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Electronic Kapitza conductance due to inelastic electron-boundary scattering

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Inelastic electron scattering at the interface between a conducting film and an insulating substrate provides a new channel for energy transfer from the film electrons to the substrate phonons. Its contribution to the Kapitza conductance is found to be $\hbar u \gamma / k_B \tau_{e-ph}$, where u is the sound velocity, γ is the Sommerfeld constant, and τ_{e-ph} is the electron-phonon energy relaxation time in the film. This mechanism is significant for conductors with strong electron-phonon coupling, or for an interface with a small value of the phonon transparency. The results of the theory agree in order of magnitude with the observed decrease of the Kapitza conductance at the transition to the superconducting state. They can explain a universal minimal value of the conductance for pairs of materials with rather different acoustic impedances (metallic films on diamond substrate). This scattering mechanism results in the nonequilibrium component of the photoresponse with the ps decay time proportional to the film thickness that has been recently observed in YBaCuO ultrathin films. [S0163-1829(98)52336-3]

I. INTRODUCTION

As it was first found by Kapitza,[1] a heat current I_h passing through a boundary results in a temperature jump ΔT at the interface. The proportionality coefficient relating the temperature jump to the heat current is known as Kapitza conductance (σ_K),

$$I_h = \sigma_K \Delta T. \tag{1}$$

Up to now the Kapitza conductance has been measured for many pairs of materials. Recent interest to the thermal conductance between conducting (metallic, superconducting, or semiconducting) film and dielectric substrate has sprung from thin-film device applications.[2]

If acoustic impedances of the two media are not very different, the Kapitza conductance is well described by the acoustic mismatch theory.[3] There exist, however, experimental results that cannot be explained within the framework of the acoustic mismatch theory (for an extensive review, see Ref. 4). In the general case the acoustic mismatch model gives only a low limit of the boundary conductance. Discrepancy between experimental data and predictions of the acoustic mismatch theory is most significant for the metal-insulator contacts. For materials with rather different acoustic impedances this discrepancy reaches two orders of magnitude.[4,5]

Another well-known experimental fact, which cannot be explained by the acoustic mismatch theory, is that the Kapitza conductance between a superconducting film and a substrate depends on the superconducting energy gap. In the superconducting state the Kapitza conductance turns out to be smaller than in the normal state, and for different pairs of materials the conductance change varies from a few percent to several times.[4]

Recent studies of the film-substrate Kapitza conductance are based on measurements of the transient bolometric photoresponse.[2,5–8] The value of the phonon Kapitza conductance, which was obtained in experiments with relatively thick films of high-T_c superconductors,[6–8] corresponds to the value found from stationary measurements.[9] But the ps time of the photoresponse decay in ultrathin YBaCuO films[10] (thickness $d = 150-300$ Å) gives the phonon conductance value being more than one order of magnitude as larger than the value found in Refs. 6–9. Up to now there has not been any interpretation of these data.

The experimental results mentioned above are evidence of the significant role the conducting electrons play in the boundary conductance. In the present paper we suggest a new universal mechanism of the electronic Kapitza conductance. In our model an electron is scattered from the confining potential of the interface and emits a phonon, which moves from the interface into the substrate. Due to this process the energy transfers from the film electrons to the substrate phonons. To our knowledge, the suggested mechanism has not been considered previously in papers[11–14] devoted to the electron contribution to the Kapitza conductance.

II. CALCULATION OF CONDUCTANCE

In this section details of calculations are presented. Due to a large value of the transferred momentum and local character of the interaction, inelastic electron-boundary scattering may be considered in the same way as the inelastic electron-impurity scattering.[15] The corresponding Hamiltonian has a form

$$H_{int} = \sum_{\mathbf{p},\mathbf{k},\mathbf{q},n,\mathbf{R}_{bd}} \gamma(\mathbf{k},\mathbf{q},n) c^+_{\mathbf{p}} c_{\mathbf{p}-\mathbf{k}} (b_{\mathbf{q},n} + b^+_{-\mathbf{q},n}) \times \exp[-i(\mathbf{k}-\mathbf{q})\mathbf{R}_{bd}], \tag{2}$$

where $c^+_{\mathbf{p}}$ is the creation operator of an electron with momentum $\mathbf{p}$, $b^+_{\mathbf{q},n}$ is the creation operator of a phonon with a wave vector $\mathbf{q}$ and polarization index n, and $\mathbf{R}_{bd}$ are the equilibrium positions of atoms at the boundary. The vertex of the electron-boundary scattering is

$$\gamma(\mathbf{k},\mathbf{q},n)=-iV_{e-bd}\frac{\mathbf{k}\mathbf{e}_n}{(2MN\omega_{\mathbf{q},n})^{1/2}}, \tag{3}$$

where V_{e-bd} is the boundary scattering potential, $\mathbf{e}_n$ is the phonon polarization vector, M is the ion mass, and N is the ion concentration.

To calculate the energy flux from film electrons to substrate phonons due to inelastic electron-boundary scattering we will find the corresponding collision integral. Employing the Keldysh technique to this problem, we describe the electron and phonon subsystem by advanced (A), retarded (R), and kinetic (K) Green's functions. To calculate the heat flux from electrons to phonons we will assume that the electron and phonon subsystems have different temperatures θ and T, respectively. The collision integral is expressed through the electron self-energy $\Sigma(\mathbf{p},\epsilon)$ as

$$I=\int\frac{d\mathbf{p}}{(2\pi)^3}[G^A(\mathbf{p},\epsilon)-G^R(\mathbf{p},\epsilon)]\{[2n(\epsilon)-1]\times[\Sigma^A(\mathbf{p},\epsilon)-\Sigma^R(\mathbf{p},\epsilon)]-\Sigma^K(\mathbf{p},\epsilon)\}. \tag{4}$$

In the pure case, $Td, Tl>u_l,u_t$ (l is the electron mean free path, u_l and u_t are velocities of longitudinal and transverse phonons), we can can neglect the contribution of the interference diagrams, which consist of vertices of inelastic and elastic scattering.[15] The collision integral based on the self-energy diagram with two vertices $\gamma(\mathbf{k},\mathbf{q},n)$ is

$$I(\epsilon)=\frac{8}{\pi\nu}\int\frac{d\mathbf{p}d\mathbf{q}d\omega}{(2\pi)^7}\gamma^2\mathrm{Im}\,G^A(\mathbf{p},\epsilon)\mathrm{Im}\,G^A(\mathbf{p}+\mathbf{q},\epsilon+\omega)\times\mathrm{Im}\,D^R(\mathbf{q},\omega)R(\epsilon,\omega,\theta,T), \tag{5}$$

$$R(\epsilon,\omega,\theta,T)=N(\omega,T)n(\epsilon,\theta)[1-n(\epsilon+\omega,\theta)]-[1+N(\omega,T)][1-n(\epsilon,\theta)]n(\epsilon+\omega,\theta), \tag{6}$$

$$n=[\exp(\epsilon/\theta)+1]^{-1},\quad N=[\exp(\omega/T)-1]^{-1}. \tag{7}$$

As in the case of electron-impurity scattering one can express the scattering potential V_{e-bd} in terms of the corresponding electron momentum relaxation time or the electron mean free path, which is equal to $2d$ in the case of the diffusion scattering at the interface. Therefore, the collision integral $I(\epsilon)$ is proportional to d^{-1}, and the heat flux from electrons to phonons due to inelastic electron-boundary scattering per unit square of the film is independent on d. It may be calculated as

$$\tilde{I}_h=\int d\epsilon\epsilon\nu(\epsilon)dI(\epsilon,\theta,T). \tag{8}$$

Assuming that one half of the phonons is emitted into the substrate, and calculating the integral in Eq. (8) we find the heat flux from electrons to substrate phonons,

$$I_h=\frac{\pi^4}{40}\frac{\nu(0)}{p_F^2}\left(\frac{\beta_l}{u_l}+\frac{2\beta_t}{u_t}\right)\left[\theta^4J_1\left(\frac{\theta}{T_D}\right)-T^4J_1\left(\frac{T}{T_D}\right)\right], \tag{9}$$

where $J_1(y)=(15/\pi^4)\int_0^{1/y}dx\ x^3N(x)$; β_l and β_t are the constants of interaction with longitudinal and transverse phonons: $\beta_l=(2/3\epsilon_F)^2[\nu(0)/2MNu_l^2]$, where $\nu(0)=mp_F/\pi^2$ is the electron two-spin density of states; $\beta_t=\beta_l(u_l/u_t)^2$. These constants describe coupling of electrons with thermal phonons. In the ''jelly'' model the kinetic constant β_l may be presented in terms of the electron-phonon renormalization constant λ, as $\beta=\lambda(2p_F/q_D)^2$, where q_D is the Debye wave vector. In the low-temperature limit $T\ll T_D$ (T_D is the Debye temperature), the integral J_1 approaches 1.

It is important to note that due to inelastic boundary scattering, electrons interact with transverse phonons as well as with longitudinal phonons. The second term in parentheses in Eq. (9) corresponds to the interaction with transverse phonons. A typical value of the ratio u_l/u_t is 2–3, therefore the contribution of transverse phonons is approximately one order of magnitude larger than the contribution of longitudinal phonons.

The electronic Kapitza conductance obtained from Eq. (9) is given by

$$\sigma_K^e=\frac{\pi^4}{10}\frac{\nu(0)T^3}{p_F^2}\left(\frac{\beta_l}{u_l}+2\frac{\beta_t}{u_t}\right)J_2\left(\frac{T}{T_D}\right), \tag{10}$$

where $J_2(y)=-(15/4\pi^4)\int_0^{1/y}dx\ x^4(dN(x)/dx)$.

Ignoring the decrease in the electron-phonon coupling at the interface, one can express the conductance through the electron-phonon energy relaxation rate in a pure bulk metal,[2,16,17]

$$\frac{1}{\tau_{e-ph}}=\frac{7\pi\zeta(3)}{2}\frac{\beta_lT^3}{(p_Fu_l)^2}J_3\left(\frac{T}{T_D}\right), \tag{11}$$

where $J_3(y)=[2/7\zeta(3)]\int_0^{1/y}dx\ x^2(N(x)+n(x))$. In the low-temperature limit, $T<T_D$, we obtain

$$\sigma_K^{el}=\frac{3\pi\hbar}{35\zeta(3)k_B}\frac{\gamma u_l}{\tau_{e-ph}}\left[1+2\left(\frac{u_l}{u_t}\right)^3\right], \tag{12}$$

where γ is the Sommerfeld constant. In the general case, the right-hand side of Eq. (12) is multiplied by $J_2(T/T_D)/J_3(T/T_D)$.

It is instructive to compare the heat flux due to inelastic electron-boundary scattering [Eq. (9)] and the flux due to ''pure'' electron-phonon scattering. In the latter case the square of the matrix element is proportional to ω_q, that is $\sim T$. Because of the momentum conservation, the number of phonons participating in scattering is proportional to T^2. As a result, the heat flux is proportional to θ^5-T^5. For the inelastic boundary scattering the matrix element squared is proportional to $\omega_q^{-1}\sim T^{-1}$ [Eq. (3)], and all thermal phonons ($\propto T^3$) participate in scattering, because the momentum is not conserved in the system of electrons and phonons.

If other processes of the electron relaxation would be absent, according to Eq. (12) the electron relaxation rate due to inelastic electron-boundary scattering is given by

$$\frac{1}{\tau^*}=\frac{3\pi}{35\zeta(3)\tau_{e-ph}q_Td}\left[1+2\left(\frac{u_l}{u_t}\right)^3\right], \tag{13}$$

where $q_T = T/u_l$ is the wave vector of a thermal longitudinal phonon. The obtained electron relaxation rate corresponds to the electron energy relaxation rate due to inelastic electron-impurity scattering in an impure metal (see Refs. 2, 15, and 17), if one puts the electron mean free path to be of the order of the film thickness.

According to Eq. (13), as $2(u_l/u_t)^3 \sim 25$, the electron energy relaxation due to inelastic scattering from boundaries dominates over bulk relaxation due to the pure electron-phonon interaction in a wide region of parameters d and T (just as inelastic electron-impurity scattering dominates in impure films[17,18]). Thus we expect boundary scattering to play a significant role in the electron dephasing in ultrathin films and nanostructures.

Vibrating boundaries as well as vibrating impurities result in the interaction of electrons and transverse phonons. However, the Eliashberg function may be significantly changed near an interface. In pure films the Eliashberg function determined by vibrating boundaries may be found from temperature-dependent resistivity.

Recently, the interaction of electrons and transverse phonons has been studied in Au, Al, Nb, Be, and NbC films.[16–18] The resistance of all films contains a significant interference term $\delta\rho = BT^2\rho_0$ (ρ_0 is the residual resistivity), which prevails over the Bloch-Gruneisen term at low temperatures (up to 100 K in impure films). For every film the constant of interaction β_t was obtained from the coefficient B. Then values of β_t were used to calculate a contribution of transverse phonons to the electron dephasing rate. The calculations are in good agreement with experimental data. This shows that in films with a short electron mean free path the interaction of electrons with transverse phonons controls the relaxation rate over a broad temperature range. Electron-boundary scattering in pure thin films plays the same role as electron-impurity scattering in impure films. Probably the effects of inelastic electron scattering from boundaries have been observed in Au films, where the electron mean free path was estimated to be equal to a film thickness.[16,17]

Taking into account that the electron interaction with transverse phonons due to vibrating boundaries prevails over analogous interaction with longitudinal phonons, we can express the Kapitza conductance [Eq. (10)] in terms of the measured coefficient B,

$$\sigma_K^e T^{-3} = \frac{\pi^2}{5}\frac{\beta_t}{v_F u_t} = \frac{3}{40} B v_F \nu(0). \tag{14}$$

The last equation allows a quantitative prediction of the electronic Kapitza conductance from the T^2 term in resistivity of pure films.

III. COMPARISON WITH EXPERIMENTS

Here we consider some experimental data in the light of the proposed model. Let us first directly estimate a value of the electronic Kapitza conductance. For Al, Au, Be, and Nb films we take values of the coefficient β_t, determined from resistivity measurements in Refs. 16 and 17. Parameters of materials are presented in Table I (see Ref. 17). For comparison we also present values of β_t^{th}, calculated from available parameters in Ref. 18. Values of σ_K^e for the boundaries of Al, Au, Be, and Nb films and dielectric substrates with analogous acoustic properties are calculated by Eq. (14).

TABLE I. Parameters of the metals and the electronic Kapitza conductance.

Metal	β_t	β_t^{th}	v_F 10^7 cm/s	u_t 10^5 cm/s	$\sigma_K^e T^{-3}$ W/cm^2 K^4
Al	4.7	3.0	13	3.1	0.03
Au	1.4	0.6	14	1.2	0.02
Be	4.3	0.2	22	9	0.006
Nb	10	6.6	2.7	1.7	0.6

As one can see, the electronic Kapitza conductance may vary over a wide range. The measured total conductance is sensitive to a method of sample preparation, but for most of the metal-insulator contacts, $\sigma_K^e T^{-3}$ ranges from 0.1 to 0.01 W/cm^2 K^4.[4] Therefore, we expect a very significant effect of electrons for Nb contacts.

In many measurements the conductance was observed to decrease when a metal was driven superconducting.[4] In the frame of the proposed model a simple explanation of the effect is that superconducting electrons are not available to transport heat. We consider the conductance at an indium-sapphire interface. Samples prepared by different methods were studied in Refs. 19–21. When in magnetic field, indium is in the normal state, a value of the conductance at 1 K was found to range from 0.01 to 0.07 W/cm^2 K. A maximal decrease during the normal-superconducting transition $\delta\sigma_K = 0.011$ W/cm^2 (30% of σ_K in the normal state) was observed for a rough interface.[21] For an optically polished interface, $\delta\sigma_K(1\text{ K})$ was found to be 0.008 W/cm^2 K.

To evaluate σ_K^e in In, we used the following parameters: $\gamma = 1.69$ mJ/mol K^2,[22] and $u_l = 2.7\times10^5$ cm/s, $u_t = 0.9\times10^5$cm/s.[23] The measured value of $\tau_{e-ph}(T_c = 3.4\text{ K})$ is 0.8–2 ns (see Ref. 24). Then using Eq. (12), we find the electronic Kapitza conductance at $T = 1$ K is 0.01–0.03 W/cm^2 K. In the present theory, the value of σ_K^e is two to three times larger than the measured $\delta\sigma_K$, because we do not take into account a decrease in coupling near the interface. We also expect that diffusion motion of electrons near a rough interface makes the interaction time longer and enhances the coupling.

At nitrogen temperatures the electron energy relaxation time τ_{e-ph} in metals is determined from the fs thermomodulation measurements. For most metals a value of the relaxation time was found to be near 1 ps.[25,26] The value of the electronic Kapitza conductance [Eq. (12)] is evaluated as 2000–6000 W/cm^2 K. That is in good agreement with the measured Kapitza conductance for a pair of materials with rather different acoustic impedances, like metal and diamond.[4,5] To be more specific, we consider a gold-diamond interface. The Kapitza conductance was measured to be 2000 W/cm^2 K at 100 K, which is one order of magnitude larger than the result of the acoustic mismatch theory.[5] Taking $\gamma = 0.73$ mJ/mol K^2,[22] $u_l = 3.2\times10^5$ cm/s, $u_t = 1.2\times10^5$ cm/s,[16] and the value of τ_{e-ph} (100 K) is 0.5 ps,[25] we obtain from Eq. (12) that σ_K^e is equal to 2800 W/cm^2 K.[27]

The inelastic electron-boundary scattering plays a significant role also in nonequilibrium phenomena in thin films of

high-T_c superconductors, where a complicated many-component photoresponse is observed.[2,6,8,28] Up to now two components of transient photoresponse are well identified. The fast nonequilibrium component has a decay time of ~ 1 ps that corresponds to the electron energy relaxation time due to the electron interaction with longitudinal phonons. The bolometric component has a ns decay time, which is proportional to the film thickness. The new component was found in Ref. 10, where the transient photoresponse of ultrathin YBaCuO films to laser pulsed radiation with pulse duration of 500 fs and 40 ps was measured. The observed ps decay time of 40 ps for 15-nm-thick film is too short to be attributed to the bolometric component. But the relaxation time found in Ref. 10 turned out to be proportional to the film thickness and, therefore, it cannot be assigned to the bulk electron-phonon relaxation.

We suggest the following interpretation of the new photoresponse component. Since the electron temperature in the resistive photoresponse is near the transition temperature (70–90 K in Ref. 10), while the operating temperature (T_0) was 25 K, the correlation signal was measured in strong nonequilibrium conditions. In the discussed time scale, electrons interact with part of the phonon modes, and the heat capacity of these modes is significantly smaller than the total phonon heat capacity. In the framework of the Debye model, these phonon modes are associated with longitudinal phonons; thus, 1 ps after the pulse electrons have already reached equilibrium with longitudinal phonons of the film. The decay of the new component is determined by cooling of electrons and longitudinal phonons together due to inelastic electron-boundary scattering. The corresponding energy balance equation is

$$(C_e + C_{l,ph})\,\frac{\partial \theta}{\partial t} = -\sigma_K^e(\theta - T_0), \tag{15}$$

where θ is the temperature of electrons and longitudinal phonons in the film, T_0 is the substrate temperature, and $C_{l.ph}$ is the specific heat capacity of longitudinal phonons. It is convenient to present σ_K^e as C_e/τ^* [Eq. (13)]. Then the decay time of this photoresponse component is

$$\tau = \frac{35\zeta(3)}{3\pi}\,\frac{1 + C_{l,ph}/C_e}{1 + 2(u_l/u_t)^3}\,q_T d\,\tau_{e-ph}. \tag{16}$$

To evaluate the decay time for YBaCuO film we take the ratio u_l/u_t equal to 2 (this coefficient is approximately the same for YBaCuO films and substrates[8,29]), then $C_{t,ph}/C_{l,ph} = 2(u_l/u_t)^3$ is equal to 16 (where $C_{t,ph}$ is the electron heat capacity of transverse phonons). The ratio of phonon and electron heat capacities at the transition temperature is 41,[28] and therefore, the ratio of $C_{l,ph}/C_e$ is 2.4. For the film with thickness 15 nm, the parameter $q_T d$ is ~ 50. Substituting $\tau_{e-ph} = 1$ ps (Refs. 2 and 28) into Eq. (16), we get a value of τ is 45 ps, which is in agreement with measured decay time of 40 ps.[10]

In conclusion, we calculated the electronic Kapitza conductance due to inelastic electron-boundary scattering [Eq. (12)]. This mechanism is very important for many applications of ultrathin films and nanostructures.

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