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The effect of pulse duration on the interplay of electron heat conduction and electron–phonon interaction: Photo-mechanical versus photo-thermal damage of metal targets

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ABSTRACT

Here we present a theoretical work on short pulse laser interaction with metal targets. With the help of Two Temperature Model we consider in detail the interplay of two competitive mechanism of the laser deposited energy dissipation: the fast electron heat conduction and the electron–phonon interaction processes. For a range of pulse durations the modelling included a complex description of these processes as the functions of the electron and phonon temperatures. It was shown that for transitional type of metals the maximum surface temperature determined for a range of pulse durations at fixed fluence exhibits peak behavior and the corresponding pulse duration is close to the electron–phonon relaxation time. In contrast, having complex deviation of heat conductivity from its linear growth with the electronic temperature, the group of noble metals shows different behavior in maximum surface temperature depending on the fluence regime. Based on the results, an experimental approach in measuring the electron–phonon relaxation time is suggested and a general tendency of photo-mechanical versus photo-thermal damage of metal targets is deduced.

1. Introduction

The effect of laser pulse duration on the character of laser damage (melting, ablation, and spallation) has been considered in several experimental and theoretical papers for the last decade [1,2]. It was justified that ns laser pulses results in a relatively slow heating under electron–phonon equilibrium conditions and eventually in a significant damage of surrounding material due to the excess of thermal energy (melting) [3]. This process therefore was attributed to photo-thermal damage and opposed to the photo-mechanical effect where ps or fs laser pulses were used to generate a strong nonequilibrium between electrons and phonons. Fast, within tens of ps, heating due to hot electrons resulted in significant stresses development inside of the material. The relaxation of these stresses led to a local mechanical material damage/modification with very sharp edges [4,5]. Apparently, the regime of photo-mechanical influence should be a working mode for the purpose of nanostructuring [6,7]. The understanding of laser energy dissipation channels [8] under highly nonequilibrium conditions is therefore of a great importance for the purpose of both technology and fundamental science.

Although, it was discussed that it is the strength of the electron–phonon coupling that plays a determinant role in the character of the laser induced processes [1], its quantitative definition is still a big challenge and only several predictions based on theoretical models were made till nowadays [9,10]. Obviously, that the strength of the electron–phonon coupling is unequivocally related to the electron–phonon relaxation time. The complex and transient character of the electronic and lattice temperature behavior, however, during the short laser pulses make the experimental determination of this time difficult. Additionally, the fast electron heat conduction mechanism is a strong function of the electronic temperature T_e and can drastically alter the energy dissipation during the electron–phonon equilibration process. In several theoretical attempts to study the electron heat conduction mechanism [10–12], it was emphasized that depending on the level of electron excitation (temperature), the conductivity can be approximated either as a constant ($T_{ph} \sim T_e \ll T_F$), or a linear function of T_e ($T_{ph} < T_e \ll T_F$), or a function attributed with the electron–electron collision process ($T_{ph} \ll T_e < T_F$), or a function that behave like plasma conductivity $\sim T_e^{5/2}$ ($T_{ph} \ll T_e \sim T_F$).

In this paper we perform a theoretical investigation of the response of metal targets to a short pulse laser irradiation. For this purpose we utilize Two Temperature Model [4,13] and carry out the calculations on the example of several metals: Al, Ni, Cu, and Au. Particularly, we will consider the interplay of two competitive

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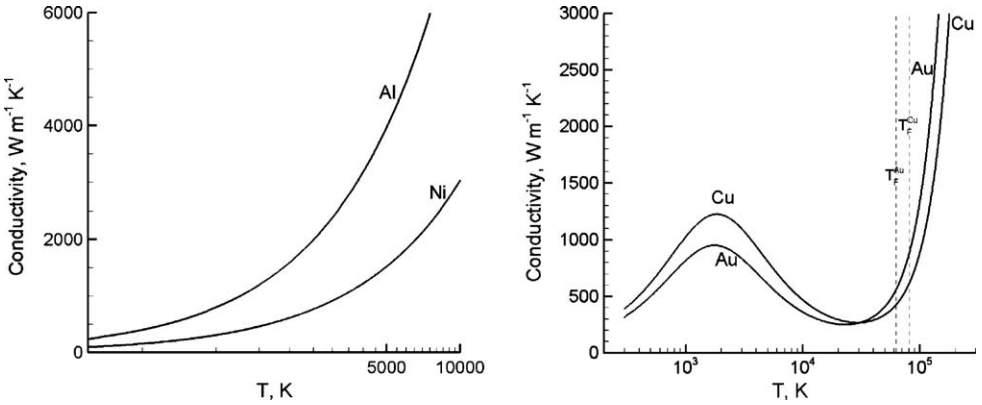

Fig. 1. The electron heat conductivities for: (a) Ni (T_F = 136,000 K) and Al (T_F = 135,000 K); (b) Au (T_F = 64,000 K) and Cu (T_F = 81,000 K).

processes of dissipation of the laser deposited energy: fast electron heat conduction and the electron–phonon relaxation processes.

2. The model

In all calculations we used the well-known Two Temperature Model (TTM) [13], in which two coupled differential equations describe the temperature dynamics of two interacting subsystems: the electrons and phonons. Among the four metals chosen to test the process of energy dissipation of two of them, Au and Cu, exhibit pure properties of noble type of metals and the rest, Ni and Al, belong to the group of transitional metals. Noble metals have a relatively weak strength of the electron–phonon coupling that results in quite high values of the electronic temperatures upon laser light absorption. Transitional metals, on the other hand, exhibit much stronger electron–phonon coupling and much lower electronic temperatures are reached during the laser–matter interaction process.

The heat conduction mechanism in this work was considered in 1D that corresponds to the case of wide laser spot (~10 μm) on a metal surface as compared to the laser light absorption depth (~10 nm). The corresponding form of TTM is shown below, Eq. (1):

$$\begin{cases} C_e(T_e)\dfrac{\partial T_e(z,t)}{\partial t} = \dfrac{\partial}{\partial z}k_e(T_e,T_{ph})\dfrac{\partial}{\partial z}T_e(z,t) - G(T_e,T_{ph})(T_e - T_{ph}) \\ \quad + S(z,t) \\ C_{ph}(T_{ph})\dfrac{\partial T_{ph}(z,t)}{\partial t} = G(T_e,T_{ph})(T_e - T_{ph}) \end{cases} \quad (1)$$

where heat capacities C_{ph} and C_e were taken as functions of phonon T_{ph} and electron T_e temperatures correspondingly. The dependence of C_{ph} on T_{ph} was based on experimental measurements [14]. The dependence of the electron heat capacity C_e and the strength of the electron–phonon interaction G on T_e was accounted for with different theoretical predictions and qualitatively did not affect the results of our calculations: either we used $G = const$ and $C_e = \gamma T_e$ [15] or more complex dependence was considered when both $G = f(T_e)$ and $C_e = f(T_e)$ [16]. The electron heat conduction k_e exhibits a strong dependence with temperatures of electrons and phonons and was described with two approximations, depending on the material and, as a result, the temperature of electrons developed upon the absorption of the laser pulse. Namely, if the excitation of electrons leads to the temperature comparable with Fermi temperature T_F, like it is the case of Au and Cu ($T_e \sim 10^4$ K), the dependence of k_e as suggested in Ref. [11] is given by Eq. (2):

$$k_e = C\frac{\left(\vartheta_e^2 + 0.16\right)^{5/4}(\vartheta_e + 0.44)\vartheta_e}{\left(\vartheta_e^2 + 0.092\right)^{1/2}\left(\vartheta_e^2 + b\vartheta_{ph}\right)} \quad (2)$$

where $\theta_e = T_e/T_F$ and $\theta_{ph} = T_{ph}/T_F$ and C, b are material constants, listed in Refs. [11,17]. If, however, the excitation is relatively low and the maximum electronic temperature remains negligible compare with Fermi temperature T_F, like it was in the case of Ni and Al ($T_e \sim 10^3$ K), an ordinary linear dependence of k_e with electron temperature T_e can be used [12], Eq. (3):

$$k = k_0\frac{T_e}{T_{ph}} \quad (3)$$

where k_0 is a material constant. The heat conductivities for the materials used in the present calculations for T_{ph} are shown in Fig. 1a and b.

The fluences used in all calculations were chosen to be relatively low, below the threshold for material modification (melting), and were hold constant for a particular set of calculations where only pulse duration was varying. The source term $S(z,t)$ was defined similar to that in Ref. [1] as well as the method for accounting the ballistic transport of nonequilibrium electrons in Au by adding the ballistic range of 105 nm to the optical absorption depth. The latter results in a long range temperature gradient establishing in Au upon laser heating as compared with other materials. Since working at the relatively low fluences, however, the dependence of the ballistic range value on T_e was not accounted for.

3. Numerical results and discussions

3.1. Transitional metals Ni and Al

The system of TTM equations was solved numerically by the finite-differences method with the initial and boundary conditions as they are for free standing films. The film thickness L was chosen so that by the end of modeling the rise of electron temperature on the back surface would not exceed 1% of its initial value. Thus, the calculations were carried out on what is usually understood by thick or bulk metal targets (with L values 300, 400, 500, and 2000 nm for Ni, Al, Cu, and Au materials, respectively). The time step was chosen based on Neumann stability criterion $\Delta t \leq 0.5(\Delta X^2)C_e(T_e)/K_e(T_e,T_{ph})$ and ranged from 0.01 fs for Au to 1 fs for Ni.

For each particular metal we perform a set of calculations at fixed absorbed fluence for a range of pulse durations from 100 fs to