Brief Reports

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Transient effects in the electron temperature and phonon distribution in thin metal films

M. N. Wybourne
Department of Physics, University of Oregon, Eugene, Oregon 97403

N. Perrin
Laboratoire de Physique de la Matière Condensée de l'Ecole Normale Supérieure, 24 rue Lhomond, 75231 Paris CEDEX 05, France

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We present a model to describe the evolution of the phonon energy density and the electron temperature in a heated metal film in which spontaneous phonon decay is present. The phonon-decay processes are shown to have several effects: First, the characteristic temperature of the electron system is lower than predicted by other models; second, the electrons reach a steady state more quickly than they do without the phonon processes; and finally, the high-frequency component of the longitudinal-phonon spectra is attenuated.

I. INTRODUCTION

Joule heating of thin metal films has been used for many years as a convenient method of producing phonon pulses with which to study phonon propagation in solids.[1] Recently, heat-pulse experiments on the nanosecond time scale have been used to examine the dynamics of phonons within the metal films themselves.[2] In these experiments it was observed that at high-excitation power densities phonons generated in the film by electron relaxation processes undergo spontaneous decay before escaping from the film. A steady-state analysis of the experiments was performed using the Perrin and Budd model[3] modified to include three-phonon decay processes.[4] We now extend the analysis to study the temporal evolution of both the electron temperature and the phonon distribution within the metal film. We will assume that the electron system has a well-defined temperature at all times.

II. MODEL

The evolution of the electron temperature T_e may be described by the equation

$$C_e\frac{dT_e}{dt}=\frac{E^2}{\rho}-\sum_{\mathbf{q}}\hbar\omega(q)\left[\frac{N_L(q,T_e)-N_L(q)}{\tau_{e\text{-ph}}(q)}\right], \tag{1}$$

where C_e is the specific heat of the electrons (γT_e), E is the applied electric field, ρ is the resistivity of the film, $\tau_{e\text{-ph}}(q)$ the electron-phonon relaxation time for phonons of wave-vector magnitude q, and $N_L(q,T_e)$ is the Bose distribution of longitudinal phonons at the electron temperature. The nonequilibrium phonon distribution $N_L(q)$ is obtained by evaluating

$$\frac{\delta N_j(q)}{\delta t}=\frac{N_j(q,T_e)-N_j(q)}{\tau^j_{e\text{-ph}}(q)}+\frac{N_j(q,T_0)-N_j(q)}{\tau^j_x} \tag{2}$$

for the longitudinal model ($j=L$), where T_0 is the substrate temperature. The scattering rate τ_x^{-1} is a combination of the phonon escape rate from the film $(\tau_b^j)^{-1}$ and the relaxation rates $\tau_1(q)^{-1}$, $\tau_2(q)^{-1}$, and $\tau_3(q)^{-1}$ that describe the three spontaneous down conversion processes, $L\rightarrow L+T_2$, $L\rightarrow T_1+T_1$, and $L\rightarrow T_2+T_2$, respectively.

In the calculations we have used the usual expression to describe the rate of phonon escape from the film to the substrate, $(\tau_b^j)^{-1}=v_j/4\eta d$, where v_j is the sound velocity of mode j, d is the film thickness, and η is an acoustic mismatch parameter which we assume to be frequency independent. The relaxation rates $\tau_1(q)^{-1}$, $\tau_2(q)^{-1}$, and $\tau_3(q)^{-1}$ are all proportional to q^5 and depend on the second- and third-order elastic constants of the metal film in a nontrivial way. Complete expressions for the phonon down-conversion rates are given in Refs. 5, 6, and 4. In order to simplify the problem, we consider the film to be a simple metal with a spherical Fermi surface. In this case, if we neglect umklapp processes, the electrons can only couple to the longitudinal phonons. Furthermore, if we assume the metal to be clean, that is $ql>1$, where l is the elastic mean-free path of the electrons, the electron-phonon relaxation rate is given by[7]

$$\tau_{e\text{-ph}}^{-1}\equiv\Xi^2m^2q/2\hbar^3\pi\rho_D\ , \tag{3}$$

where Ξ is the deformation potential, m is the electron mass, and ρ_D is the mass density of the film.

III. RESULTS

We have solved numerically the coupled equations (1) and (2) to calculate the electron temperature in a gold film on sapphire as a function of time after the onset of excitation. The parameters we have used are as follows: electron density is 6×10^{28} m^{-3}, $\rho=5\times10^{-8}$ Ω m, $\rho_D=19\,300$ Kg m^{-3}, and $\Xi=3.77$ eV. From the elastic constants of gold we estimate the velocities of sound to be $v_L=3.42\times10^3$ m s^{-1} and $v_T=1.32\times10^3$ m s^{-1} and $\eta=12.5$. Using these parameters we find that $(\tau_b^L)^{-1}=68/d$ s^{-1}, $(\tau_b^T)^{-1}=26/d$ s^{-1}, where d is the film thickness, and that the relaxation rates describing the three-phonon processes are $\tau_1(q)^{-1}=7\times10^{-41}\,q^5$ s^{-1}, $\tau_2(q)^{-1}=2\times10^{-40}q^5$ s^{-1}, and $\tau_3(q)^{-1}=4\times10^{-40}\,q^5$ s^{-1}. The magnitude of the isotropic zone-boundary wave vector q_{ZB} for gold was estimated from the low-temperature lattice constant of 0.39 nm and the fcc structure to be 1.72×10^{10} m^{-1}.

Numerical convergence of the data was checked by calculating the electron temperature using several different time intervals. It was found that convergence to better than 1% was achieved when the time interval was reduced below $\sim10^{-2}$ the value of the fastest phonon relaxation process.

The model has been applied to a 10-nm-thick film subject to an electric field $E=1.55\times10^5$ V m^{-1}. Figure 1 shows the evolution of the electron temperature calculated with and without the three-phonon processes. The minimum relaxation time for the phonon down-conversion process is $\tau_3(q_{ZB})=1.7$ ps. At times less than $\tau_3(q_{ZB})$ the electron temperature is unaffected by the presence of phonon down conversion. At times longer than all of the relaxation times in the problem, the electron temperature reaches a steady-state value T_{es}^* that is considerably lower than the value T_{es} attained without the phonon decay. This may be understood as enhanced heat loss from the electron system by the three-phonon processes. Furthermore, the heightened heat loss also acts to bring the electron system into the steady state more rapidly.

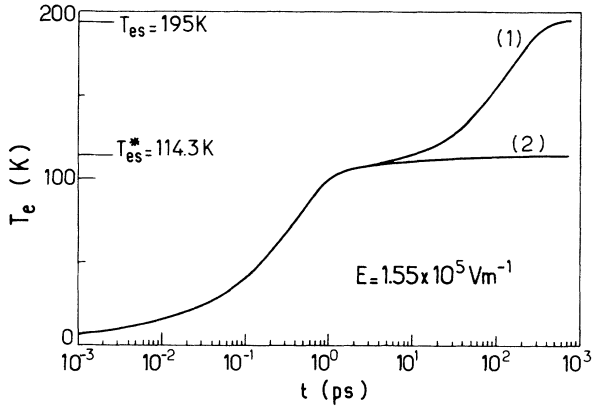

FIG. 1. The electron temperature in the gold films as a function of time after the electric field is applied (1) without and (2) with three-phonon processes.

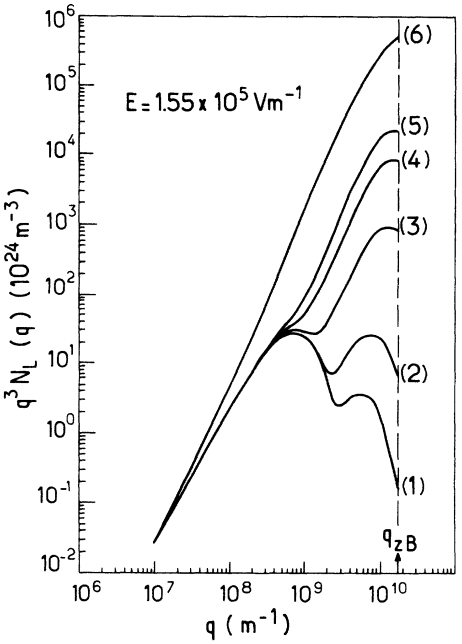

FIG. 2. The evolution of the longitudinal-phonon energy density spectrum without three-phonon processes. The times are in picoseconds: (1) 0.125, (2) 0.25, (3) 1.0, (4) 3.75, (5) 8.75, and (6) 601.25.

An interesting feature of the evolution of the electron temperature without phonon decay is the nonuniform increase of the electron temperature with time. This is no-

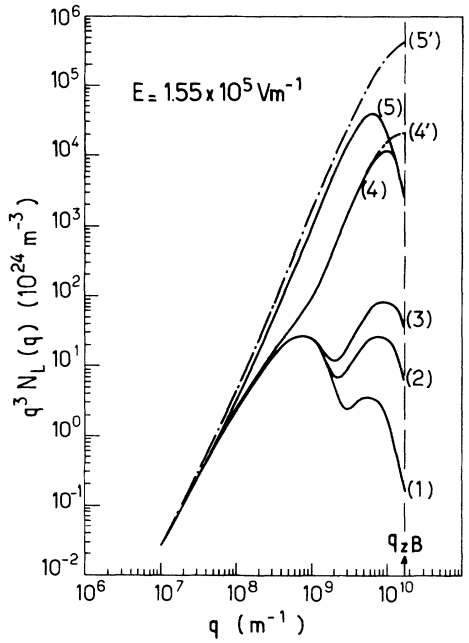

FIG. 3. The evolution of the longitudinal-phonon energy density spectrum with three-phonon processes. The times are in picoseconds: (1) 0.125, (2) 0.25, (3) 0.375, (4) 8.75, and (5) 301.25. To show the effect of the phonon processes clearly, (4') and (5') are the 8.75- and 601.25-ps curves from Fig. 2.

ticed in Fig. 1 as a leveling off of the electron temperature at times ~ 1 ps. We can interpret this behavior as follows: at times less than 1 ps the electron system gains energy from the electric field yet does not transfer much energy to the phonons since electron-phonon scattering is very improbable at these times. The small amount of phonon heating that does occur on this time scale is seen in Fig. 2 but the rate is insufficient to prevent the electron system from continuing to rise in temperature. At later times the probability of electron-phonon coupling increases until at approximately 3 ps the energy heating the electron system is balanced by the energy leaving the electrons to heat the phonon system. Since the phonon escape time is 147 ps, the combined electron-phonon system then continues to heat under the action of the electric field until phonon escape to the substrate causes the whole system to reach a steady state at ~ 600 ps. Figures 2 and 3 show the nonequilibrium energy density spectra for the longitudinal phonons calculated without and with three-phonon processes, respectively; it is apparent that at times less than $\tau_3(q_{ZB})$ the spectra are independent of the phonon-decay processes. A complete discussion of the evolution of the nonequilibrium longitudinal-phonon spectra and the generation of transverse phonons will be presented elsewhere.

IV. CONCLUSION

We have shown that for thin metal films excited by short duration electric fields, the steady-state electron temperature and phonon spectra are dependent on three-phonon processes within the films. Without these processes both the electron temperature and the phonon spectrum take longer to reach the steady state and the electron temperature is considerably overestimated.

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