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Phonon trapping in thin metal films

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We describe a simple electron-heating experiment in which the phonon escape from thin metal films is changed *in situ*. This provides clear evidence that the phonon-escape rate can significantly affect electron heating. From the results we have determined that phonon generation by the heated electrons and phonon escape have comparable rates.

The nonequilibrium properties of metallic microstructures have been of considerable recent interest.^{1,2} Experiments to study these properties have used electric fields to heat the electron system above the ambient temperature of the lattice. With this technique it has been demonstrated that the electron and phonon temperatures can be made very different. This has raised questions about the appropriate temperature to be used to describe, for instance, the electron-phonon interaction in the nonequilibrium situation.³ The additional complications of nonthermal distributions of electrons and phonons have also been discussed.^{4,5} In the analysis of the nonequilibrium heating experiments, one approximation has been consistently applied: the phonons have always been considered to escape from the microstructure in a time less than the electron-phonon relaxation time. Within this approximation the lattice temperature of the microstructure remains the same as that of the substrate on which it is supported. Electron-heating experiments using different substrate materials have indicated that this is not necessarily a valid approximation.² In this Rapid Communication we describe an experiment in which the phononescape time from a thin metallic film to the surroundings is changed in situ. The results show very clearly that the phonon occupations in the film and the substrate can be very different.

The samples used for the experiments were 30-nm-thick Au₆₀Pd₄₀ alloy films that were thermally evaporated onto a silicon substrate at room temperature and at a pressure of 5×10^{-6} Torr. The typical sheet resistance of the films was $R_{\Box}=10 \ \Omega$. The sample geometry was defined by electron-beam lithography⁶ and was designed for true four terminal resistance measurements. The active sample region was 1 μ m wide and 10 μ m long. Some of the samples were made free standing by removing the silicon beneath the active region using a CF₄-O₂ reactive ion etch for approximately 40 s. The experiments were performed with the samples mounted inside a sealed copper chamber that was thermally attached to a temperature controlled bath of helium. The sample mounting arrangement provided good thermal contact between the substrate and the chamber walls. The chamber could either be evacuated or filled with liquid helium so that the exposed surface area of the films was either in a vacuum or liquid helium. The process of changing the conditions from vacuum to helium was carried out at low temperature so that the phonon boundary conditions could be modified without cycling the sample to room temperature.

Four terminal resistance measurements were made over the temperature range 1-4 K using a low-frequency ac bridge. To provide electron heating a dc electric field of up to 6.5 V/cm was applied across the sample. The ac sensing current was always kept low enough that its contribution to the heating remained very small.

Under equilibrium conditions we observed a temperature dependent resistance which had a minimum R_{0eq} , at a temperature $T_0 = 7$ K. Below T_0 the resistance increased logarithmically with the substrate temperature and was consistent with the resistance corrections reported for similar alloy films by other workers.^{7,8} We found no change in the equilibrium temperature dependence of the resistance with the sample chamber evacuated or filled with liquid helium.

Figure 1 shows the striking difference between the response of the resistance $\Delta R/R_{0eq}$ to an applied dc field when the film surface was in a vacuum and when it was in liquid helium. The electron heating by the applied electric field appears to be significantly reduced when the free-surface area of the sample is covered with helium. We believe this simple result provides strong evidence that the phonon-escape time from the film cannot be neglected in electron-heating experiments.

The change in resistance $\Delta R/R_{0eq}$ as a function of substrate temperature T_s for several electric fields is shown in Figs. 2(a) and 2(b) for vacuum and helium, respectively. The data shown in Fig. 1 are the set of data points at $T_s = 1.5$ K. Comparing the data in Fig. 2(a) and 2(b), the reduction in the resistance change with applied field when

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FIG. 1. The change in resistance with electric field of a supported film in vacuum (\bullet) and covered with liquid helium (\blacksquare). $T_s = 1.5$ K.

helium is present is seen to occur over the whole temperature range studied. As additional evidence of the importance of the phonon-escape mechanism, we observe a distinct change in $\Delta R/R_{0eq}$ as the helium temperature passes through the λ point [Fig. 2(b)]. This is also observed in the data for the free-standing film shown in Fig. 3. The phenomenon responsible for this change is uncertain at present but we suggest it may be associated with either



FIG. 2. (a) The change in resistance of a supported film in vacuum as a function of the substrate temperature. The fields applied were 0.15, 0.64, 1.28, 1.91, 2.55, 3.19, 3.83, 4.47, 5.11, 5.74, and 6.38 V cm⁻¹ (top to bottom). (b) The change in resistance of a supported film covered in helium as a function of the substrate temperature. The fields applied were 0.15, 0.64, 1.28, 1.91, 2.55, 3.19, 3.83, 4.47, and 5.11 V cm⁻¹ (top to bottom).



FIG. 3. The change in resistance of a free-standing film surrounded by liquid helium as a function of the helium temperature. The fields applied were 0.26, 0.49, 0.98, 1.47, 1.96, 2.45, 2.93, and 3.42 V cm⁻¹ (top to bottom).

the changed excitation spectrum or the changed thermal conductivity of the helium. However, crossing the λ point is only expected to change the phonon-loss mechanism from the sample to the helium which confirms the sensitivity of electron-heating experiments to the phonon-escape time.

To discuss these results in more detail we will consider first the data shown in Fig. 1. The straightforward rate equation 9

$$\frac{dn(\omega)}{dt} = \frac{n(\omega, T_e) - n(\omega)}{\tau_{e-\text{ph}}} + \frac{n(\omega, T_s) - n(\omega)}{\tau_{esc}}$$

can be used to describe the relaxation of energy from the electron system to the phonon system in the film and subsequently to the substrate. $n(\omega)$ is the nonequilibrium phonon distribution and the Bose distributions of the phonons at the electron temperature T_e and the substrate temperature are $n(\omega, T_e)$ and $n(\omega, T_s)$, respectively. For moderate electric fields, two characteristic times are required to determine the relaxation of energy; the phononescape time from the film to the substrate τ_{esc} , and the electron-phonon relaxation time τ_{e-ph} , taken to be of the Pippard form.¹⁰ Under the steady-state conditions of the present experiments, the power per volume being transferred from the film to the substrate is

$$\frac{E^2}{\rho} = \int D(\omega) \hbar \omega \frac{n(\omega, T_e) - n(\omega, T_s)}{\tau_{e-ph} + \tau_{esc}} d\omega$$

where E is the electric field applied to the film, ρ is the resistivity, and $D(\omega)$ the phonon density of states. From this equation it is clear that the two characteristic times act in series and form a bottleneck in which the longest dominates the overall relaxation of energy from the film. Since it seems unlikely that the electrons in the film couple directly to the helium, the process of allowing helium into the chamber can be considered to introduce a loss path for phonons that is in addition to the phonon escape into the substrate. The two routes for phonon loss will act in parallel with the combined effect of reducing the phonon-escape time from the film. If, as is usually assumed, $\tau_{esc} \ll \tau_{e-ph}$, the energy relaxation bottleneck would be dominated by τ_{e-ph} and the reduction of the phonon escape

cape time by the additional helium loss path would be expected to have a negligible effect on the heating data. This is obviously not the case and shows the importance of the phonon escape in electron-heating experiments. Our data therefore imply that τ_{esc} is at least comparable to τ_{e-ph} . Under these conditions, as the electrons are heated by the field we expect $T_e > T_p > T_s$ where, for succinctness, we have used an effective temperature T_p for the phonons in the film even though they are not in equilibrium. A full description using the nonequilibrium phonon distribution will be given elsewhere.

An interesting feature, shown in both Figs. 2(a) and 2(b), is that in the presence of the electric field the steady-state resistance of the films can be reduced below R_{0eq} . This effect has been observed previously but in the present experiments we have found that the amount by which the resistance falls below R_{0eq} depends critically on the rate of phonon escape from the samples, and thus on the phonon occupation within the film. Liu et al.¹ explained the lowering of the resistance below R_{0eq} by considering the sum of three effects to describe the overall temperature dependence of the resistance: a Drude term that includes the electron-phonon inelastic lifetime, $\tau_{e,\text{ph}}^{i}$ ¹¹ electron-electron interactions; and weak localization. In equilibrium, at temperatures well above T_0 , the Drude term is dominant and since $\tau_{e-ph}^i \propto T^{-\beta}$ this gives a power-law dependence $R \propto T^{\beta}$. In the present experiments we find $\beta = 3$. At temperatures well below T_0 , the Drude term becomes smaller than the other terms which provide the observed logarithmic resistance rise. Therefore, the combination of the three terms gives a resistance minimum under equilibrium conditions. When the system is in the steady state, the relative contribution of each term is changed because they each have a different dependence on T_e and T_p . In particular, τ_{e-ph}^i becomes a function of T_e and T_p .³ Consequently, since changes of τ_{esc} affect both T_e and T_p , τ_{esc} will modify τ_{e-ph}^i in the steady state. In order to simplify the situation we follow the argument presented by Liu et al.,¹ that for small temperature differences between the electrons and the phonons, the effective temperature describing the electron-phonon interaction remains close to the phonon temperature. Therefore, for the present discussion, we will suppose that in the steady-state τ_{e-ph}^{i} is only a function of T_{p} . The drop of the resistance below R_{0eq} is now easily explained by two

competing processes: A net logarithmic contribution providing a decreasing $\Delta R/R$ with T_e due to electron localization and electron-electron interactions, and a Drude contribution providing an increasing $\Delta R/R$ with T_p . Clearly, the minimum in the nonequilibrium situation depends on the relative values of T_e and T_p which in turn depend on τ_{esc} . Therefore, if phonons are trapped in the film the resistance can still drop below R_{0eq} , but at high enough electric fields T_p will increase and the Drude term will dominate over the logarithmic terms. The data in Fig. 2 are decreasing with electric field, even at the highest fields, which indicates that even though the Drude term is increasing, it is not the dominant term at the fields applied.

Another interesting effect shown in Fig. 2(a) is the positive slope of the high-field data with T_s . Previous electron-heating measurements have yielded data that is independent of the substrate temperature at high electric fields. 12-14 In such cases the explanation has been that the resistance is a function of T_e and that T_e becomes independent of the substrate temperature above some critical value of electric field. We suggest that the cause of the positive slope in our data is the temperature dependence of the Drude contribution via τ_{e-ph}^{i} . At each higher substrate temperature, the Drude contribution is somewhat larger which will result in a smaller negative $\Delta R/R$ for a given electric field. The relative insensitivity of the data to T_s above the lambda point [Figs. 2(b) and 3] suggests that the temperature dependence arises predominantly from T_p , that is, phonons trapped in the microstructure.

In conclusion, using a very simple experiment we have clearly demonstrated that electron-heating experiments are affected by the process of phonon escape from a metal structure. This indicates that the usual assumption of rapid phonon escape from films and wires is not always valid and needs careful consideration for each metalsubstrate combination. The breakdown of this assumption also leads to a nonequilibrium phonon distribution within the film which must be taken into account in a complete analysis of the problem. This will be discussed in a subsequent paper.

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