SIMULATION OF ABSORPTION OF FEMTOSECOND LASER PULSES IN SOLID-DENSITY COPPER

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Annual Workshop on the Non-ideal Plasma Physics

November 23-24, 2011, Moscow, Russia

Summary

We present a simulation of absorption of femtosecond laser pulses by a copper target. The modeling involved thermodynamic functions calculated by using a first-principles full-potential linear muffin-tin orbital method and chemical-picture-based model of dense plasma utilizing a superconfiguration approach. The results of the simulation are compared to experimental and other theoretical data.

The effects of laser-pulse-induced growth of the electron-ion energy exchange and metal bond hardening are analyzed.

Further work on detailed improvement of the presented theoretical model is discussed.

Energy balance equations of a metal layer absorbing laser light

Two-temperature model [*]:

$$C_e(T_e)\frac{\partial T_e}{\partial t} = \frac{\partial}{\partial z}\left(\kappa_e(T_e,T_i)\frac{\partial T_e}{\partial z}\right) - \Delta Q_{ei}(T_e,T_i) + Q_L(z,t),$$

$$C_i(T_i)\frac{\partial T_i}{\partial t} = \Delta Q_{ei}(T_e,T_i)$$

$$\Delta Q_{ei} = \gamma(T_e)\cdot(T_e - T_i),$$

$$Q_L(z,t) = \omega_L \,\mathrm{Im}\left[\varepsilon(z,\omega_L,t)\right]\frac{|\mathbf{E}(z,t)|^2}{8\pi}$$

[*] S.I. Anisimov, B.L. Kapeliovich, T.L. Perel'man, Sov. Phys. JETP, **39**, 375 (1974); D. Fisher et al. PRE **65**, 016409 (2001)

Helmholtz equation for the electric field amplitude

Maxwell eqs. for the normally incident fs-laser EMW

Helmholtz eq. for the electric field amplitude *E*(*z*,*t*)

$$\frac{d^2E(z,t)}{dz^2} + \frac{\omega_L^2}{c^2}\,\varepsilon(z,\omega_L,t)\,E(z,t) = 0,$$

$$\varepsilon = 1 + 4\pi \sum_Z n_Z\,\alpha_Z + i\,\frac{4\pi}{\omega_L}\,\sigma, \qquad [*]$$

$$\varepsilon = 1 + \delta\varepsilon_1 + i\delta\varepsilon_2 - \frac{\omega_p^2}{\omega_L\,(\omega_L + i\nu_{\text{eff}})} \qquad \text{cf. } [\dagger]$$

[*] R.M. More, H. Yoneda, H. Morikami. JQSRT, **99**, 409 (2006); J. Clérouin et al. PRB, **71**, 064203 (2005).
[†] M.V.Veysman et al. JPB, **41**, 125704 (2008)

Momentum relaxation rate of conduction electrons

$$\nu_{\text{eff}} = \min\left\{\nu_{\text{met}}, \nu_{\text{Sp}}, \nu_{\max}\right\} \rightarrow \nu_{\text{eff}} = \nu_{\max}\left[1 + \left(\frac{\nu_{\max}}{\nu_{\text{met}}}\right)^6 + \left(\frac{\nu_{\max}}{\nu_{\text{Sp}}}\right)^6\right]^{-1/6} \quad [*]$$

1. $\nu_{\text{met}} = \nu_{e,ph} + \nu_{e,e}$

Eidmann et al. [†]: for ion (lattice) $\theta_D \le T_i < T_m$

$$\nu_{e,ph} \approx k_{e,ph}\frac{e^2 k_B T_i}{\hbar^2 \upsilon_F}$$

Lifshitz, Pitaevskii [‡]: @ $T_e \sim E_F$

$$\nu_{e,e} \approx \sqrt{\frac{2E_F}{a_l^2 m_e}} \cdot \left(\frac{k_B T_e}{E_F}\right)^2$$

2. $\nu_{\text{Sp}} = \dfrac{4\sqrt{2\pi}}{3}\dfrac{\langle Z\rangle e^4 m_e n_e}{(m_e k_B T_e)^{3/2}}\,\Lambda, \qquad \Lambda_{\min}=2 \quad [\bigstar]$

3. $\nu_{\max} = \upsilon_e / r_0^{(i)}, \qquad \upsilon_e = \sqrt{\upsilon_F^2 + 3k_B T_e / m_e} \qquad [*, †]$

[*] M.V.Veysman et al. JPB, **41**, 125704 (2008)
[†] K. Eidmann et al. PRE, **62**, 1202 (2000).
[‡] E.M. Lifshitz, L.P. Pitaevskii. *Physical Kinetics*. (Pergamon, 1981).
[★] Y. Lee, R. More, Phys. Fluids **27**, 1273 (1984).

Laser light absorption

$$A = \frac{1}{F_L}\int_0^{t_{max}} dt \int_0^{\infty} Q_L(z,t)\,dz$$

$t_{max} = 2\tau_L$ for a sine-squared pulse [*] and
$t_{max} \geq 4\tau_L$ for a Gaussian pulse

To calculate A — find the temperature dependences of variables entering energy balance eqs.

Electron heat conductivity $\kappa_e(T_e,T_i)$ — interpolation between the cold-metal and hot-plasma limits [†, ‡]:

$$\kappa_e(T_e,T_i) = \alpha\vartheta_e \frac{(\vartheta_e^2+0.16)^{5/4}\cdot(\vartheta_e^2+0.44)}{(\vartheta_e^2+0.092)^{1/2}\cdot(\vartheta_e^2+\beta\vartheta_i)}. \qquad \vartheta_{e,i} = k_B T_{e,i}/E_F$$

Fitting parameters α & β ⇐ comparison to experimental reference data @ T_0 = 300 K; for Cu α = 377 W/(m K), β = 0.139 [†]

$C_i(T_i)$ ⇐ wide-range semiempirical EOS [★] allowing for the anharmonicity of the ion-lattice vibrations. Reproduces the Dulong-Petit limit $C_i = 3k_B n_i$ @ $T_i \sim T_0$ from which $C_i(T_i)$ slowly decreases with T_i ↑ to the ideal-gas limit.

[*] D. Fisher et al. PRE **65**, 016409 (2001)
[†] S.I. Anisimov, B. Rethfeld. Proc. SPIE **3093**, 192 (1997).
[‡] C. Schäfer, H.M. Urbassek. PRB, **66**, 115404 (2002)
[★] A.T. Sapozhnikov, A.V. Pershina. VANT, Ser.: Metodiki i Programmy... **4**, 47 (1979) [in Russian]

Electron-temperature-dependent properties

All other T_e-dependent properties were found by using the FP-LMTO method [*] and the CP-SC model of dense plasmas [†]

FP-LMTO:

- 3*s*-, 3*p*-, 3*d*-, and 4*s*-electrons of Cu = valence electrons
- exchange-correlation functional with gradient corrections
- 92 bands above the Fermi energy ⇒ accuracy in specific energy ≤ 0.01 eV/atom

Effective average ionization $\langle Z \rangle (T_e)$ in the FEG approximation:

$$\langle Z \rangle (T_e) = \frac{m_e^{3/2}}{\sqrt{2}\pi^2 \hbar^3 n_i} \int_0^\infty g f_e(\mathcal{E}, \mu_e, T_e) \sqrt{\mathcal{E}}\, d\mathcal{E},$$

$$f_e(\mathcal{E}, \mu_e, T_e) = 1/\{1 + e^{(\mathcal{E}-\mu_e)/k_B T_e}\}, \quad g = 2$$

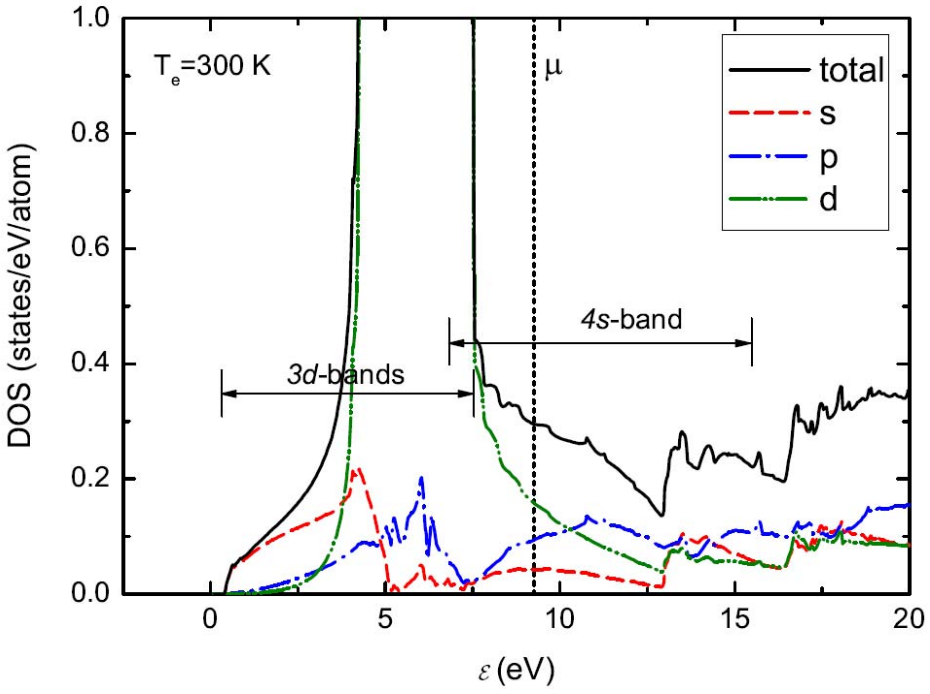
[*] S.Yu. Savrasov. PRB, **54**, 16470 (1996); G.V. Sin'ko, N.A. Smirnov. PRB, **74**, 134113 (2006).
[†] P.A. Loboda, V.V. Popova, A.A. Shadrin. CPP, **49**, 738 (2009).

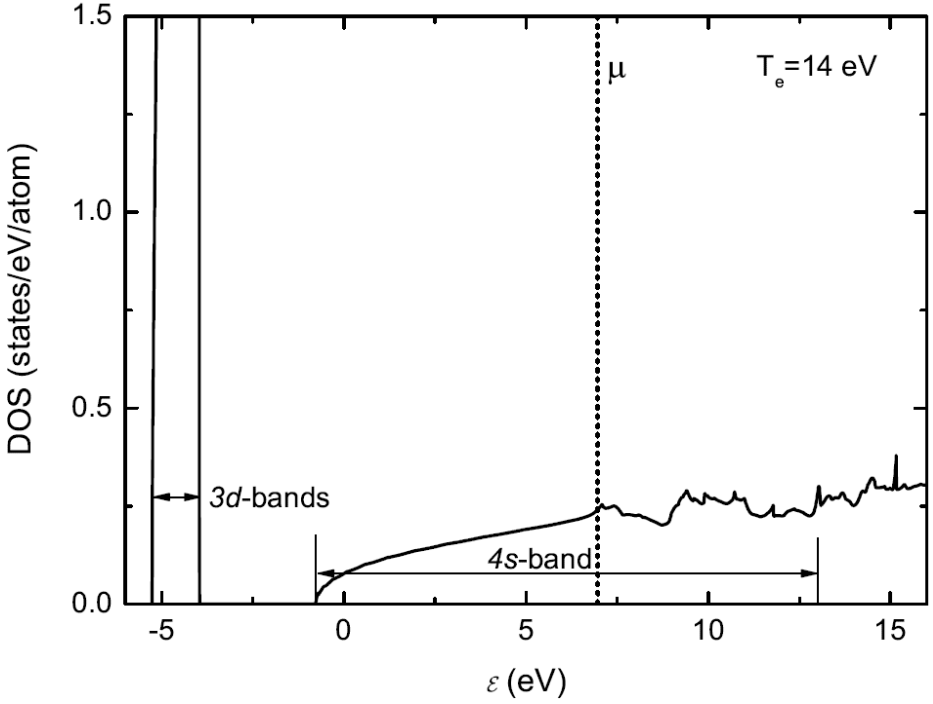
Electron-temperature-dependent properties

FP-LMTO: @ $T_e = T_0$ $E_F = 9.25$ eV, $\langle Z \rangle = 1.51$

(cf. $E_F = 6.9$ eV @ $\langle Z \rangle = 1$ from general estimates) — effect of 3*d*-electrons on μ_e.

FP-LMTO: $T_e \uparrow \Rightarrow$ drastic change of the electron DOS $g(\mathcal{E})$





Electron-temperature-dependent properties

Cu valence-electron DOS structure — very similar to that of Au.

Au: excitation of 5*d*-electrons with $T_e\uparrow \Rightarrow$ decrease of screening of the ion Coulomb potential $\Rightarrow$ metal-bond hardening & ion-lattice stabilization [*].

Experimental confirmation [†, ‡]: @ fs-laser $I_L \leq 10^{13}$ W/cm^2 ultrathin golden foil preserved its crystal structure for several ps.

Hardening of the metal bonding $\Rightarrow$ hardening of the phonon modes $\Rightarrow \theta_D\uparrow$.

Debye-Lindemann theory $\Rightarrow$ $T_m(T_e) \propto \theta_D^2(T_e)\uparrow$

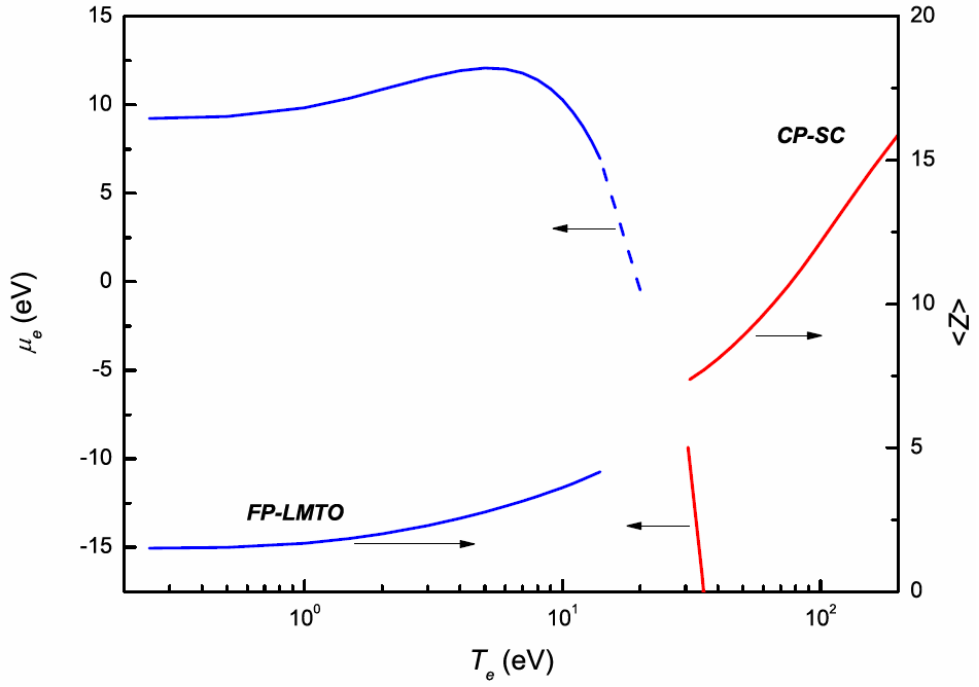
Similar metal-bond hardening with $T_e\uparrow$ should also occur for Cu.

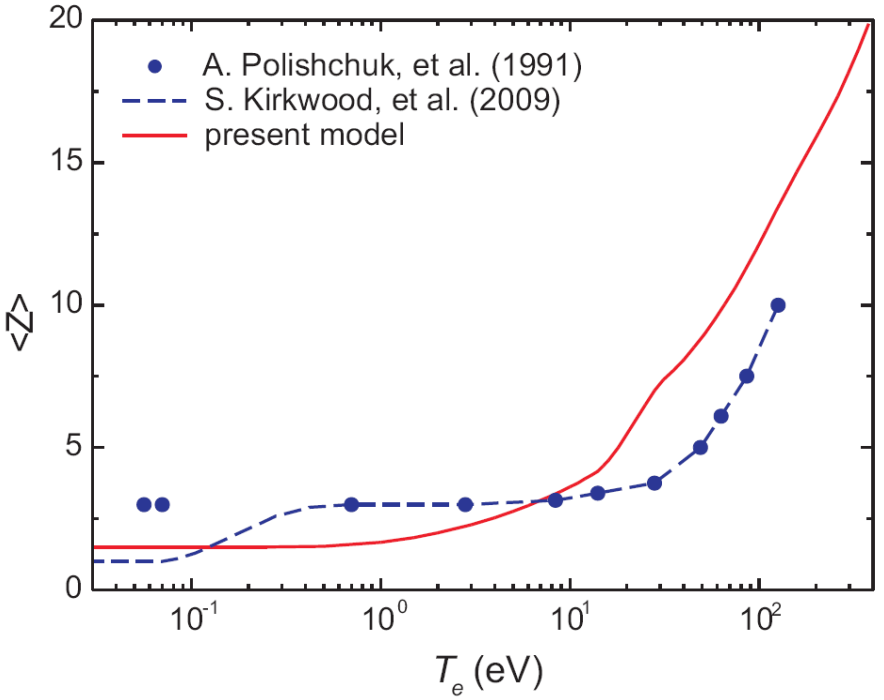
Assumption to initiate the modeling: heated surface layer of Cu is in the crystalline state as long as $k_B T_e \leq 1.5\, E_F \approx 14$ eV

[*] V. Recoules et al. PRL **96**, 055503 (2006).
[†] Y. Ping et al. PoP **15**, 056303 (2008).
[‡] R. Ernstorfer et al. Science **323**, 1033 (2009).

Electron-temperature-dependent properties

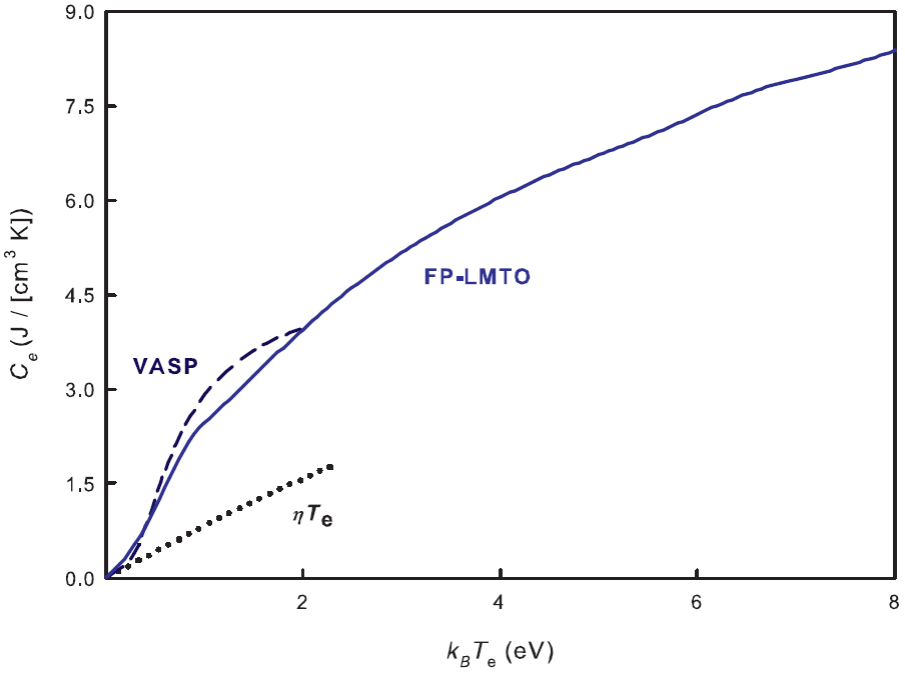
$\mu(T_e)$ and $\langle Z \rangle$ of solid-density Cu:

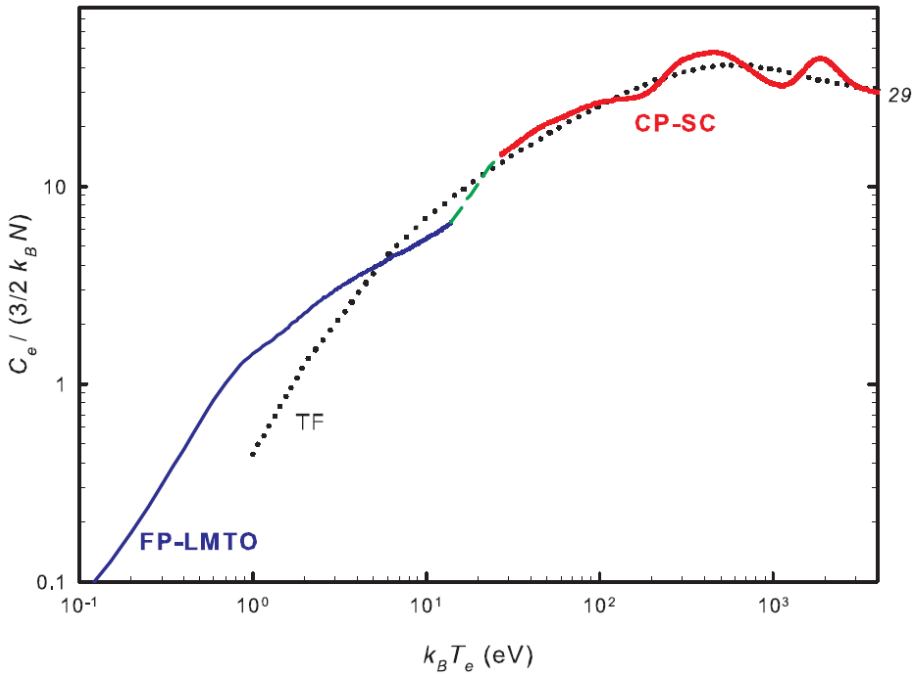




Electron-temperature-dependent properties

Electron heat capacity per unit volume $C_e(T_e) = \partial E_e(T_e)/\partial T_e$





Widely used linear dependence: $C_e = \eta T_e \; (\eta = 96.8 \; \mathrm{J/(m^3\,K^2)})$

Electron-temperature-dependent properties

$G(T_e)$ — electron-ion coupling factor $\gamma(T_e)$ in the crystalline state [*]:

$$G(T_e) = \frac{\pi \hbar k_B \lambda_{ph} \langle \omega_{ph}^2 \rangle}{g(E_F)} \int_{-\infty}^{+\infty} g^2(\mathcal{E}) \left(\frac{\partial f_e}{\partial \mathcal{E}} \right) d\mathcal{E}$$

λ_{ph} — electron-phonon coupling parameter,

$\langle \omega_{ph}^2 \rangle$ — second moment of the phonon spectrum, $\langle \omega_{ph}^2(T_e) \rangle \approx \theta_D^2(T_e)/2$ [*]

1st stage: $\lambda_{ph} \langle \omega_{ph}^2 \rangle$ = 29 meV^2 (pump-probe experiment @ $T_e \leq 1000$ K [†])

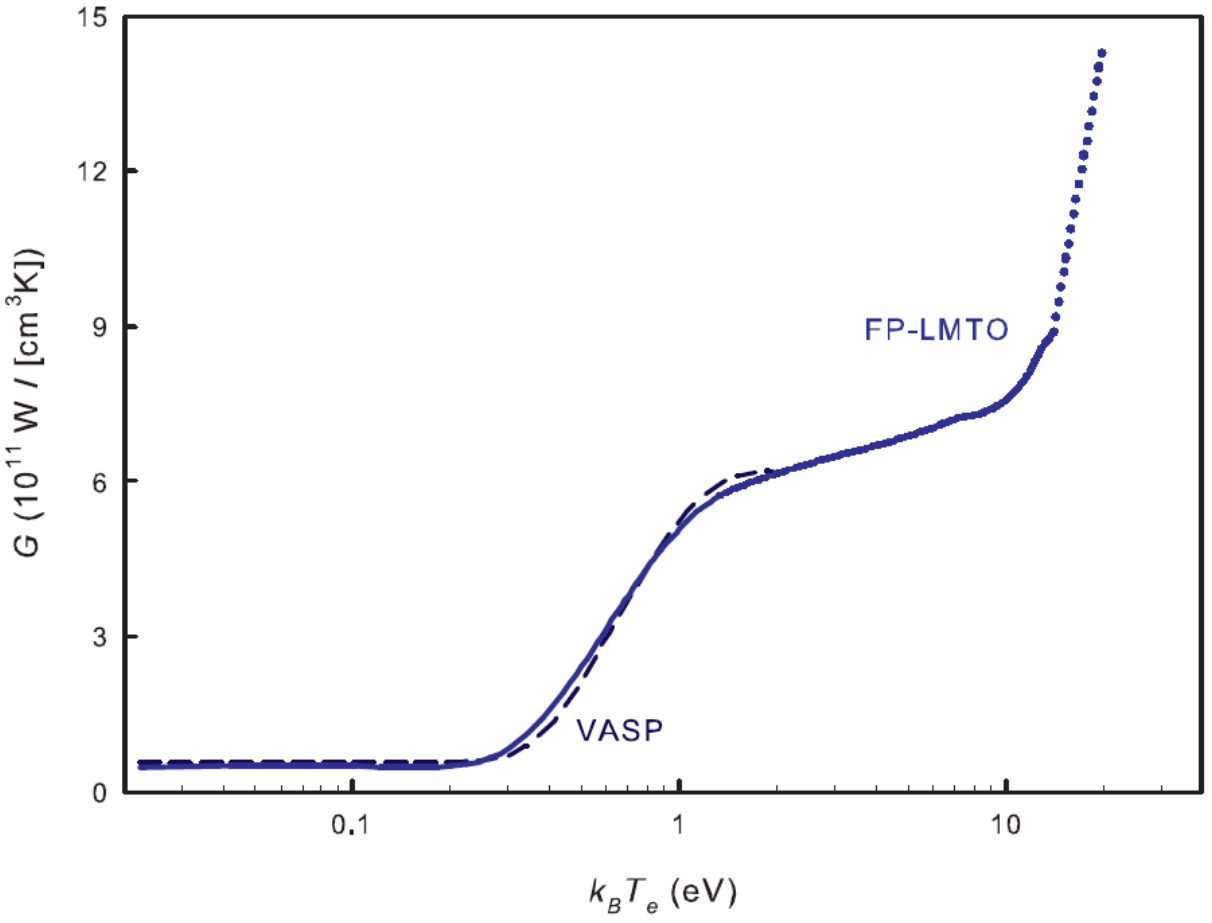
$\gamma_{e,i}(T_e)$ — electron-ion coupling factor $\gamma(T_e)$ in the plasma state [‡]:

$$\gamma_{e,i}(T_e) = \frac{3 m_e k_B}{M^2} \nu_{\text{eff}}^{\gamma}$$

[*] Z. Lin, L.V. Zhigilei, V. Celli. PRB, **77**, 075133 (2008).
[†] S.D. Brorson et al. PRL, **18**, 2172 (1990).
[‡] K. Eidmann et al. PRE, **62**, 1202 (2000).

Electron-temperature-dependent properties

Electron-ion coupling factor $G(T_e)$:



VASP calculation: Z. Lin, L.V. Zhigilei, V. Celli. PRB, **77**, 075133 (2008).

Electron-temperature-dependent properties

Plasma electron frequency:

FP-LMTO: $$\omega_p^2(T_e) = \frac{8\pi e^2\hbar^2}{3\Omega_c}\hat{g}(E_F)\left\langle \upsilon^2(T_e)\right\rangle$$ [*, †]

$\hat{g}(E_F)$ — electron DOS @ the Fermi level / atom / spin state

$\left\langle \upsilon^2(T_e)\right\rangle$ — mean-square band velocity: averaging $\upsilon^2_{\vec{k}\lambda} = \hbar^{-2}[\partial E_\lambda(\vec{k})/\partial \vec{k}]^2$ over all the valence bands λ and wave vectors ***k***, ($E(\boldsymbol{k})$ — T_e-dependent band energy of the crystal).

CP-SC: $$\omega_p^2(T_e) = \frac{4\pi e^2 n_i}{m_e}\langle Z\rangle(T_e)$$

[*] L.F. Mattheiss, L.R. Testardi, W.W. Yao, PRB, **17**, 4640 (1978).
[†] S.Y. Savrasov, D.Y. Savrasov. PRB, **54**, 16487 (1996)

Electron-temperature-dependent properties

Dielectric permittivity:

@ λ_L = 800 nm $\delta\varepsilon_2 = 0$

$\delta\varepsilon_1 \approx \varepsilon_\infty > 0$ ion-core polarizability: weak infrared-freq. dependence, approximated by a constant value — not small for the noble metals @ T_0.

But $\delta\varepsilon_1 \downarrow$ as $T_e \uparrow$ $\Rightarrow$

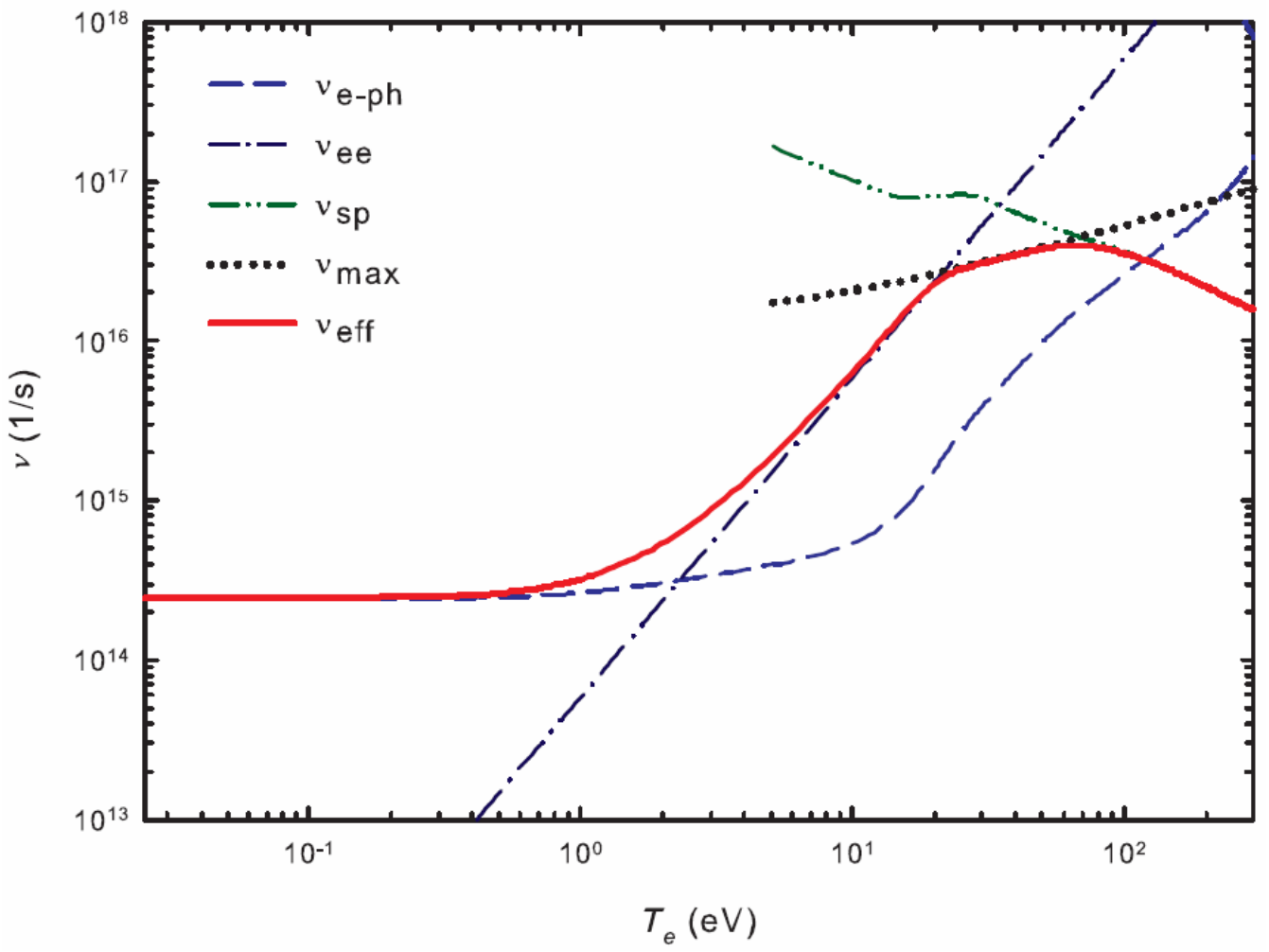
more appropriate to implicitly include the ion-core polarizability @ low T_e by setting $\delta\varepsilon_1 = 0$ and properly choosing $k_{e,ph}$ factor: $\delta\varepsilon_1 \to 0$ as $T_e \uparrow$ —

$\nu_{\text{eff}} \approx \nu_{e,ph} \to \nu_{e,e}$ as the electron temperature grows from T_0 to several eV.

$k_{e,ph} \cong 5.3$ — by fitting the ref. value of the reflection coefficient of metallic Cu @ T_0

Electron-temperature-dependent properties

Collision frequencies:



Simulation of absorption of 50 fs laser pulses by a copper target (1st stage [*])

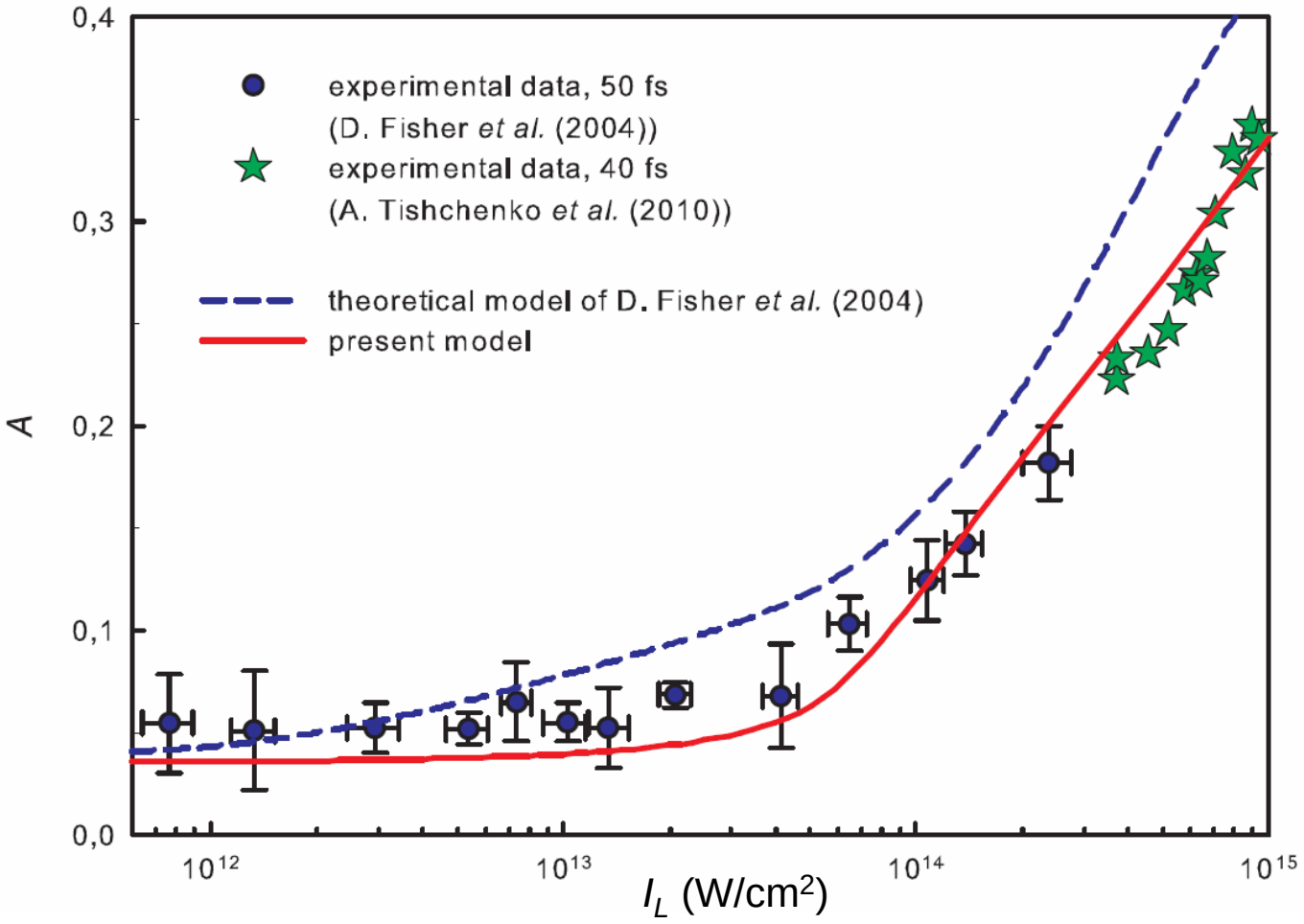
Modified version of the 1D ERA hydrocode [†] — implements new theoretical data on thermodynamic functions of the target material + improved description of relaxation and energy-transfer rates

Table 1: Maximum electron and ion temperatures at the surface of the copper target heated by the 50 fs laser pulses at the intensities $I_L = 10^{13} - 10^{15}$ W/cm^2 as calculated by the ERA code.

I_L, W/cm^2	T_e, eV	T_i, eV
10^{13}	0.97	0.04
10^{14}	10.9	0.4
10^{15}	44.5	5.3

[*] P.A. Loboda, N.A. Smirnov, A.A. Shadrin, N.G. Karlykhanov. HEDP, 7, 361-370 (2011)
[†] N.A. Barysheva, A.I. Zuev, N.G. Karlykhanov, et al., Zh. Vychislit. Mat. I Mat. Fiz. (Sov. J. Of Comput. Math. & Math. Phys **22**, 401 (1982) [in Russian].

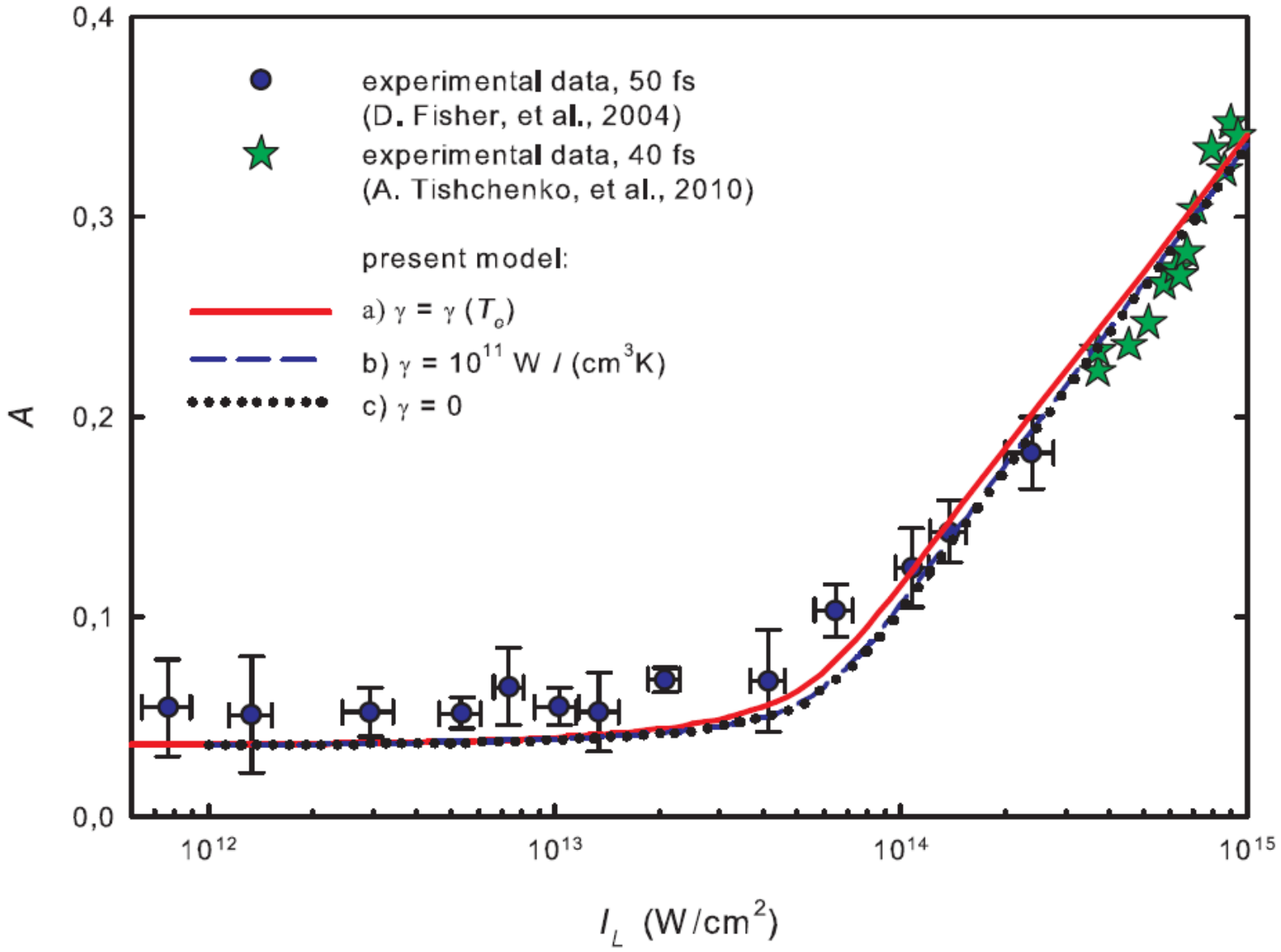
Simulation of absorption of 50 fs laser pulses by a copper target (1st stage)



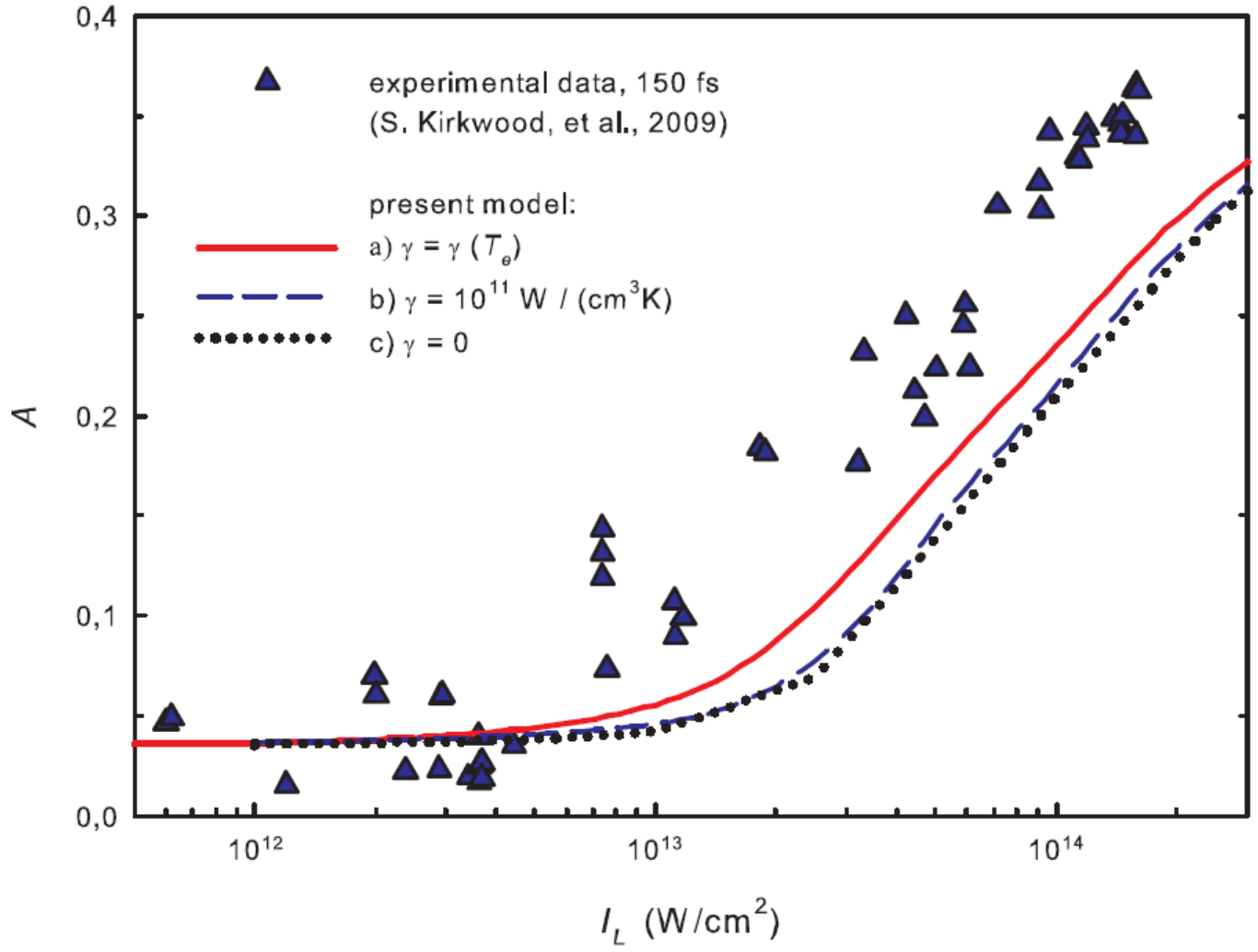
D. Fisher, *et al*. In: Proc. XXVIII ECLIM conf., Rome, Italy (2004); LPB **23**, 391–393 (2005)

A. S. Tishchenko, *et al*. In: Abstr. 10th Intl. conf. “Zababakhin Scientific Talks”, Snezhinsk, Russia (2010)

Simulation of absorption of 50 fs laser pulses by a copper target (1st stage)



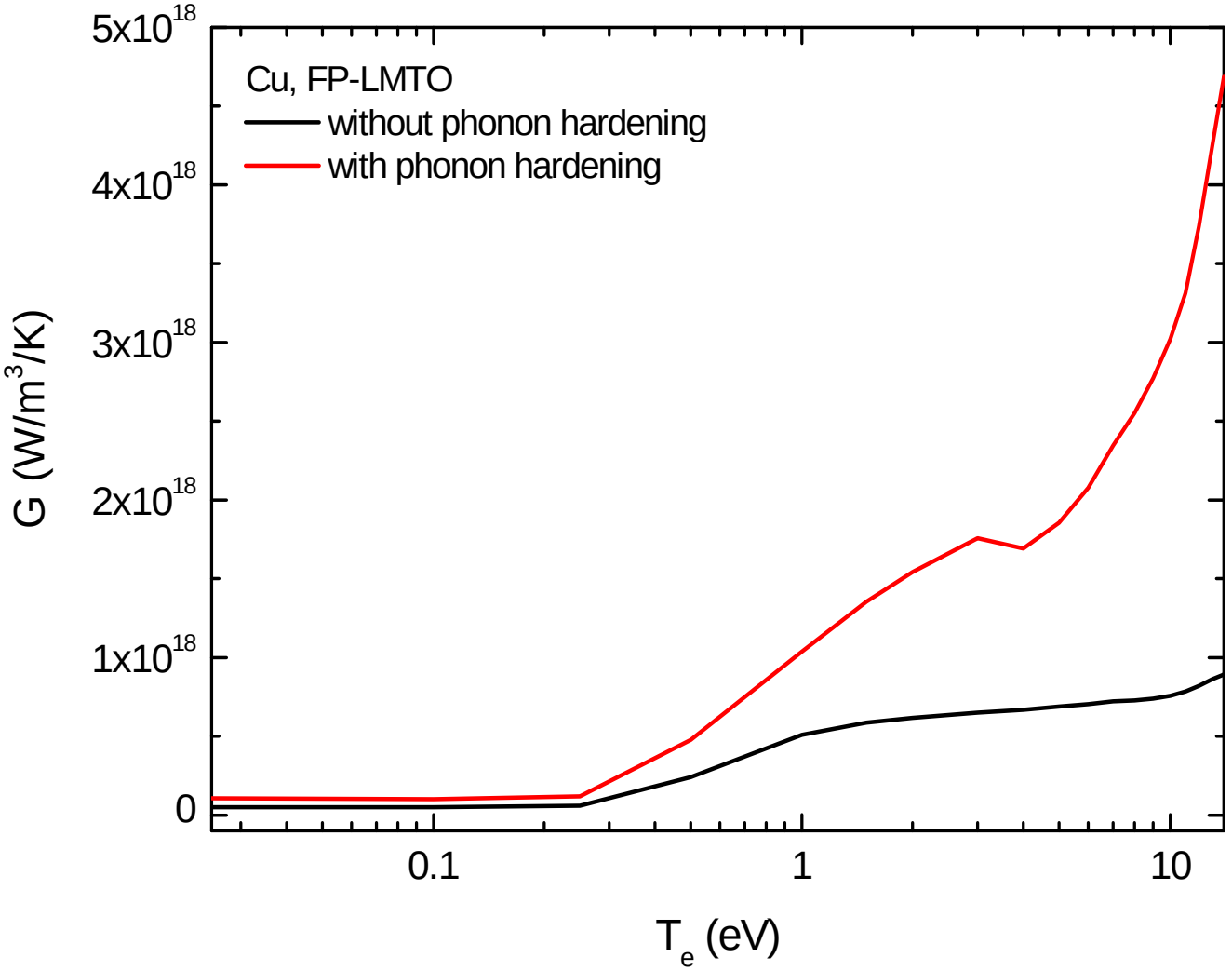
Simulation of absorption of 150 fs laser pulses by a copper target (1st stage [*])



[*] P.A. Loboda, N.A. Smirnov, A.A. Shadrin, N.G. Karlykhanov. HEDP, 7, 361-370 (2011)

S.E. Kirkwood, Y.Y. Tsui, R. Fedosejevs, A.V. Brantov, V.Y. Bychenkov. PRB **79**, 144120 (2009)

Modification of the electron-ion coupling factor due to phonon hardening (2nd stage)



To get $G(T_e)$ with phonon hardening we used:

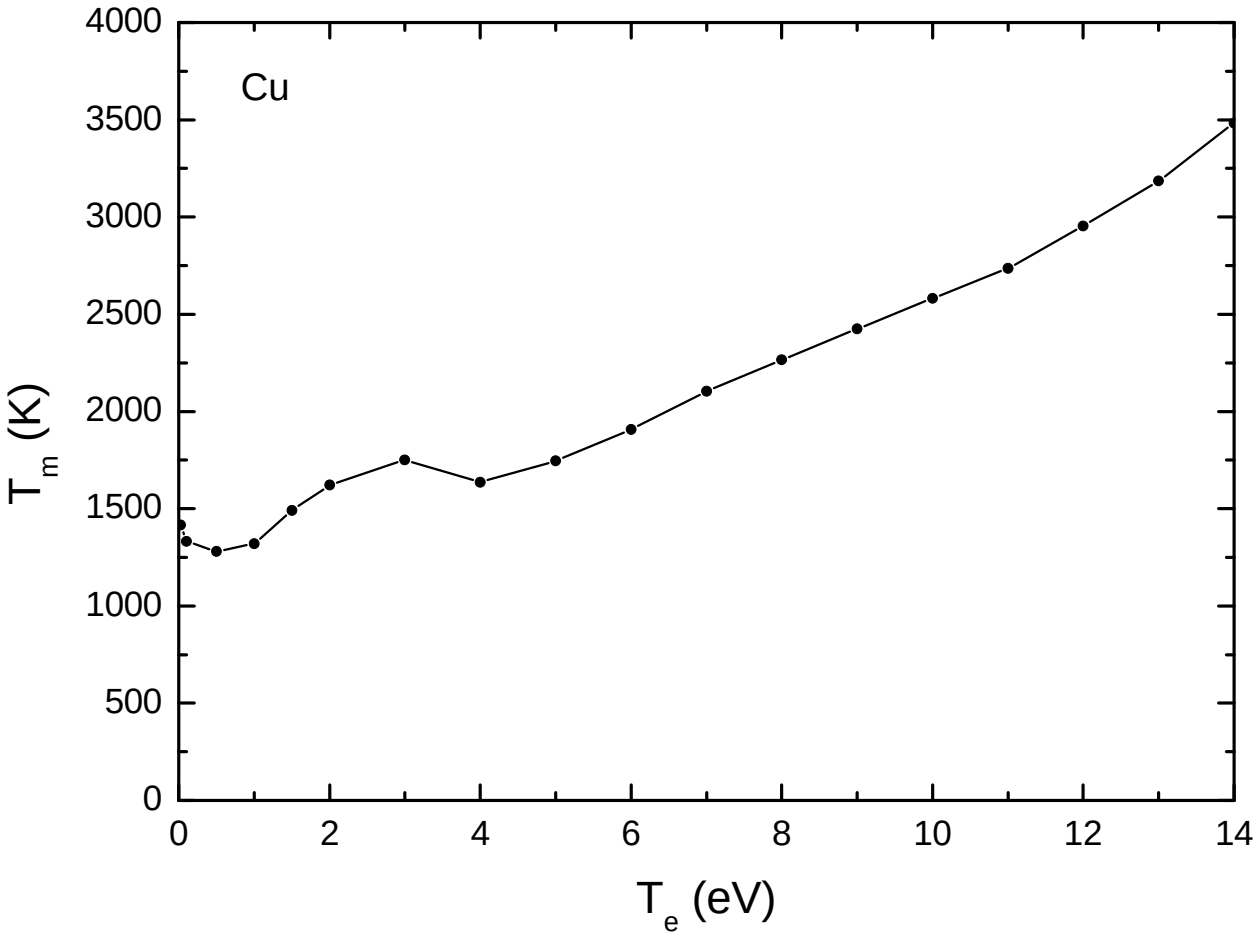
λ_{ph} — electron-phonon coupling parameter was taken from *ab initio* calculations [*]: λ_{ph} =0.14;

$$\langle \omega_{ph}^2(T_e) \rangle \approx \theta_D^2(T_e)/2$$

$\theta_D(T_e)$ was calculated from elastic constants [†] with the FP-LMTO code.

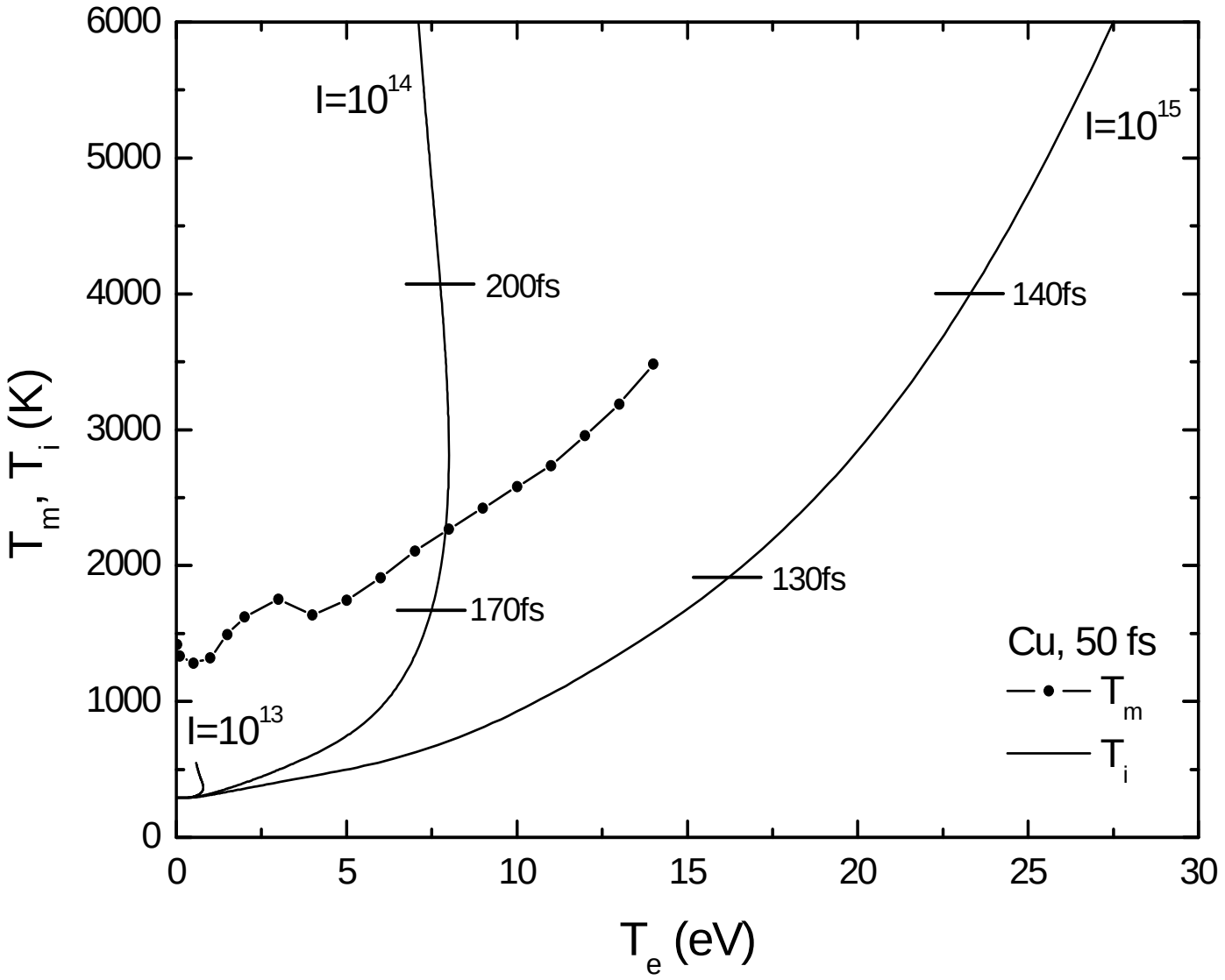
[*] S.Y. Savrasov, D. Y. Savrasov, PRB, **54**, 16487 (1996).
[†] G. V. Sin'ko and N. A. Smirnov, J. Phys.: Condens. Matter, **14**, 6989 (2002).

Evolution of the melting temperature with T_e from the FP-LMTO calculations

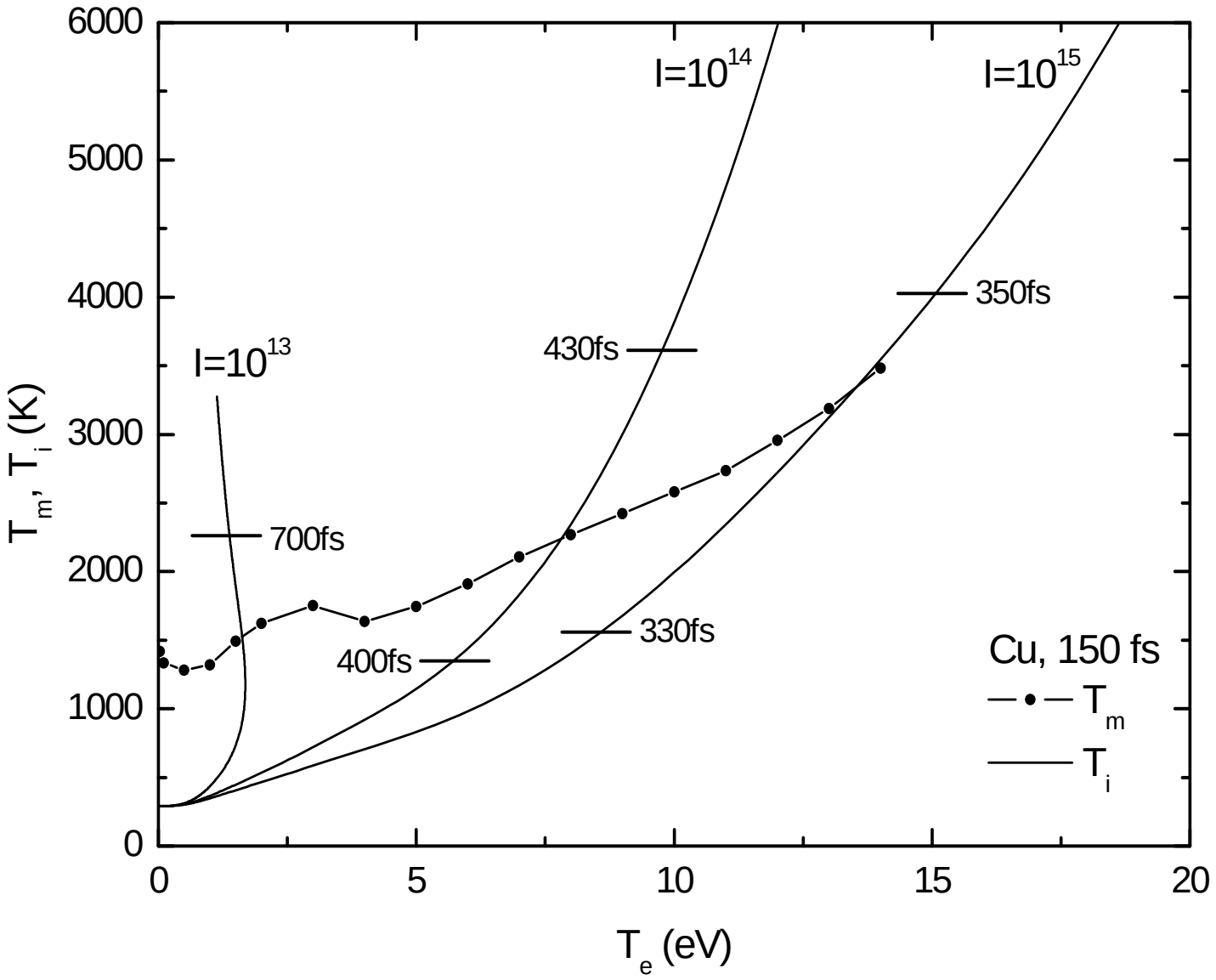


Melting curve was found from Lindemann criterion: $T_m\left(T_e\right) = CV^{2/3}\Theta_D^2\left(T_e\right)$, where the constant *C* was chosen using the experimental value of the melting temperature at zero pressure.

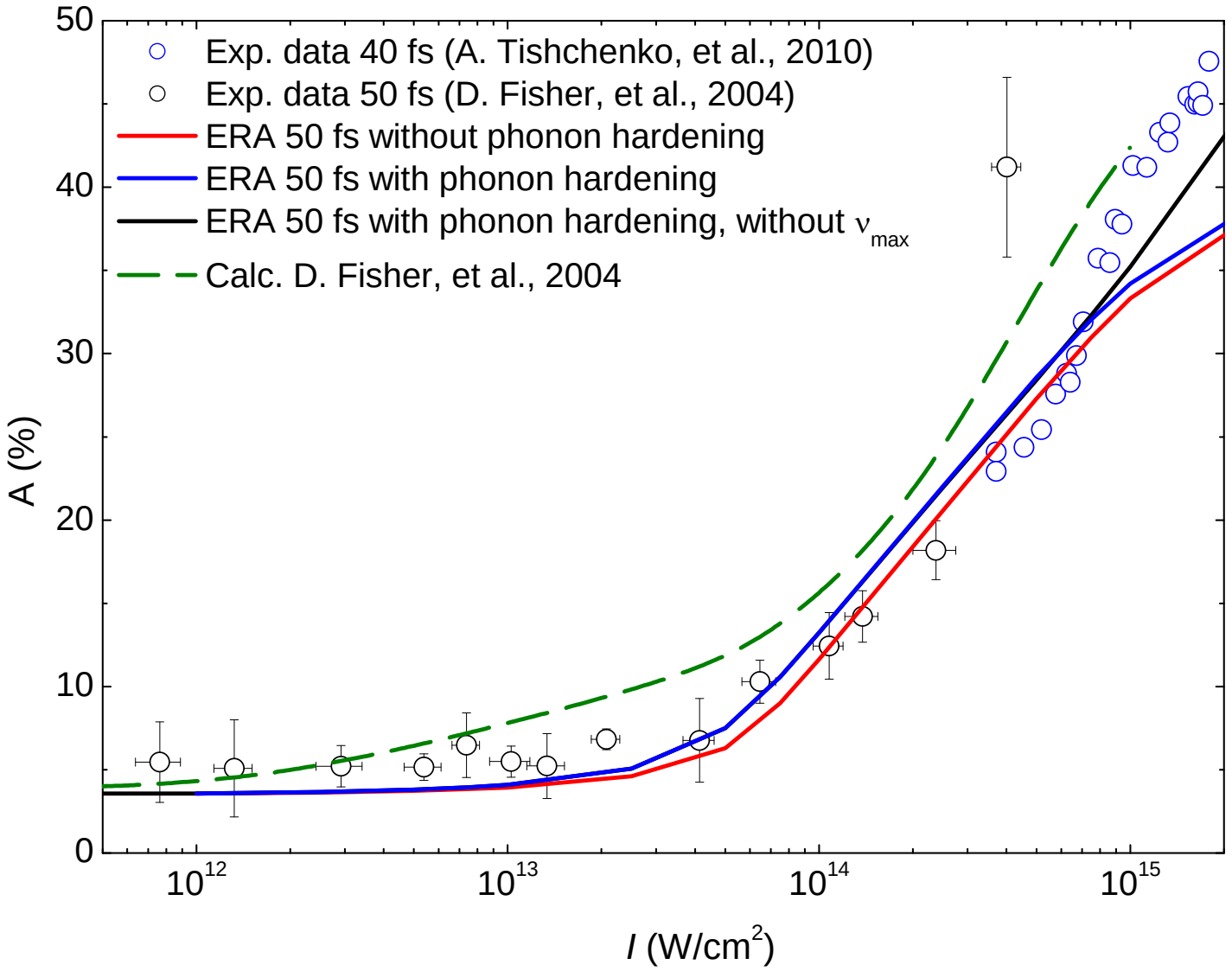
Evolution of T_i & T_m with T_e



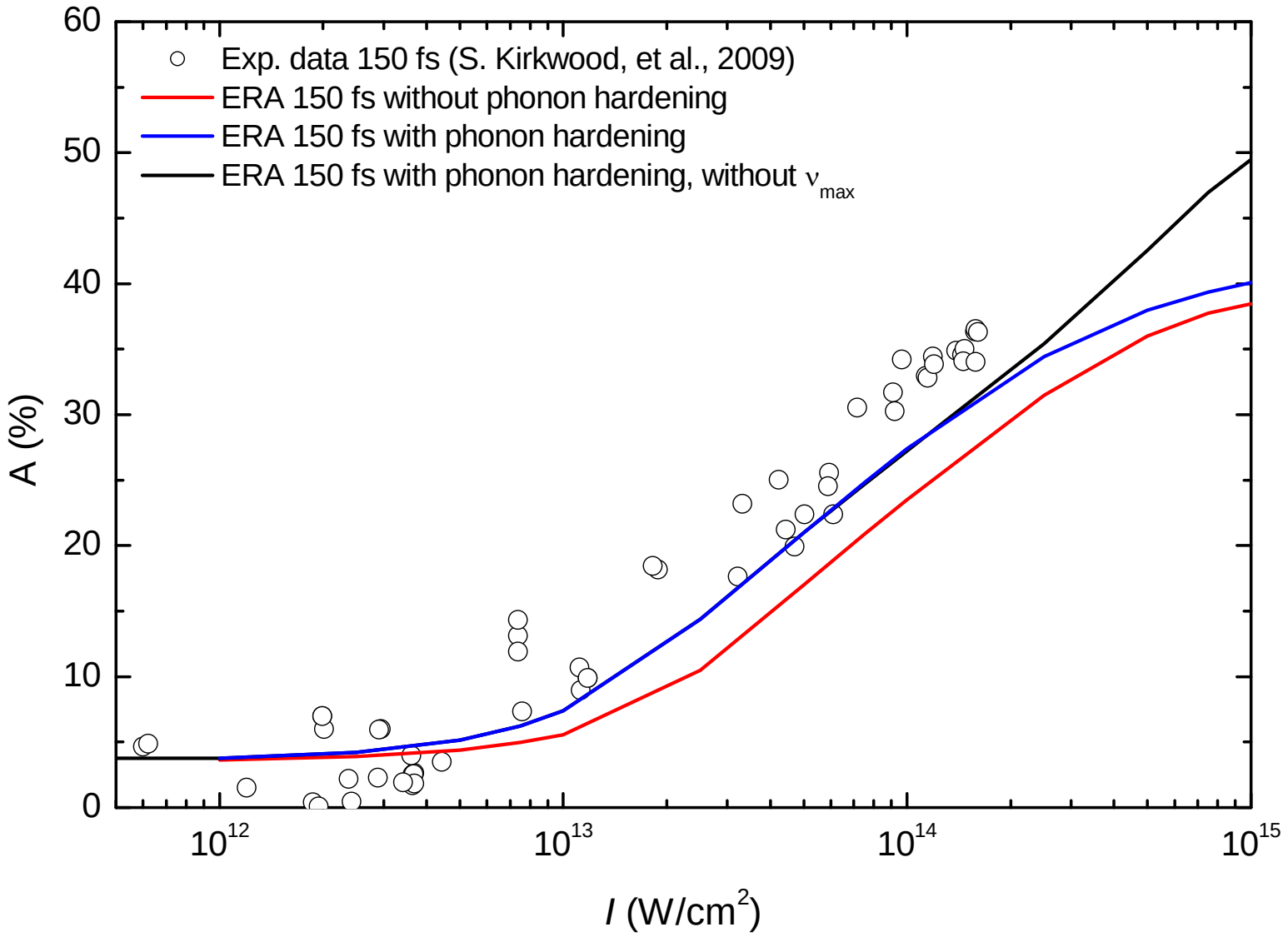
Evolution of T_i & T_m with T_e



Simulation of absorption of 50 fs laser pulses by a copper target (2nd stage)



Simulation of absorption of 150 fs laser pulses by a copper target (2nd stage)



Conclusions

- Theoretical description of the absorption of the 800 nm, 50 fs and 150 fs normal-incidence Ti:Sa laser pulses by a solid copper target at laser intensities of up to $I_L \leq 10^{15}$ W/cm^2 has been done. Numerical modeling involved thermodynamic functions calculated by using first-principles FP-LMTO method and chemical-picture-based model of dense plasma utilizing superconfiguration approach CP-SC.
- Implementation of the new theoretical model in the ERA hydrocode enabled to restrict the number of the fitting parameters to 4 (3) that are determined from the comparisons to the room-temperature reference data.
- Good agreement was obtained with the experimental absorption coefficients of fs-laser pulses by metal copper targets measured at the intensities $I_L \sim 10^{12}$ -- 10^{15} W/cm^2.
- For the 50 fs laser pulses the electron-ion energy exchange can be neglected with a loss of accuracy of less than 10 -- 15%, which is better than typical experimental errors. For the 150 fs laser pulses disregarding the electron-ion energy exchange leads to an underestimate of the laser absorption up to ~ 25 -- 50%.
- Dependences of the melting temperature and electron-ion coupling factor on the electron temperature have been obtained. Copper-target layer absorbing fs-laser light remains in the metallic state more than the halftime of the total laser-pulse action even at τ_L = 150 fs and $I_L = 10^{15}$ W/cm^2.
- Phonon-mode hardening with growing electron temperature displays a pronounced effect on the absorption driving the 150-fs modeling into better agreement with the experiment.