Thermal and non-thermal explosion in metals ablation by femtosecond laser pulse: classical approach of the Two Temperature Model

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Abstract. We propose a classical Two Temperature Model TTMc where we consider the metal film during the irradiation like an ideal plasma. The numerical results are comparing to those finding by the existing TTM and the experimental data. In our model The cooper is taken as a target irradiated by a single laser pulse with 120 fs at 800 nm wavelength in air room. Our numerical results shown that there are a thermal and non-thermal explosion successively occurs in metal ablation by ultrashort laser pulse.

1. Introduction

Metal ablation by ultrashort laser pulse has taken a spacial attention siense 1990s. On the one hand, because the new regim of lasers rather femtosecond lasers opened a new field of industriel-thechnology with a vaste applications developped owing to the high quality of ablation hole in contrast to those obtained by conventional (nanosecond) regim. In the other hand, this kind of laser pulse have opened a new posibilities of fundamental investigations in physics.

it has been shown that the quality of ablated holes induced by femtosecond laser pulse is much better than those produced by nanosecond or even more large pulses [1, 6], where the main charactiristic of femtosecond laser ablation of metals are: very rapid creation of plasma phase and no heat effect zone around the craters at low fluence [1, 2], and a thin layer of liquid appear at high fluence [3]. Even if all authors are in agreement with the fact that the matter ejected in liquid form is necessarily due to phase explosion whether by nanosecond pulse [1, 4] or at high fluence by femtosecond pulse [3, 5], we must investigate the physical mecanism responsible of the metal ablation by femtosecond (fs) laser pulse at low and high fluence.

Many works have been published in this field to answer this quenstion where it was pro-posed that the ablation mecanism is always due to phase explosion e.i. thermal explosion basing on the existing quantum Two Temperature Model (TTMq) [5, 7, 8]. But the problem is that this model does not give any information on the ablation state at low fluence (aroud ablation thershold), also the phase explosion model does not explane the absense of liquid around the
crater at low fluence. In this effect, Gamaly et al. [9, 10] proposed a new model called Coulomb Explosion (CE) where they explained that the metal ablation mechanism at fs laser pulse consists in the ion acceleration in the electrostatic field created by hot electrons escaping from the target.

CE is very interesting, since it explains clearly the ablation mechanism of solids by ultra-short lasers, without thermal physics. Gamaly has succeeded in giving explanation for the lack of liquid phase at the surface during ablation. However, the model presents an inconvenient concerning the nature of the solid material. Since 2002, Stoian et al. [11] demonstrated that this kind of explosion is dominant only in the case of dielectrics. In 2006, Dachraoui’s et al. [12] calculations shown that this kind of explosion exists by evidence in all solids but at different rates: it is very important in dielectrics, moderately weak in semi-conductors, and very weak in metals because of the screening effect.

In this paper we try to contribute to resolve this problem by giving an answer to the rising question: what is the physical mechanism responsible of the ablation by ultra-short laser pulses? we propose a new classical Two Temperature Model (TTMc) which agree with the experimental data at low and high fluence.

2. Theoretical Model

2.1. quantum Two Temperature Model TTMq

Considering that a copper target is normally irradiated by a femtosecond laser pulse, the evolution of electron and lattice temperatures can be described by the famous one-dimensional Two Temperature Model called quantum Two Temperature Model (TTMq) [7, 13]:

\[ C_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial z}(k_e \frac{\partial T_e}{\partial z}) - G_q(T_e - T_l) + S \tag{1} \]

\[ C_l \frac{\partial T_l}{\partial t} = G_q(T_e - T_l) + S \tag{2} \]

where the subscript e and l denote, respectively, electron and lattice. C is the heat capacity, k is the thermal conductivity, \( G_q \) is the electron-phonon coupling factor, S is laser heat density and t is the time. The laser beam is propagated along the z-axis. The heat density S can be expressed as [7]:

\[ S = 0.94 \frac{1 - R}{t_p(\delta + \delta_b)(1 - e^{-d/(\delta + \delta_b)})} F_{exp} \left[ - \frac{z}{(\delta + \delta_b)} - 2.77 \left( \frac{t}{t_p} \right)^2 \right] \tag{3} \]

\( t_p \) is the FWHM (full width at half maximum) pulse width, \( \delta = 1/\alpha \) the absorption depth, \( R \) the reflectivity, \( d \) the thickness of the sample, and \( F \) the fluence. \( \delta_b \) is the ballistic range. For the parameter values of \( C_e = \gamma T_e \) where \( \gamma = 97 \ J/(m^3K^2) \) [14, 15] is the electron heat capacity constant and \( C_l = 3.46 \times 10^6 J/(m^3K) \) [15].

\( \alpha \) is the absorption coefficient and \( R \) is normal-incidence reflectivity. \( k_e \) is the electron thermal conductivity value, it was calculated using the general solution provided by Animisov et al. [13]. \( G_q \) is the electron-phonon coupling factor related to the rate of the energy exchange between the electrons and the lattice, which can be expressed as [5]:

\[ G_q = G_0 \left[ \frac{A_e}{B_l} (T_e + T_l) + 1 \right] \tag{4} \]
we adapt the existing TTM Eq.1 and Eq. 2 to our assumption where we propose a new target during the irradiation like an ideal plasma (electrons + ions).

2.2. classical Two Temperature Model TTMc

We propose a new classical Two Temperature Model (TTMc) where we consider the cooper first ionization potential, Debye temperature and mass molar for copper respectively [22].

\[ \lambda_{collisions} \]

where \( \lambda \) is the Debye length.

The amount \( f_{e\text{ph}}(T_e) = (\frac{m_e^*}{M})^{1/2} \frac{J_i}{T} \frac{1}{T_D} \) is called the collision frequency for momentum exchange which can be expressed as [10]:

\[ f_{e\text{ph}}(T_e) = (\frac{m_e^*}{M})^{1/2} \frac{J_i}{T} \frac{1}{T_D} \]  \hspace{1cm} (5)

where \( m_e^* \) is the electron effective mass [21], \( J_i = 7.72eV, T_D = 343K \) and \( M = 63.54 \) a.m.u are first ionization potential, Debye temperature and mass molar for copper respectively [22].

\[ C_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial z}(ke \frac{\partial T_e}{\partial z}) - G_e(T_e - T_i) + S \]  \hspace{1cm} (6)

\[ C_i \frac{\partial T_i}{\partial t} = G_e(T_e - T_i) + S \]  \hspace{1cm} (7)

\( G_e \) is the electron-ion coupling factor related to the rate of the energy exchange between the electrons and ions.

We define an average relaxation time \( \tau_e(T_e) \) for the exchange of momentum between electrons and ions such as \( G = \frac{\sigma_1}{\tau_e(T_e)} \) [17]. The amount \( f_{e\text{el}}(T_e) = 1/\tau_e(T_e) \) is the collision frequency for momentum exchange which can be expressed as: \( f_{e\text{el}}(T_e) = n_e \sigma_1 v_{e}^* \), \( \sigma_1 \) is called the cross-section for momentum transfer between the electron and ions once ionized, given by the relation \( \sigma_1 = 4\pi p_0^2 \ln \Lambda \) where \( p_0 = e^2/(4\pi\varepsilon_0 m_e^* v_e^* ) \) [18] is called the critical impact parameter; \( p_0 \) is the distance at which the two particles should be placed so that the potential energy is equal to twice the initial kinetic energy. \( v_e^* = \sqrt{3k_B T_e}/(m_e^*) \) is the average quadratic velocity of electrons.

The amount \( \ln \Lambda = \ln(\lambda_D/p_0) \) [6] is called the Coulomb logarithm; it is related to the plasma parameter, this factor measures the relative importance of distant collisions compared to near collisions, \( \lambda_D \) is the Debye length.

Initial and boundary conditions are expressed as:

\[ \frac{\partial T_e}{\partial z} \big|_{z=0} = \frac{\partial T_e}{\partial z} \big|_{z=d} = \frac{\partial T_{l,i}}{\partial z} \big|_{z=0} = \frac{\partial T_{l,i}}{\partial z} \big|_{z=d} \]  \hspace{1cm} (8)

3. Results and discussion

Our model was simulated using MATLAB software, the results were compared to those obtained by the TTMq model and with the experimental data.

Figure 1(a) represent the evolution of electron and lattice temperatures after a femtosecond laser irradiation, modelling by TTMq model. We see that the lattice reach 90% \( T_C \) (\( T_C = 7696K \) is the critical temperature of phase explosion (thermal explosion)) at (3 J/cm²) after vibrational (equilibration) relaxation time. Because the emitted phonons carry little energy, the electron-phonon collision takes many scattering processes, and therefore several picoseconds, before the carriers and the lattice reach thermal equilibrium [19]. This fluence \( (F = 3) \) called the strong ablation threshold where the matter start to be ejected in liquid form during the ablation. It is
Figure 1. (a) the evolution of electron and lattice temperatures, (b) lattice temperature versus time and position in a copper sample for an absorbed fluence just above the thermal ablation threshold (3 J/cm²)

similarly equal to Mannion’s et al. experimental results [20] where they find $F = 3.19$ J/cm². However, as mentioned above, it has been observed experimentally [2, 5, 6, 20] a very clear ablation at low fluence about of $F = 0.5$ J/cm², where there is no trace of liquid on the surface, therefore, it has been suggested that the matter has been ejected in plasma form.

In Figure 1(b) we represent the lattice temperature versus the time and the position in a copper sample for an absorbed fluence just above the thermal ablation threshold (3 J/cm²). It is the contour of the thermal penetration of lattice temperature during electron-phonon collision time.

Figure 2(a) represent the evolution of electrons and ions temperature after the irradiation by femtosecond laser, obtained by TTMc model proposed.

Figure 2. (a) the evolution of electrons and ions temperature, (b) electrons temperature versus time and position in a copper sample for an absorbed fluence just above gentle ablation threshold (0.53 J/cm²)
When the metal surface irradiated by a femtosecond laser, the electrons will absorb the laser energy and then transfer it to set of ions, but we remark that at \((0.53 \text{ J/cm}^2)\), the electrons reach a classical critical temperature \(T_{CC}\) greater than or equal to \(E_b\) where \(E_b = 3.125 \text{ eV/atom}\) is the binding energy \([10]\). So, \(T_{CC} = \frac{E_b}{k_b} = 3.62 \times 10^4 K\). therefore, we suggest that ions absorb electrons energy in momentum exchange at sub-vibrational time via electron-ion collision and stay thermally cold (Figure 2(a)), so there will be a random motion of ions which can be considered as a non-thermal liquid (plasma) that will escape or leave the material, so it may be called a non-thermal explosion. This critical fluence is in good agreement with the ablation threshold (also called gentle ablation) in various experimental results \([5, 6, 20]\).

In Figure 2(b) we represent the electron temperature versus the time and the position in a copper sample for an absorbed fluence just above the non-thermal ablation threshold \((0.53 \text{ J/cm}^2)\). It represents the contour of penetration of electron temperature during electron-ions collision time.

Figure 3 represents a logarithmic evolution of depth in function of fluence. We observe that our theoretical results obtained by TTMc model are in good agreement with the experimental data.

But since the thermal explosion is obviously present at high fluence, we suggest that at low fluence all the ablation inside the laser focal volume is due to non-thermal explosion, but with increasing fluence the ablation in the side of the spot laser have to be caused by thermal explosion, because electron-phonon collision frequency increase in contrast the electron-ions collision frequency decrease (Fig. 4).

Finally, we can say that at low fluence all the crater volume due to the non-thermal explosion (N-TE) and the mater is ejected in a plasma form, and at high fluence, deeper region of crater due to N-TE and the inter side region due to thermal explosion (TE) and the mater successively ejected in plasma form and in liquid form.
4. Conclusion
In this paper, we highlight a recent problem in the modern physics, that one concerning the physical mechanism of the metal ablation by femtosecond laser pulse, where it has been shown that there is a large difference between the quality of the crater produced by short or ultrashort laser pulse. Therefore, we can say that there are a large difference also in the physical mechanism corresponding.

It experimentally observed that no trace of liquid around the crater during the irradiation by fs pulse at low fluence and the presence of a thin liquid layer at high fluence. The TTMq model show that at high fluence there is a thermal explosion (TE), so the matter is ejected in a liquid form, but at low fluence no information is given. Our proposed model TTMc show that there is a non-thermal explosion (N-TE) at low fluence due to the creation of a plasma phase at sub-vibrational time, during electron-ion collision when the material keep thermally cold. At high fluence there is always a non-thermal explosion (N-TE) but exist also a phase explosion (TE) because of the different rate in collision frequency value.

As a conclusion, basing to our model TTMc, metal ablation at low fluence is necessary due to a non-thermal explosion which produce a very clear ablation. In contrast, at high fluence, both non-thermal and thermal explosion successively occurs.

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