Surface heating of electrons in cluster plasma irradiated by intense ultrashort laser pulses

V.P. Krainov

Moscow Institute of Physics and Technology, 141700 Dolgoprudny, Moscow Region, Russia

Femtosecond laser pulses have proven to provide valuable insight into the dynamics of microscopic systems. Applied to atomic clusters even a single pulse of varying pulse duration can reveal how and when energy from the laser pulse is transferred effectively to the cluster. When an electron is heated in laser field? A free electron does not absorb energy from the monochromatic electromagnetic field. The reason is that the energy conservation law and the momentum conservation law cannot be satisfied simultaneously. Moreover, a free electron does not absorb practically energy from the ultra short laser pulse containing only two-three field periods if this pulse does not contain breaks and the integral of the field strength over all pulse duration is zero. There are two ways for laser energy absorption by an electron. The first is strong distortion of laser pulse: superposition of incident and reflected waves, deep focusing etc. The second is sharp change of electron motion. A third body should produce sharp change in the electron motion. There are several types of such third bodies. The main experimental observables for energy transfer to atomic clusters and the major theoretical approaches are contained in review papers [1-2]. In atomic clusters irradiated by femtosecond laser pulses, plasmas with high density and high temperature are created. The heating can be caused by inverse induced bremsstrahlung, i.e. determined by the electron–ion collisions inside the cluster. Inverse bremsstrahlung (IB) heating, an important process in the laser–matter interaction, involves two different kinds of interaction — the interaction of electron with the external laser field and the electron–ion interaction. An electron can both absorb and emit laser photon, but the probability of absorption is a little larger than the probability of emission. The average energy that is absorbed at one electron-ion collision in the monochromatic linearly polarized laser field with the field strength amplitude E and with the frequency ω is twice average ponderomotive (oscillation) energy of an electron in the electromagnetic field.

In order to derive the absorption rate we should multiply (1) by frequency of electron-ion collisions ν_{ei}. In the case of strong laser field when the oscillation (quiver) electron velocity $V_q = eE/m\omega$ is larger than the thermal (drift) electron velocity $V_d = \sqrt{T_e/m}$ (T_e is the electron temperature in cluster plasma), the frequency of electron-ion collisions strongly decreases with the field strength [3]: $\nu_{ei} \sim E^{-3}$. Thus, the absorption rate decreases as E^{-1}. It should be noted that free electrons in cluster plasma can be always considered as classical particles because of sufficiently high electron temperature.

In the case of cluster plasma this situation occurs typically near the peak of femtosecond laser pulse when spherical cluster strongly expands and the frequency of surface plasmon $\sqrt{4\pi e^2 n_e / 3m}$ (n_e is the electron number density) becomes to be equal to the laser frequency (so called Mie resonance). Coulomb forces in the positively charged cluster appearing due to its outer ionization produce this expansion. The role of hydrodynamic expansion is small. The inner electric field strength strongly increases due to Mie resonance. Hence, the electron-ion collision frequency strongly decreases.

Then the second mechanism of electron heating appears [4-5]. An electron can be quasi elastically reflected from cluster surface. In metal clusters, this reflection is explained by electric image force [6]. In the case of Van der Waals atomic clusters, the main reason is positive charge of cluster due to its outer ionization. Surface damping mechanism removes practically the Mie resonance. The inner electric field becomes small near the peak of laser pulse. Thus, free electron can move freely inside the cluster from one collision with surface to other one. However, near the cluster surface electric field coincides practically with the external laser field. At each reflection in the presence of the laser field an electron achieves the twice average ponderomotive energy (1) from laser field. Megi et al [4] choose for the electron-surface collision frequency the simple qualitative expression

$$\nu_{es} = \frac{V}{R} \tag{1}$$

where R is the cluster radius and $V = \sqrt{V_d^2 + V_q^2}$. Thus, they assume that the velocities V_d and V_q are perpendicular to each other. Micheau et al [5] used this model at the analysis of electron heating to describe the interaction of intense laser pulses with rare-gas clusters.

The electron heating can occur also when an electron goes through the cluster surface. So called vacuum heating mechanism (or Brunel mechanism) is considered in [6] in application for clusters: an electron is ejected by the laser field from the cluster and in a half of a laser period this electron returns back to the cluster; it acquires the kinetic energy of the order of the ponderomotive energy. This mechanism is significant when the excursion length $eE / m\omega^2$ is less than the cluster radius R. In the case of the cluster radius R = 20 nm, this corresponds to the laser field intensity less than an atomic value 3×10^{16} W/cm^2.

In this paper, we consider in detail the process of electron reflection from cluster surface and electron heating, in comparison to qualitative estimates of Megi et al [4].

The goal of this research is to determine the electron energy gain at one reflection from the cluster surface in the presence of laser field near cluster surface (see also our paper [7]). Inside the cluster we assume the free electron motion. Of course, this gain depends on the reflection angle θ . Unlike Megi et al [4], we do not assume that drift velocity is perpendicular to quiver velocity.

Let us introduce notations $\mathbf{V}_d$ for the drift electron velocity and $\mathbf{V}_q = \frac{\mathbf{E}}{\omega}$ for the amplitude of the electron quiver velocity. Here $\mathbf{E}$ and ω are the field strength amplitude and field frequency of the linearly polarized electromagnetic field. For sakes of simplicity we use here atomic units $e = m = 1$.

Further we should average the energy gain over the time moment of the reflection t_0 (this averaging is equivalent to the averaging over the field phase)

$$\langle \delta\varepsilon \rangle_{t,t_0} = \frac{e^2 E^2}{2m\omega^2}\left(1-\cos\theta\right) \ . \tag{2}$$

We restore here electron charge and mass. This expression is valid for average electron heating at one electron–ion collision when an electron scatters on the angle θ . After averaging on the angle θ one obtains that at each collision with an atomic ion, or with plasma surface an electron absorbs twice ponderomotive energy $e^2E^2/2m\omega^2$ (see Eq. (1)). Thomas Bornath (private communication) has obtained this expression for partial case when $\mathbf{V}_d \parallel \mathbf{V}_q$.

In this approach we assumed that ionization process occurs mainly nearly cluster surface. From kinetic energy distributions, they found that multiply charged ions were generated near the cluster surface. Their results suggest that charges are inhomogeneously redistributed in the cluster to lower the total energy stored in the clusters.

Thus, electron energy gain due to reflection of an electron from cluster surface is

$$\Delta\varepsilon_{es}(\theta) = \frac{1}{2}V^2(t=+\infty) = \frac{\pi E_0^4 \tau^2}{16\omega^4 R^2}\left(1-\cos\theta\right). \tag{3}$$

Averaging this expression over the solid angle: $\frac{1}{4\pi}\int d\Omega\left(...\right)$, finally one obtains

$$\delta\varepsilon_{es} = \frac{\pi}{16}\frac{e^2E^2}{m\omega^2}\left(\frac{eE\tau}{m\omega R}\right)^2 \tag{4}$$

When the electron kinetic energy is larger than the potential energy of its attraction to the cluster with the charge Q, this electron is not reflected from the cluster surface, but goes out the cluster. Hence, according to Eq. (4) the cluster charge Q (the number of ejected electrons) is determined from the relation

$$\Delta\varepsilon_{es} = \frac{\pi E_0^4 \tau^2}{16\omega^4 R^2} = \frac{Q}{R}. \quad (5)$$

Other $N - Q$ electrons are remaining inside the cluster. Here

$$N = Zn_i \frac{4\pi}{3} R^3 \quad (6)$$

is the total number of electrons produced during the laser pulse in the cluster. The quantity n_i is the number density of atoms in the cluster, Z is the final charge multiplicity of atomic ions inside the cluster after inner ionization process. Reflection of an electron from cluster surface is possible when the kinetic electron energy $\varepsilon < Q / R$.

In above consideration we assumed the sharp cluster surface. Linear and nonlinear light scattering and absorption in free-electron nanoclusters with *diffuse surface* was investigated recently in Ref. [8], especially the properties of the linear Mie resonance (the width and position). Authors of Ref. [9] using PIC simulations demonstrated that the competition between surface collisions and radiation damping is responsible for the maximum in the size dependent lifetime of the Mie surface plasmon. Besides of this, the plasma waves are excited by electrons recolliding with the cluster surface and travel toward the center, where they collide and break [10]. These new aspects of role of cluster surface in the electron heating require further analysis. Detailed discussion of our results can be found in Ref. [11[.

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