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

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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 [*]:

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

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

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

\[
Q_L(z, t) = \omega_L \text{Im} [\varepsilon(z, \omega_L, t)] \frac{|E(z, t)|^2}{8\pi}
\]

Helmholtz equation for the electric field amplitude

Maxwell eqs. for the normally incident fs-laser EMW

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

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

$$\varepsilon = 1 + 4\pi \sum Z n_Z \alpha_Z + i \frac{4\pi}{\omega_L} \sigma.$$ [*]

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

[†] M.V.Veysman et al. JPB, 41, 125704 (2008)
Momentum relaxation rate of conduction electrons

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

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

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

\[ \nu_{e,ph} \approx k_{e,ph} \frac{e^2 k_B T_i}{\hbar^2 \nu_F} \]

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

\[ \nu_{e,e} \approx \sqrt[2]{\frac{2 E_F}{a_1^2 m_e} \left( \frac{k_B T_e}{E_F} \right)^2} \]

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

3. \( \nu_{\text{max}} = \nu_e/r_{0}^{(i)}, \quad \nu_e = \sqrt{\nu_F^2 + 3 k_B T_e / m_e} \quad [*, \dagger] \)

[*] M.V. Veysman et al. JPB, 41, 125704 (2008)
Laser light absorption

\[ A = \frac{1}{F_L} \int_0^{t_{\text{max}}} dt \int_0^\infty Q_L(z, t) \, dz \quad \text{for} \quad t_{\text{max}} = 2 \tau_L \quad \text{for a sine-squared pulse} \quad \text{[*]} \]
\[ t_{\text{max}} \geq 4 \tau_L \quad \text{for a Gaussian pulse} \quad \text{[†]} \]

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 \([†, ‡]\):

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

Fitting parameters \( \alpha \) & \( \beta \) \leftrightarrows \text{comparison to experimental reference data} @ \, T_0 = 300 \text{ K}; \] for Cu \( \alpha = 377 \text{ W/(m K)}, \beta = 0.139 \text{ [†]} \)

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

[*] D. Fisher et al. PRE 65, 016409 (2001)
[★] A.T. Sapožnikov, 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:

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

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

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

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

$g = 2$

[†] P.A. Loboda, V.V. Popova, A.A. Shadrin. CPP, 49, 738 (2009).
Electron-temperature-dependent properties

FP-LMTO: \( T_e = T_0 \) \( E_F = 9.25 \text{ eV} \), \( <Z> = 1.51 \)
(cf. \( E_F = 6.9 \text{ eV} \) @ \( <Z> = 1 \) from general estimates) — effect of 3d-electrons on \( \mu_e \).

FP-LMTO: \( T_e \uparrow \Rightarrow \) drastic change of the electron DOS \( g(\varepsilon) \)
Cu valence-electron DOS structure — very similar to that of Au.

Au: excitation of $5d$-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/cm² 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

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: $C_e = \eta T_e$ ($\eta = 96.8 \text{ J/(m}^3\text{ K}^2)$)
Electron-temperature-dependent properties

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

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

\( \lambda_{ph} \) — electron-phonon coupling parameter,

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

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

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

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

Electron-temperature-dependent properties

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

Electron-temperature-dependent properties

Plasma electron frequency:

**FP-LMTO:**

\[
\omega_p^2(T_e) = \frac{8\pi e^2 \hbar^2}{3\Omega_c} \hat{g}(E_F) \left\langle \nu^2(T_e) \right\rangle
\]

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

\(\left\langle \nu^2(T_e) \right\rangle\) — mean-square band velocity: averaging

\[
v_{k\lambda}^2 = \hbar^{-2} \left[ \frac{\partial E_{\lambda}(k)}{\partial k} \right]^2
\]

over all the valence bands \(\lambda\) and wave vectors \(k\),

\((E(k) — T_e\text{-dependent band energy of the crystal})\).

**CP-SC:**

\[
\omega_p^2(T_e) = \frac{4\pi e^2 n_i}{m_e} \left\langle Z \right\rangle (T_e)
\]

Electron-temperature-dependent properties

Dielectric permittivity:

@ $\lambda_L = 800$ 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: $\delta\varepsilon_1 \rightarrow 0$ as $T_e \uparrow$ $\rightarrow$

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

$k_{e,ph} \approx 5.3$ — 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.

<table>
<thead>
<tr>
<th>$I_L$, W/cm$^2$</th>
<th>$T_e$, eV</th>
<th>$T_i$, eV</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^{13}$</td>
<td>0.97</td>
<td>0.04</td>
</tr>
<tr>
<td>$10^{14}$</td>
<td>10.9</td>
<td>0.4</td>
</tr>
<tr>
<td>$10^{15}$</td>
<td>44.5</td>
<td>5.3</td>
</tr>
</tbody>
</table>

[*] P.A. Loboda, N.A. Smirnov, A.A. Shadrin, N.G. Karlykhanov. HEDP, 7, 361-370 (2011)
Simulation of absorption of 50 fs laser pulses by a copper target (1st stage)


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

present model:

a) $\gamma = \gamma (T_0)$

b) $\gamma = 10^{11} \text{ W cm}^{-3}\text{K}^{-1}$

c) $\gamma = 0$
Simulation of absorption of 150 fs laser pulses by a copper target (1st stage [\*])

![Graph showing absorption vs laser intensity]

- Experimental data, 150 fs (S. Kirkwood, et al., 2009)
- Present model:
  - $\gamma = \gamma (T_e)$
  - $\gamma = 10^{11} \text{ W} / (\text{cm}^3 \text{K})$
  - $\gamma = 0$

[*] P.A. Loboda, N.A. Smirnov, A.A. Shadrin, N.G. Karlykhanov. HEDP, 7, 361-370 (2011)
Modification of the electron-ion coupling factor due to phonon hardening (2nd stage)

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

$\lambda_{ph}$ — electron-phonon coupling parameter was taken from ab initio calculations [*]:

$\lambda_{ph} = 0.14$;

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

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

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

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

![Graph showing the evolution of $T_i$ and $T_m$ with $T_e$.](image)
Evolution of $T_i$ & $T_m$ with $T_e$

![Graph showing the evolution of $T_i$ and $T_m$ with $T_e$.](image)
Simulation of absorption of 50 fs laser pulses by a copper target (2nd stage)

- Exp. data 40 fs (A. Tishchenko, et al., 2010)
- Exp. data 50 fs (D. Fisher, et al., 2004)
- ERA 50 fs without phonon hardening
- ERA 50 fs with phonon hardening
- ERA 50 fs with phonon hardening, without $v_{\text{max}}$
- Calc. D. Fisher, et al., 2004

Graph showing absorption $A$ (%) vs. intensity $I$ ($\text{W/cm}^2$)
Simulation of absorption of 150 fs laser pulses by a copper target (2\textsuperscript{nd} stage)

![Graph showing absorption vs. intensity for different conditions: Exp. data 150 fs (S. Kirkwood, et al., 2009), ERA 150 fs without phonon hardening, ERA 150 fs with phonon hardening, ERA 150 fs with phonon hardening, without $v_{\text{max}}$.](image-url)

- Exp. data 150 fs (S. Kirkwood, et al., 2009)
- ERA 150 fs without phonon hardening
- ERA 150 fs with phonon hardening
- ERA 150 fs with phonon hardening, without $v_{\text{max}}$
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/cm² 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/cm².

- 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 $\tau_L = 150$ fs and $I_L = 10^{15}$ W/cm².

- 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.