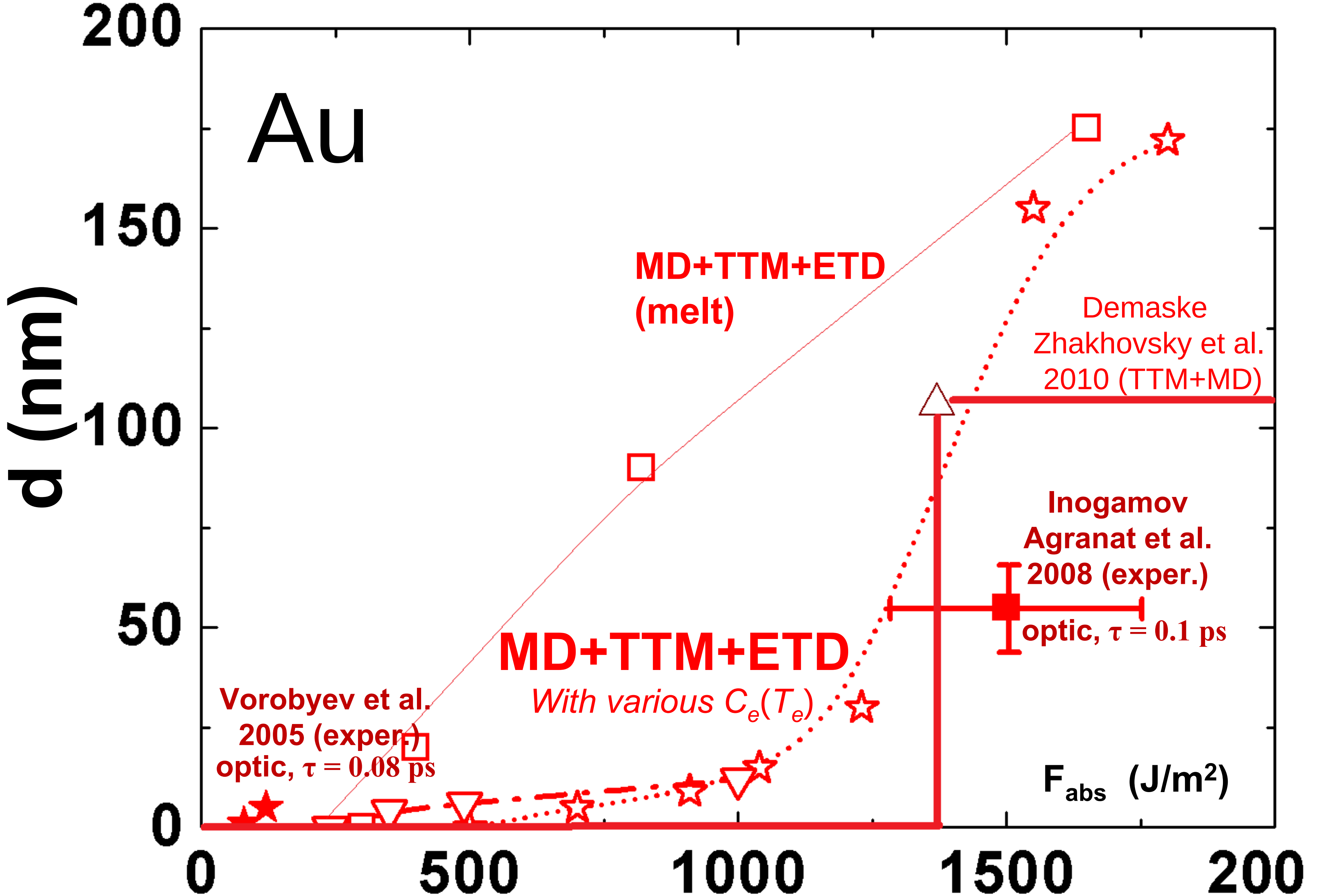
$$P_{e\ \text{free}} = A \cdot C_e T_e = B \cdot T_e^2$$

$$\mathbf{B} = \frac{\nabla P_{e\ \text{free}}}{\rho_{ion}}$$

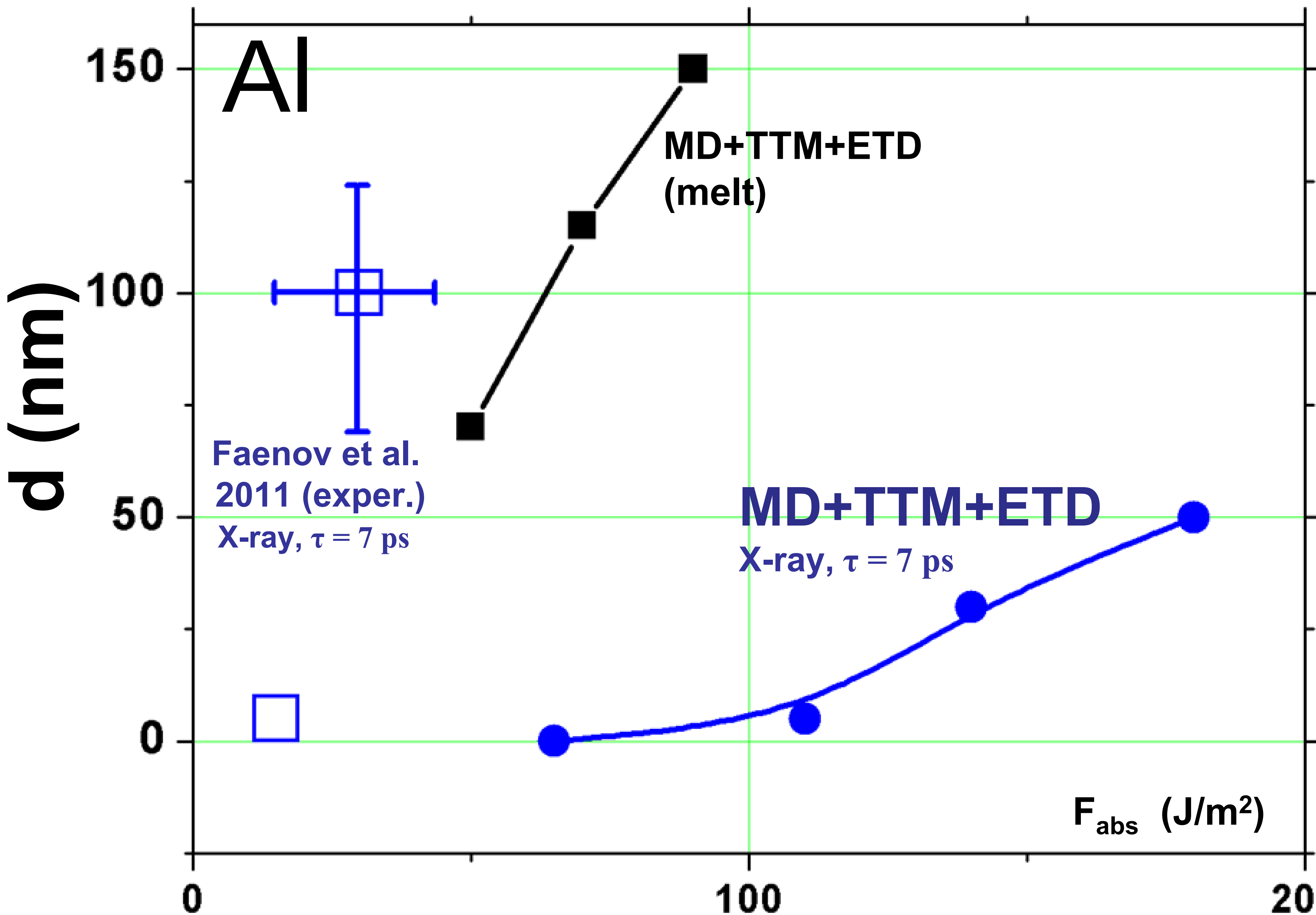
**Electron blast force**

[Falkovsky, Mishchenko , JETP (1999)]

# Dependence of crater depth on absorbed fluence



## Dependence of crater depth on absorbed fluence



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

$$Q=I(t)\exp(-x/l)$$

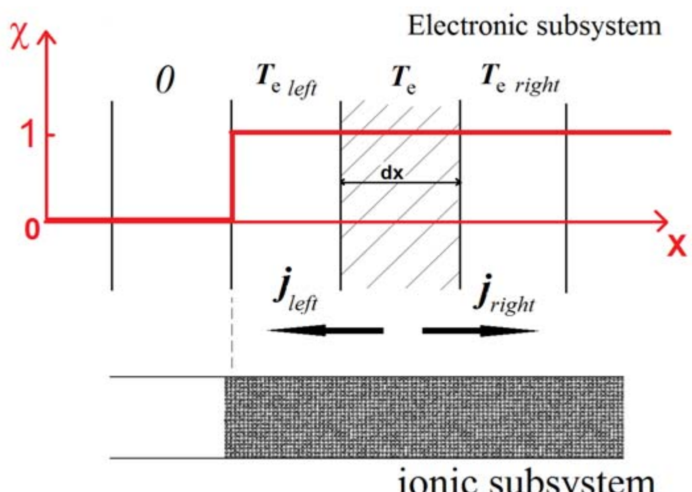
$$C_e(T_e)=C_0+C_1\cdot T_e+C_2\cdot T_e^2$$

$$k_e(T_e)=k_0+k_1\cdot T_e+k_2\cdot T_e^2+k_3\cdot T_e^3$$

$$\nabla\left(k_e(T_e)\cdot\nabla T_e\right)=\frac{j_{right}-j_{left}}{dx}=\frac{\chi(x)\cdot k_e(T_e)\frac{dT_e}{dx}\Big|_{right}-\chi(x)\cdot k_e(T_e)\frac{dT_e}{dx}\Big|_{left}}{dx},$$

$$\chi(x)\cdot k_e(T_e)\frac{dT_e}{dx}\Big|_{right}=\chi(x_{right})\cdot k_e\left(\frac{T_{e,right}+T_e}{2}\right)\cdot\frac{T_{e,right}-T_e}{dx},$$

$$\chi(x)\cdot k_e(T_e)\frac{dT_e}{dx}\Big|_{left}=\chi(x_{left})\cdot k_e\left(\frac{T_{e,left}+T_e}{2}\right)\cdot\frac{T_e-T_{e,left}}{dx},$$



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# Integrated continuum-atomistic modeling of nonthermal ablation of gold nanofilms by femtosecond lasers

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hot electron blast force.[6,12] However, the hot electron blast force has never been included in the modeling of ultrafast laser material ablation.[8,9,15–17]

A better understanding of the ultrashort thermomechanical behavior of materials can enable further optimization of the laser material processing. The present work investigates the ultrafast nonthermal laser ablation of Au nanofilms using a coupled two-temperature model (TTM) and molecular dynamics (MD). This approach is well suited for ultrafast thermomechanical response of nanofilms since it does not require *a priori* knowledge of lattice thermomechanical properties.

The TTM-MD model for ultrafast laser heating of metals is formulated as[8]

$$C_e(T_e)\frac{\partial T_e}{\partial t} = \nabla[K_e(T_e)\nabla T_e] - G(T_e - T_l) + S, \quad (1)$$

$$m_i\frac{d^2\boldsymbol{r}_i}{dt^2} = \boldsymbol{F}_i - \xi m_i \boldsymbol{v}_i^T, \quad (2)$$

$$\xi = \frac{G(T_l - \bar{T}_e)V_c}{\sum_{k=1}^{N_V} m_k(\boldsymbol{v}_k^T)^2}, \quad (3)$$

where $t$ denotes time, $C$ is heat capacity, $T$ is temperature, $K$

The hot electron blast force $\boldsymbol{B}$ generated in the early nonequilibrium thermal state is given by[18]

$$\boldsymbol{B} = 2\nabla(C_eT_e)/3. \quad (4)$$

To include the blast force in the MD simulation, Eq. (2) is modified to

$$m_i\frac{d^2\boldsymbol{r}_i}{dt^2} = \boldsymbol{F}_i - \xi m_i \boldsymbol{v}_i^T + \frac{\boldsymbol{B}V_c}{N_V}. \quad (5)$$

The MD model for the Au nanofilms is created from a bulk fcc Au crystal with $x$, $y$, and $z$ denoting the [1 0 0], [0 1 0], and [0 0 1] crystallographic directions, respectively. Let $z$ be the film thickness direction. The initial dimensions of the model along $x$, $y$, and $z$ are $4.08\times4.08\times100$ nm$^3$ with free boundaries in the $z$ direction and periodic boundaries in the $x$ and $y$ directions. The problem is simplified as a uniaxial strain case[6,8] with the laser source term in Eq. (1) given by[6,9]

$$S(z,t) = 0.94\frac{J_{\text{abs}}}{t_pz_s}\exp\left[-\frac{z}{z_s} - 2.77\left(\frac{t-2t_p}{t_p}\right)^2\right], \quad (6)$$

where $J_{\text{abs}}$ is the absorbed laser fluence, $t_p$ is the laser duration, and $z_s$ is the optical penetration depth. The embedded atom method[19] is used for the interatomic interactions, and the stresses are calculated by the virial theorem.[20] Before the laser irradiation, the MD system is thermally equilibrated to

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# Heating model for metals irradiated by a subpicosecond laser pulse

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We propose a model describing the heating and ablation of a metallic target irradiated by a subpicosecond laser pulse. It takes into account the temperature equilibration between the electrons and ions and the density variation of the target material during the heating process. A simple analytical equation of state is developed, which allows one to calculate the total pressure in the he
The thermodynamic behavior of a nonequilibrium syst
cohesion limits are introduced. The model is applied for a
a sub-ps laser pulse. Aluminum and copper targets are c
process is due to breaking the nonequilibrium cohesion li
subpi-
e short
ons, in
trongly

sider $Z$ as parameter for each material. The ion pressure is written in a standard form

$$P_i = \Gamma(V) k_B T_i / V, \tag{11}$$

where $\Gamma(V)$ is the Grüneisen factor, a function of $V$, which will be defined later with respect to the pressure characteristics at the critical point. To simplify the following equations, $V$ is normalized by $V_0$, the ion-specific volume at $T_i=0$. It is defined as $V_0 = A/N_A\rho_0$, where $A$ is the molar mass, $N_A$ is the Avogadro number, and $\rho_0$ is the cold mass density. The temperatures $k_B T_{i,e}$ are normalized by the Fermi energy at zero temperature, $\varepsilon_{F_0}$. The normalized quantities are denoted by an overbar—for example, $\bar{P}_i$.

$$\bar{P}_e = \frac{2}{5} Z \bar{V}^{-5/3} \left( 1 + \frac{5\pi^2}{12} \bar{V}^{4/3} \bar{T}_e^2 \right). \tag{13}$$

For a cold metal at low temperatures, chemical bonds support the equilibrium. In order to account for this effect, following More *et al.*,[11] we account for the binding pressure in a semiempirical form

$$\bar{P}_b = - b_1 b_2 \bar{V}^{-2/3} \exp[b_2(1 - \bar{V}^{1/3})]. \tag{14}$$

The total normalized pressure is the sum of Eqs. (11), (13), and (14), $\bar{P} = \bar{P}_i + \bar{P}_e + \bar{P}_b$. Two constants $b_1$ and $b_2$ are defined by the condition of equilibrium at zero temperatures: $\bar{P}=0$ for $\bar{V}=1$ and $\bar{T}_{e,i}=0$. Also we request that our EOS reproduce the average value of the bulk modulus $B_s = -V(\partial P/\partial V)_{T_{e,i}}$ in our domain of temperatures. Then

$$b_1 = \frac{4Z^2}{30Z - 75\bar{B}_s}, \quad b_2 = 3 - \frac{15\bar{B}_s}{2Z}. \tag{15}$$

Here $\bar{B}_s$ is normalized by $\varepsilon_{F_0}/V_{i0}$. We note here that in our EOS the repulsive force is provided only by the Pauli principle in the electron pressure. The interatomic electrostatic repulsion is neglected. Strictly speaking, this approach is