Electromagnetic particle-in-cell (PIC) method for modeling the formation of metal surface structures induced by femtosecond laser radiation

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**A R T I C L E   I N F O**

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**A B S T R A C T**

The particle in cell (PIC) method coupled to the finite-difference time-domain (FDTD) method is used to model the formation of laser induced periodic surface structures (LIPSS) at the early stage of femtosecond laser irradiation of smooth metal surface. The theoretical results were analyzed and compared with experimental data taken from the literature. It was shown that the optical properties of the target are not homogeneous and the ejection of electrons is such that ripples in the electron density were obtained. The Coulomb explosion mechanism was proposed to explain the ripples formation under the considered conditions.

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**1. Introduction**

Periodic nanostructures in form of ripples on semiconductors irradiated by a ruby laser have been observed since 1965 [1]. And, it has been shown that remarkable properties of these nanostructures can appear on semiconductors, metals or dielectrics. They are generally perpendicular to the laser polarization with spatial period slightly above the laser wavelength. They appear at fluence close the ablation threshold and for any pulse width. Although the original structure is always ripples, various morphologies such as beads and spikes can be observed. All these structures are commonly called laser induced periodic surface structures (LIPSS). In last decade, some scientists are interested on the LIPSS formation mechanisms from fundamental point of view for their optimal applications in nanotechnologies as in photovoltaic and catalyze [2]. In this context, main phenomena invoked to explain the LIPSS formation are the diffraction of the focusing lens [1], interference of the incident waves with surface ones diffused by the microscopic roughness of irradiated surface [3–6] or by the laser-induced plasma or surface plasmons and polaritons [7–9]. The commonly considered mechanism is that of the optical interference suggested for the first time by Emmony et al. in 1973 [10] and then formalized by Sipe et al. in 1982 [11]. Thus, the electromagnetic wave transmitted parallel to the surface by the roughness interferes with the incident wave to form a pattern of interference fringes which are perpendicular to the polarization direction. This theory is quite satisfactory in the case of ripples produced by pulses of nanosecond or more. Experiments with femtosecond (fs) lasers have showed that the spatial period could be shorter than the wavelength. Thus, two types of ripples can appear called low spatial frequency (LSF) and high spatial frequency LIPSS (HSFL). Recently, Bonse et al. [5,6] have improved this theory by including nonlinear effects and proposing the second harmonic generation process to explain the formation of HSFL. The complex refractive index has been corrected by adding contributions of multiphotonic ionization and the Kerr effect [5,6,12]. On the other hand, Sakabe et al. [13,14] have proposed a new explanation based on the Coulomb explosion of the ionic cloud due to the surface wave plasma induced by the process of stimulated Raman scattering. In this inelastic process, a photon is annihilated and gives rise to a surface plasma and a photon with lower frequency. The dispersion relation of surface plasmons showed that wavelength of the induced ripples varies between 0.85λ and 0.5λ (λ is the laser wavelength). Experimentally, these results are in agreement with the measurements carried out only for fluences slightly above the ablation threshold. Below this fluence, the ripples with spacing smaller than 0.5λ have been observed. From the theoretical point of view and in fs
regime, it seems difficult to describe the physics of the interaction due to the incomplete knowledge of thermodynamic properties of the material. Then, the approach usually used is that based on the hydrodynamic two-fluid model, even though in this configuration the non-equilibrium state of the system is not considered. In this context of non-equilibrium, we report in this paper a new approach based on the particle in cell method (PIC), which presents the advantage that it recovers the Vlasov transport equation and provide more convenience in the numerical treatment. One application has been already described in the case of an electrostatic model [15], and in this work, we present an improved model that couples the PIC part and the electromagnetic part. To do so, we have organized this paper as follows: in Section 2, we introduce the theory of the modeling. The simulation results concerning the electron density profile and morphologies of nanostructure formation under fs laser pulse are presented in Section 3. We focus on the spatial period of ripples as a function of laser intensity and comparisons between our results and those of experiments found in the literature. Finally, Section 4 concludes this work and gives perspectives inspired from our results.

2. Modeling

We consider that the free electrons referred to as conduction electrons in the metal target move in the background of immobile positive metal ions. This can be justified by the fact that in the interaction of a femtosecond laser with a solid, electrons absorb the major part of the electromagnetic energy by inverse bremsstrahlung process. It is only after a relaxation time of the order of a few picoseconds, that the electronic gas transfers part of its energy to the lattice. This hypothesis, which is valid only at the beginning of the interaction, suggests using the PIC method well known in the plasma physics. This method is based on the concept of a pseudo-particle i.e. the evolution of a system of a large number of particles is equivalent to the evolution of a smaller number of pseudo-particles, each of these is a set of neighboring particles in the phase space of the system. In this context, the relativistic electron motion equations are as follows

\[
\frac{du}{dt} = -\frac{e}{m_e} \vec{E}
\]

\[
\frac{d\vec{r}}{dt} = \gamma^{-1} \vec{u}
\]

\[
y = \sqrt{1 + \frac{u^2}{c^2}}
\]

where \(-e\) and \(m_e\) denote the electrical charge and the rest mass of the electron, respectively; \(u\) is the electron velocity and \(c\) represents the light celerity in the vacuum and \(\vec{E}\) denotes the electric field.

To solve Eqs. (1), we need to known the electrical field which is calculated from the resolution of Maxwell equations. The current density is then deduced from the Ohm law \(j = \sigma \vec{E}\) where the electrical conductivity \(\sigma\) is given by the Drude model [16–18] as:

\[
\sigma = \frac{\hbar \omega_{pe}^2}{2 \pi c^2 (\omega^2 + \omega_{pe}^2)}.
\]

\(\nu\) denotes the collision frequency of free electrons, \(\omega_{pe}\) and \(\omega\) are the electronic plasma and the laser frequencies, respectively. Let us note that \(\nu\) and \(\omega_{pe}\) depend on the electron density \(n_e\) and \(\nu\) are defined as the geometrical average given by [16]:

\[
\nu^{-1} = \nu_{e-e}^{-1} + \nu_{e-ph}^{-1}
\]

\(\nu_{e-e}\) and \(\nu_{e-ph}\) are electron–electron and electron–phonon collisions frequencies, respectively expressed by

\[
\nu_{e-e} = \frac{1}{3(2\pi)^{3/2}} \frac{Z_{av} e^4 n_e n_e}{\sigma_0^2 (m_e k_B T_e)^{3/2}} \ln \Lambda
\]

\[
\nu_{e-ph} = 2k_e \alpha_f c^2 \frac{\hbar^2}{m_e}
\]

where \(Z_{av}\) is the ionization rate, \(\Lambda\) the Coulomb logarithm, \(k_e\) is a numerical constant of about 10, \(\alpha_f = 1/137\) and \(v_F\) is the Fermi velocity.

To solve the above equations, we used the “leap-frog” algorithm already described in [19,20], associated with the finite difference time domain (FDTD) algorithm for the electromagnetic part [21]. According to this, the 2D simulation domain is divided into a grid of \(1739 \times 187\) square cells. At the boundaries, we applied perfectly matched layer (PML) conditions of absorption. The spatial step (\(\Delta x = 11.5\) nm) and the temporal step (\(\Delta t = 0.019\) fs) are determined by the stability criterion of FDTD method formulated by \(\Delta t = \Delta x^2 / 2c\) combined with the stability criterion of the leapfrog discretization \(\omega_{pe} \Delta t < 2\). The spatial interpolation of the electric field and the density calculation are realized according to the Cloud-In-Cell algorithm [19].

3. Simulations and results

In this section we present simulation results showing the electron density profile and spatial ripple period versus laser wavelength and intensity. We consider a copper target \((n_{\text{Cu}} = 6 \times 10^{28} \text{ m}^{-3})\) initially at room temperature \(300\) K with large thickness \((1\) μm) as compared to the depth of the region heated by the shock wave. The pseudo-particles and particles per pseudo-particle numbers are taken \(1.5 \times 10^5\) and 50, respectively. The choice of these computational parameters is imposed by the need to obtain a good compromise between the pertinence of the model, available space memory and computing time. Simulations are carried out using 70fs Gaussian pulse (full width at half maximum) centered at 100fs with a simulation time of 230fs corresponding to the total duration of a single pulse. Fig. 1 illustrates the geometry of the computational domain and the spatial profile of the laser intensity applied on this surface target considered to be perfectly flat. Fig. 2a shows temporal development of the spatial electron density distribution (along y). We observe a shock wave represented by an abrupt density peak which propagates toward the
inside of the target. The same behavior concerning the evolution of the mass density in the 1D hydrodynamic model of the ablation has been reported [16]. This figure also shows a portion of the target devoid of its cloud of conduction electrons. Fig. 2b clearly indicates the domain that carries a positive electrical charge of about \( e \eta_{el} = 9.6 \times 10^{-8} \, \text{C.m}^{-2} \). The electrostatic repulsion of the positive ions lattice increases strongly. We can assume that this repulsion is able to break the atomic bands and then causes strong ion ejections.

Fig. 3a and b represents electron density profiles obtained at the end of the simulations (230 fs) with wavelengths 0.6 and 0.8 \( \mu \text{m} \), respectively. Structures in the form of waves appear in the craters for intensities greater than \( 1 \times 10^{19} \, \text{W.m}^{-2} \). They are identified with the nano-ripples observed experimentally. Thus, our results confirm that during the pulse duration, the electron density is spatially variable in contrast to studies based on interference and stimulated Raman scattering. In most simulations, the analysis of the Fourier spectrum of surface crater profiles highlights two spatial periods, one (LSFL) close and the other (HSFL) smaller to the laser wavelength as shown in Fig. 4. The variation of the ripples period with the wavelength is illustrated in Fig. 5a and b at an intensity of \( 7.7 \times 10^{19} \, \text{W.m}^{-2} \) and \( 1.5 \times 10^{20} \, \text{W.m}^{-2} \), respectively. It can be noticed that the space between ripples increases with the wavelength and intensity such as established by the experimental measurements [13,14,22].

To explain the formation of ripples on the target, we adopt the mechanism based on the Coulomb interactions and the inhomogeneous density of free electrons. In fact, the upper part of the resulting crater contains only few or even no free electrons. The Coulomb repulsion between positive ions forming the crystal lattice is no longer shielded by the presence of these electrons. At some point, the repulsion will prevail over the atomic bonds. This will result in ejection of the ions that duplicates the shape of the electron
density and explaining the observed periodicity. These structures have the form of ripples perpendicular to the laser polarization.

The interference mechanism, commonly accepted, involves the Kerr effect and the formation of HSFL involves the process of second harmonic generation (second order), in addition rough surface is assumed [12]. The mechanism of the Coulomb explosion proposed in Ref. [13], involves the stimulated Raman scattering, which is a non-linear process of the third order. Our model assumes a linear interaction (first order) of the electromagnetic wave with matter and a smooth surface. Without rejecting these two mechanisms, we suggest a much easier explanation. The explanation of the Coulomb explosion was suggested by a DFT molecular dynamic calculation on a copper dimmer. We found a dissociation of the dimmer for double ionization. The stability of the dimmer Cu$_2$ disappears when it is ionized twice (Cu$_2^{2+}$). By extrapolation, it is considered that this phenomenon persists at higher temperature and for larger clusters and surface atoms. As an indication, the Cu–Cu distance is 7.53 Å for Cu$_2^{2+}$ dimmer and the energy of dissociation is close to zero.

4. Conclusion

In order to simulate the formation of the periodic structures on solid target irradiated by an ultra-short laser pulse, we have developed in this paper an improved model which couples PIC and FDTD methods to describe the early stage of the interactions in the created plasma. Our model has been applied to the copper target to simulate electron density profiles and spatial period for various wavelength and laser intensities. We have shown the formation of LIPPS in form of ripples with two periods corresponding to either the LSFL or the HSFL structures. To explain the formation of ripples on the target, we have considered the mechanism of Coulomb repulsion due to an inhomogeneous density of free electrons. The validity of our model has been demonstrated by comparing our numerical results with available literature experimental data. The model has been applied to copper, but it can certainly be used for any metal. Our perspectives will focus on the application of this model to semi-conductor and dielectric targets [23].

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References