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A New Model of the Electron Gas Effect on the Thermoacoustics of Conductors under Laser Irradiation

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Abstract—A two-component model which accounts for electron gas pressure is proposed for describing the dynamics of thermoelastic and thermoacoustics effects in laser-irradiated conductors. The model medium represents two interpenetrating continua such that interacting particles of both exist at each point of the medium. The electron gas in the model comprises free and bound electrons of which the former obey the laws for perfect metals and the latter obey those that account for electron trapping to localized levels and for electron transitions from level to level, i.e., for jump diffusion and hopping conductivity. Unlike the classical model of thermoelasticity, the proposed model is the first to show that the electron gas pressure depends strongly on the temperature difference between the electron gas and the conductor lattice and on the change in the density of free electrons as localized species become free by the Mott mechanism. The duration of acoustic pulses in the conductor lattice is essentially dependent on the time of laser irradiation and on how long the gas and the lattice differ in temperature, with the longest acoustic pulse falling on a certain localized electron density. The model data are compared with experiments.

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1. INTRODUCTION

Modern methods of nondestructive testing and research in thermophysical and elastic properties of bulk materials, thin films, and thin film structures widely use laser irradiation [1], which allows one to trace the dynamics of laser energy conversion to heat and then to acoustic vibrations or waves in various materials. Mostly, related theoretical and experimental studies deal with thermoelastic generation of elastic strains and acoustic waves in solids. In one of the pioneering theoretical works on the subject [2, 3], a coupled problem of thermoelasticity was considered and its solutions in the linear approximation were derived for the spatial and temporal evolution of strains in solids under pulsed irradiation.

Unfortunately, all known theoretical approaches to thermoelastic processes in metals at the initial stage of laser irradiation fail to provide even a qualitative agreement with experiments [4, 5] showing that all conductors, like most metals, feature a substantial delay of deformation compared to the prediction of quite adequate theoretical models of acoustic pulses in nonconducting materials under thermal load [4].

The use of pulses ranging from micro- and nanoseconds to pico- [6–8] and femtoseconds [9] requires a new and reliable model for describing the dynamics of thermal and elastic effects in conductors under high-frequency loads.

Here we propose a new model of thermoelasticity which accounts for the effect of the electron gas dynamics on the time stretching of acoustic pulses in conductors.

Note that a quite successful attempt to explain this phenomenon is the model of a thermal piston [5] which is supposedly created by the electron gas moving in a conductor. The model has given a qualitative explanation of the delayed thermoelastic response in metals, but the nature of the phenomenon remains unclear and its

mathematical interpretation needs correction for applying the model to other processes.

In our study, a two-component model which accounts for electron gas pressure is proposed for describing the dynamics of thermoelastic effects in conductors [10–12]. The model medium represents two interpenetrating continua such that interacting particles of both exit at each point of the medium. The internal interaction forces Q_{ie} and Q_{ei} are equal, and the indices i and e stand for the ion-electron content in the two-component medium. The electron gas in the model comprises free and bound electrons [13] of which the former obey the laws for perfect metals and the latter obey those that account for electron trapping to localized levels and for electron transitions from level to level, i.e., for jump diffusion and hopping conductivity.

According to the data available [14], the effect of external action, in particular pulsed laser irradiation, is such that part of the electrons passes from one localized state to another and their relaxation is much longer than the relaxation of free electrons at the Fermi level.

Thus, one could expect that the time during which the electron gas pressure acts on the lattice of a metal will be much longer than the time of electron–phonon interaction. Such a phenomenon has been described in none of the papers known to us.

2. PROBLEM STATEMENT. BASIC EQUATIONS

The electron gas (first component) interacting with the lattice of a metal represents a continuum of variable density $\rho_e = m_e n_e$, where m_e is the electron mass and n_e is the electron density per unit volume.

The number of mobile electrons is small and falls on energy levels higher than the Fermi level. The concentration parameter is $\alpha_\vartheta = (k_B\vartheta_e/\varepsilon_F)^2$ [13], and its value at room temperature is very small. Hereinafter, k_B is Boltzmann's constant, ϑ_e is the electron gas temperature, and ε_F is the Fermi energy. The other part of free electrons $(1-\alpha_\vartheta)\rho_e$ resides on lower energy levels, and the kinematics of their mean motion is almost the same as that of the lattice.

Thus, we have the following balance equations for the number of free electrons:

$$\frac{\partial n_e}{\partial t} + \underline{\nabla}\cdot(n_e\underline{v}_e) = J_{es},$$
$$\frac{\partial n_e^F}{\partial t} + \underline{\nabla}\cdot(n_e^F\underline{v}_e^F) = -\frac{1}{\tau}n_e^F + \psi(\vartheta_e, \vartheta_i). \quad (1)$$

Here $n_e^F = \alpha_\vartheta n_e$ is the density of electrons at the Fermi level, $\underline{v}_e^F$ is their average velocity, τ is their relaxation time or residence time at the Fermi level, $\psi(\vartheta_e, \vartheta_i)$ is the source of electron gas excitation which determines the transition rate of part of the free electrons from lower energy levels to the Fermi level, and J_{es} is the excitation source of bound electrons which brings part of them to a localized state.

The momentum flux of free electrons $\underline{q}_e = n_e\underline{v}_e$, where $\underline{v}_e$ is a certain average velocity, has the form

$$n_e\underline{v}_e = n_e[(1-\alpha_\vartheta)\underline{v}_i + \alpha_\vartheta\underline{v}_e^F]. \quad (2)$$

The total number of free electrons in a perfect crystal $(J_{es} = 0)$ has the form

$$n_e = n_e^F + n_e^*, \quad (3)$$

where $n_e^F = \alpha_\vartheta n_e$, $n_e^* = (1-\alpha_\vartheta)n_e$.

The production of bound electrons $(J_{es} \neq 0)$ due to localized zones (defects, dislocations, etc.) increases the number of free electrons:

$$n_e = n_e^F + n_e^* + n_{es}. \quad (4)$$

The transition of localized electrons n_{es} to their free state greatly changes the electron gas pressure. Considering the electron gas as a degenerate gas [15], its pressure has the form

$$P_e = \frac{2}{5}n_e\varepsilon_F + \frac{\pi^2}{6}n_e\varepsilon_F\left(\frac{k_B\vartheta_e}{\varepsilon_F}\right)^2. \quad (5)$$

In view of the augend in (5), a relative change of order 10^{-3} in the density of free electrons can change the pressure in a metal by about 100 MPa. It is significant that such a pressure in the lattice of a metal can exist for a time much longer than the time of its jump with temperature, which is described by the well-known model of thermoelasticity.

The number of localized electrons n_{es} is determined from the balance equation

$$\frac{\partial n_{es}}{\partial t} + \underline{\nabla}\cdot(n_{es}\underline{v}_s) = -J_{es}, \quad (6)$$

where $\underline{v}_s$ is their average velocity between localization events. The source J_{es}, which describes their transitions between free and bound states, can be expressed as

$$J_{es} = \frac{n_{es}}{\tau_s} - (S - S_0), \quad (7)$$

where τ_s is the relaxation time of an electron between its bound and free states, S is a certain source (generator) which stimulates the transition of electrons from bound to free states. Reasoning from experimental data on the electrical conductivity of disordered metals, the transition of electrons can be considered to obey the Arrhenius law [16]. Then, the source S can be expressed as

$$S = A\exp\left(-\frac{U}{k_B\vartheta_i}\right), \quad (8)$$

where A is a constant, U is the activation energy. In the first approximation, provided that $U/(k_B\vartheta_i) < 1$, expression (8) can be written as

$$S - S_0 = B(\vartheta_i - \vartheta_{i0}),\ B = \frac{AU}{k_B\vartheta_{i0}^2}\exp\left(-\frac{U}{k_B\vartheta_{i0}}\right), \quad (9)$$

where ϑ_i is the lattice temperature, and ϑ_{i0} is its equilibrium value.

Thus, the basic balance equations for the number of free and bound electrons have the form

$$\frac{\partial n_e}{\partial t} + \underline{\nabla}\cdot(n_e\underline{v}_e) = J_{es},\ \frac{\partial n_{es}}{\partial t} + \underline{\nabla}\cdot(n_e\underline{v}_{es}) = -J_{es},$$
$$J = \frac{n_{es}}{\tau_s} - B(\vartheta_i - \vartheta_{i0}). \quad (10)$$

To the system of equations (10) we should add the momentum balance equation for Fermi electrons

$$-\frac{1}{m_e}\underline{\nabla}P_e = \frac{\mathrm{d}(n_e^F\underline{v}_e^F)}{\mathrm{d}t}. \quad (11)$$

Equations (10) and (11) supplemented with heat conduction equations for electron gas complete the problem statement. Certainly, the main difficulty in their solution concerns divergent terms and effect of lattice particle velocities on the total flow of free electrons. This difficulty, including the temperature and relationship between the above equations and momentum balance equations, greatly complicates the problem. However, some simplification can provide an analytical result. The key point of the model is that it accounts for the electron gas pressure as a factor dependent on the kinematics of electrons at the Fermi level and, what is more important, on the transition of electrons from localized to bound states. Such a transition strongly influences the first term in pressure (5) and radically changes the time dependence of the lattice response.

3. PROBLEM SOLUTION AND ITS COMPARISON WITH AVAILABLE THERMOELASTICITY MODELS

Without going into the details of solving the heat conduction equations, as they are comprehensively analyzed using a two-temperature model [17–19] and accurate asymptotic calculations [20], let us proceed from the hypothesis of semi-connectedness and results for the temperatures ϑ_i and ϑ_e.

The two-component model accounting for the effect of the electron gas dynamics on the lattice of a conductor is a natural generalization of classical thermoelasticity models. Neglect of the electron kinematics at the Fermi level, which is important at $\vartheta_e \neq \vartheta_i$ due to the Cherenkov effect [21], means that only electrons residing at lower levels contribute to the electron gas reaction with the lattice. Their kinematics is determined by the lattice kinematics, and the basic equations of the two-component model have the form

$$-\underline{\nabla}P_e = \underline{Q}_{ei},\ \underline{\nabla}\cdot\underline{\underline{\sigma}}_i = \underline{Q}_{ie} + \rho_i\frac{\partial\underline{v}_i}{\partial t}, \quad (12)$$

where $\underline{\underline{\sigma}}_i$ is the Cauchy tensor which determines the stress state of the metal lattice in view of the Duhamel–Neumann law, $\rho_i = m_i n_i$ is the mass density of lattice particles. The effect of the aforementioned electrons is quasi-static, and the system of equations (12) can be reduced to its ordinary form:

$$\underline{\nabla}\cdot(\underline{\underline{\sigma}}_i - P_e\underline{\underline{E}}) = \underline{Q}_{ie} + \rho_i\frac{\partial\underline{v}_i}{\partial t}, \quad (13)$$

where $\underline{\underline{E}}$ is the unit tensor. Assuming rather small perturbations of the lattice and electron gas, the balance equation for the number of free electrons after linearization gives

$$\tilde{n}_e = -n_e^{(0)}\varepsilon_i, \quad (14)$$

because $\underline{v}_e = \underline{v}_i$. Here ε_i is the bulk strain of the lattice, $n_e^{(0)}$ is the initial number of free electrons.

The second term in the equation of state (5) is quite small such that the pressure $\tilde{P}_e$ varies as

$$\tilde{P}_e = \frac{2}{5}\tilde{n}_e\varepsilon_F = -\frac{2}{5}n_e^{(0)}\varepsilon_i\varepsilon_F. \quad (15)$$

Equation (13), unlike the well-known one, contains a term with $\underline{\nabla}\tilde{P}_e$ but its effect appears only through the bulk strains and the correction is small, measuring about $n_e^{(0)}\varepsilon_F/E_i \ll 1$ [15], where E_i is Young's modulus of the lattice.

Thus, if the duration of thermal processes is much longer than the equalization time of ϑ_i and ϑ_e, their effect on acoustic pulses in a conductor is inessential; otherwise, at $\vartheta_e \neq \vartheta_i$, the kinematics of electrons at the Fermi level can affect the quantitative and qualitative distribution of acoustic pulses in the lattice. From the basic equations of the two-temperature model it follows that the lattice is heated with a delay $\tau = \rho_i c_i/G$, where c_i is the specific heat capacity of the lattice, G is a coefficient which determines the heat flux from the electron gas to the lattice [18]. In this case, the velocity of electrons at the Fermi level can be higher than the velocity of sound and they give up energy to the lattice, according to the Cherenkov effect, and go down to lower energy levels. Thus, the number of electrons is changed by

$$\tilde{n}_e^F = \frac{G}{\rho_e m_e c_e}\int_0^t(\vartheta_e - \vartheta_i)\exp\left(-\frac{t-\xi}{\tau}\right)\mathrm{d}\xi, \quad (16)$$

where c_e is the specific heat capacity of the electron gas. This expression is derived from equation (1) with the function $\psi(\vartheta_e, \vartheta_i)$ represented as

$$\psi(\vartheta_e, \vartheta_i) = G(\vartheta_e - \vartheta_i).$$

Note that (16) is certainly approximate. Generally, the divergent part should be allowed for, which leads to a complex diffusion-type equation.

Assuming that τ is very small compared to the laser pulse duration, expression (16) can be rewritten as

$$\tilde{n}_e^F = \frac{G\tau}{\rho_e m_e c_e}(\vartheta_e - \vartheta_i). \quad (17)$$

In view of the definition of n_e^F, the total density of free electrons varies as

$$\tilde{n}_e = \left[\frac{G\tau}{\rho_e m_e c_e}(\vartheta_e - \vartheta_i) - n_e^{(0)}\tilde{\alpha}_\vartheta\right]\frac{1}{\alpha_\vartheta^0}. \quad (18)$$

The pressure varies as

$$\tilde{P}_e = \frac{\varepsilon_F}{\alpha_\vartheta^0}\left[\frac{2G\tau}{5\rho_e m_e c_e}(\vartheta_e - \vartheta_i) - n_e^{(0)}\tilde{\alpha}_\vartheta\right]. \quad (19)$$

After some transformations, this expression can be reduced to the form

$$\tilde{P}_e = G^*(\vartheta_e - \vartheta_i), \quad (20)$$

where

$$G^* = G\tau\frac{\varepsilon_F}{\rho_e c_e m_e \alpha_\vartheta^{(0)}},\ \alpha_\vartheta^{(0)} = \left(\frac{k_B\vartheta_e^{(0)}}{\varepsilon_F}\right)^2.$$

The obtained value coincides with that derived from a similar expression for pressure [20] based on the data reported elsewhere [21].

The asymptotic behavior of acoustic pulses in the lattice of a conductor allows us to determine the time stretching of strain waves. However, a comparison of the results with experiments gives a difference in pulse durations by more than two orders of magnitude [4]. The pulse shapes and amplitudes estimated with account of electron gas pressure (5) provide a good description of the stress state in thin films [21, 22] when the laser pulse duration compares with the equalization time of the lattice and electron gas temperatures.

From the initial balance equations for free and bound electrons (10) and conclusions made elsewhere [23] we can determine the additional electron gas pressure on the lattice due to the transition of electrons from localized to free states. Let us consider the divergent term in the second equation of system (10). When a laser pulse and associated temperature gradient act in a metal, the metal experiences a Dember emf [24] whose typical value measures several microvolts per degree. Estimates show that in such fields the drift length of bound electrons during relaxation will be much smaller than other characteristic lengths of the problem (lengths of acoustic and thermal waves). Therefore, this equation can be simplified to the form

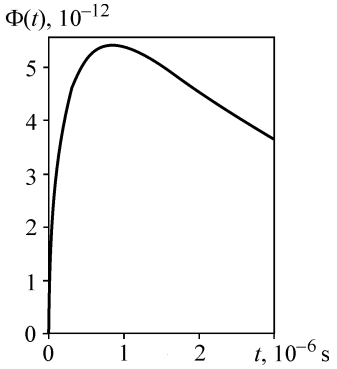

Fig. 1. Time dependence of pressure.

$$\frac{\partial n_{es}}{\partial t} = -\frac{n_{es}}{\tau_s} + B(\vartheta_i - \vartheta_i^{(0)}). \quad (21)$$

Reasoning that the initial density of localized electrons is $n_t^{(0)}$, its value at the next points in time is determined by the expression

$$n_{es}(x,t) = n_{es}^{(0)} - B\int_0^t e^{-(t-\xi)/\tau}(\vartheta_i(x,\xi) - \vartheta_i^{(0)})d\xi. \quad (22)$$

As the density of localized electrons decreases with increasing temperature, the density of free electrons increases by the same value and their pressure builds up by virtue of both summands in equality (5). At electron temperatures $T_e \le 2E_F/(\pi k_B)$, the main contributor to the pressure will be the first summand. For metals, this relation is valid up to $T_e \cong 10^3$ K. In the temperature range specified, the electron gas pressure will respond to the release of localized electrons as

$$\tilde{P}_e(x,t) = 2/5\,\varepsilon_F\tilde{n}_{es} = 2/5\,\varepsilon_F B\Phi(t), \quad (23)$$

where

$$\Phi(t) = \int_0^t e^{-(t-\xi)/\tau}(\vartheta_i(x,\xi) - \vartheta_i^{(0)})d\xi.$$

Figure 1 shows the time dependence of this function at $\tau_s = 10^{-6}$ s for a laser pulse duration of 10^{-8} s.

Thus, in the general case, the electron gas pressure is determined by two summands:

$$\tilde{P}_e = G^*(\vartheta_e - \vartheta_i) + 2/5\,\varepsilon_F B\Phi(t), \quad (24)$$

of which the first is governed mainly by the temperature difference of the electron gas and lattice, and the second by the number of free electrons. The first summand shows a substantial effect for about as long as the gas and lattice temperatures differ, and the second one exists for as long as jump diffusion provides the transition of an electron from its localized to free state. As the problem solution suggests, the time of jump diffusion compares with the travel time of an acoustic wave through the specimen thickness. A comparison shows that the

curve in Fig. 1 agrees qualitatively with experimental data [4], suggesting that the stress state of conductors under short-term laser irradiation should be calculated using formula (24) to account for the additional pressure associated with the Mott phenomenon [23].

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