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Size effect in electron–lattice energy exchange in small metal particles

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Abstract

The paper examines electron–lattice energy exchange in confined metal systems (metal islands). An expression is derived for the energy, which an electron loses per unit of time to initiate acoustic oscillations in the lattice in accordance to the Cherenkov mechanism. In confined metal systems an electron moves, oscillating, from one potential wall to the other. The expression obtained for energy exchange converts to the generally known expression for bulk metals when the distance between the potential walls is increasing. The intensity of the bulk electron–lattice energy exchange oscillates and tends to zero at reaching a certain size with decrease of the distance between the walls.

Keywords: Hot electrons; Metal islands films; Electron–solid interactions; Size effect; Nanoelectronics; Non-equilibrium; Models of non-equilibrium phenomena; Energy dissipation

1. Introduction

In recent years, there have been a number of researches studying size effects and peculiarities of the electron–lattice energy exchange in small metal particles (SMPs) [1–5]. Interest in these objects is caused by the physical properties of small particles that can differ significantly from analogous properties of bulk materials. For example, non-equilibrium (hot) electrons can be readily obtained in SMPs while relatively small power is applied. Hot electrons can be obtained under current excitation of a film [6] as well as under laser one [7,8] in island metal films that are an ensemble of SMPs coupled by tunneling. It is significant that hot electrons in island metal films can be observed in stationary conditions and without any damage of the film structure in contrast to bulk metals and continuous film. The existence of hot electrons in these films results in electron and photon emissions [8,9]. These phenomena are observed when applied electric field strength or relatively low laser irradiation intensity. However, nothing similar is observed in continuous films or bulk metals. Island metal films are very promising materials for optoelectronics (see, for example, review [3]) due to their some particular attributes.

New features appear in properties of small particles and their ensembles when a characteristic size of a small particle is comparable with one of physical values with dimensionality of length (such as the electromagnetic wave length, the electron or photon free path, the de Broglie wavelength, the depth of skin-layer and others). In particular, when the size of metal particle is smaller than the electron free path, this affects both the electron relaxation time in pulse and in energy. In the case of an asymmetrical particle, an influence of size on the relaxation time in pulse manifests itself in turning the optical conductivity into a tensor value [10,11]. The tensor character of optical conductivity consequently influences the dependence of absorption of a particle of the electromagnetic wave polarization as well as the shape of bands of plasma resonances.

As for the influence of the size effect on the relaxation time in energy, the influence reveals itself in the electron–lattice energy exchange. This energy exchange determines the electron temperature if there are hot electrons in SMPs.

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When it comes to SMPs, the electron–lattice energy exchange is determined both by bulk and by surface electron dispersion. It is supposed, as a rule (see, for example, [3]), that in SMPs, the volume contribution in the energy exchange remains constant with increasing particle size, and the surface contribution is only added to it. However, this assumption does not represent the facts.

In this work, an analytic expression has been obtained for the bulk electron–lattice energy exchange in SMPs. This expression gives the size dependence of the bulk energy exchange in SMPs. When a particle size far exceeds the size of electron's mean free path, the obtained expression transforms to the known expression for bulk metal. When a particle size decreases and becomes smaller than the electron mean free path, the bulk energy exchange in SMPs reveals oscillating dependence on the particle size and converts to zero at a critical size. The size may be considered as critical when the transit frequency of the electron oscillations between walls is compared with the maximum frequency of the lattice oscillations (the Debye frequency). This criterion has clear physical meaning.

Though contribution of the surface dispersion in the energy exchange increases with decreasing particle size, it is still smaller than the bulk contribution by 1–2 orders of magnitudes under the particle sizes of order of the electron mean free path (see estimations in [12,13]). Therefore, when the particle size diminishes and reaches the size of order of the electron mean free path, initially the summary intensity of the electron–lattice energy exchange in SMPs falls sharply (by 1–2 orders), and only then starts to rise. This peculiarity of the energy exchange in SMPs leads to the observation of hot electrons in stationary conditions in island metal films only. However, they are not observed either in bulk metal or in continuous films. Of course, hot electrons can be obtained in the last-named objects under an action of short and powerful pulses, but it is possible only during the pulse operation mode.

Let us consider now attempts to calculate the electron–lattice energy exchange in SMPs from the basic principles [2]. In these attempts, the electron–lattice interaction is interpreted as the electron–ion interaction by a perfect analogy with gas plasma. A distinction lies only in using the Fermi distribution function of electrons instead of the Maxwell distribution.

Importantly that the biggest part of the electron–ion interaction is already taken into account during determination of the Hartree–Fock self-consistent potential. As it is known, the self-consistent potential determines the dispersion law of electron in metal. The electron–ion interaction can be taken into account just as it was made in [2] only for a high-speed electron running through a particle. For this electron, the self-consistent periodic potential can be neglected. For a thermal electron, dispersion is determined by deviation of ion from its equilibrium position caused by the lattice oscillations. This type of dispersion is adequately taken into account in the model of deformation potential of Bardeen–Shokley. This approach has been used in the present work. It has been already utilized earlier in the investigation of the electron–lattice energy exchange in bulk metal [14]. While applying this approach for SMPs, the circumstance that motion of an electron becomes oscillatory (electron "runs" from one wall to another) when the particle size is smaller than the electron mean free pass also should be taken into consideration.

2. Generation of acoustic oscillations by an electron moving in the crystal lattice

In this paragraph, we will derive equation, which describes the generation of acoustic oscillations of the lattice by an electron moving in it. We remind that the basic way of the electron energy relaxation in a bulk metal is generation of the lattice acoustic oscillations by the electron according to the Cherenkov mechanism. This relaxation mechanism will be of interest to us in small metal particles.

Let us write the Hamiltonian of the electron interaction with the lattice atoms in the point $\underset{\sim}{\vec{r}}$ in the following form:

$$\widehat{H}_{\mathrm{int}} = \sum_{\underset{\sim}{\vec{n}}} \{V(\underset{\sim}{\vec{r}} - \underset{\sim}{\vec{R}}(\underset{\sim}{\vec{n}}) - \underset{\sim}{\vec{u}}(\underset{\sim}{\vec{n}})) - V(\underset{\sim}{\vec{r}} - \underset{\sim}{\vec{R}}(\underset{\sim}{\vec{n}}))\}$$

$$\approx -\sum_{\underset{\sim}{\vec{n}}} \underset{\sim}{\vec{u}}(\underset{\sim}{\vec{n}}) \frac{\partial}{\partial \underset{\sim}{\vec{r}}} V(\underset{\sim}{\vec{r}} - \underset{\sim}{\vec{R}}(\underset{\sim}{\vec{n}})). \tag{1}$$

Here, V is the atomic potential, $\underset{\sim}{\vec{R}}(\underset{\sim}{\vec{n}})$ is the radius vector of the $\underset{\sim}{\vec{n}}$th lattice point, and $\underset{\sim}{\vec{u}}(\underset{\sim}{\vec{n}})$ is small displacements of the $\underset{\sim}{\vec{n}}$th atom caused by the lattice oscillations. For the simple lattice

$$\underset{\sim}{\vec{R}}(\underset{\sim}{\vec{n}}) = \sum_{i=1}^{3} n_i \underset{\sim}{\vec{a}}_i, \tag{2}$$

where $\underset{\sim}{\vec{a}}_i$ is the translation vector.

The potential energy of lattice oscillations can be expressed through small displacements of atoms from the equilibrium position in traditional form

$$P = \frac{1}{2} \sum_{\underset{\sim}{\vec{n}},\underset{\sim}{\vec{n}}'} \widehat{A}(\underset{\sim}{\vec{n}} - \underset{\sim}{\vec{n}}')\, \underset{\sim}{\vec{u}}(\underset{\sim}{\vec{n}})\, \underset{\sim}{\vec{u}}(\underset{\sim}{\vec{n}}'), \tag{3}$$

where $\widehat{A}(\underset{\sim}{\vec{n}})$ is the force matrix.

The kinetic energy of oscillations of the lattice atoms of which mass is M, will be

$$T = \frac{M}{2} \sum_{\underset{\sim}{\vec{n}}} (\dot{\underset{\sim}{\vec{u}}}(\underset{\sim}{\vec{n}}))^2. \tag{4}$$

Taking into account the kinetic energy (4) and potential (3), as well as the energy of electron–lattice interaction (1), it is easy to obtain the lattice oscillation equation

$$M \ddot{\underset{\sim}{\vec{u}}}(\underset{\sim}{\vec{n}}) + \sum_{\underset{\sim}{\vec{n}}} \widehat{A}(\underset{\sim}{\vec{n}} - \underset{\sim}{\vec{n}}')\, \underset{\sim}{\vec{u}}(\underset{\sim}{\vec{n}}') = -\frac{\partial}{\partial \underset{\sim}{\vec{u}}(\underset{\sim}{\vec{n}})} \widehat{H}_{\mathrm{int}} = \frac{\partial}{\partial \underset{\sim}{\vec{r}}} V(\underset{\sim}{\vec{r}} - \underset{\sim}{\vec{R}}(\underset{\sim}{\vec{n}})). \tag{5}$$