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Crossover between Electron-Phonon and Boundary-Resistance Limits to Thermal Relaxation in Copper Films

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We observe a crossover from electron-phonon (e-ph) coupling limited energy relaxation to that governed by thermal boundary resistance (phonon-phonon coupling, ph-ph) in copper films at subkelvin temperatures. Our measurement yields a quantitative picture of heat currents, in terms of temperature dependences and magnitudes, in both e-ph and pp limited regimes, respectively. We show that by adding a third layer in between the copper film and the substrate, the thermal boundary resistance is increased fourfold, consistent with an assumed series connection of thermal resistances.

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I. INTRODUCTION

Investigation of energy relaxation of electrons in normal metal films is important for understanding the underlying physics as well as for applications [1–4]. Especially for mesoscopic devices at low temperature, where the dominant thermal wavelength \( \lambda \) is comparable to the device dimension, phonons in the films could be two dimensional (2D), and it has been shown both experimentally and theoretically that the reduced phonon dimension does affect the energy relaxation of electrons in thin films [5–8]. Heat transport by phonons, electrons, and photons has been studied experimentally in mesoscopic devices [9–14]. From the application point of view, a good understanding of energy relaxation in metal films is important, e.g., for calorimetry and bolometry [15]. Decreasing the heat conductance from the metal film absorber to the environment enhances the energy resolution, but on the other hand, it makes the device slower. For a transition-edge sensor, unaccounted-for thermal boundaries can affect the noise and energy resolution [16]. Finally, for a normal-metal–insulator–superconductor (N-I-S) junction cooler, quick thermalization of the secondary electrode is favorable in order to increase the cooling efficiency [17].

In a heated normal metal film on a dielectric substrate, electrons within the film relax by electron-electron (ee) interactions, and the energy is dissipated to the environment mainly by electron-phonon (e-ph) coupling to the film phonons, which is characterized by e-ph thermal coupling resistance \( R_{\text{e-ph}} \). Film phonons are coupled to the substrate phonons, which are usually considered to constitute the heat bath for the device, by phononic coupling. The corresponding thermal resistance between phonons in the film and the substrate is the thermal boundary resistance \( R_{\text{ph-ph}} \). If the ee interactions are assumed to be much faster than other processes, the energy relaxation of the electrons in the film is determined by \( R_{\text{e-ph}} \) and \( R_{\text{ph-ph}} \), with the weaker of the two governing the energy relaxation process. For thin films at low temperatures, e-ph coupling strength is weak and it becomes the bottleneck of the energy relaxation. With increasing temperature or film thickness, the e-ph coupling gets relatively stronger and the heat transport across the boundary between the film and the substrate becomes the limitation for the energy relaxation.

Electron-phonon coupling in metal films at low temperatures has been actively studied during the last decades. In particular, the effect of disorder and phonon dimensionality on the e-ph coupling strength have been intensively discussed [7,18–20]. Thermal boundary resistance between metals and dielectric substrates has also been well investigated. Experimental observations can be explained with either the acoustic mismatch model (AMM) or diffuse mismatch model (DMM) [21]. AMM describes phonon heat transfer through a flat interface between perfect crystals. In analogy to Snell’s law for the electromagnetic waves, only the phonons with the incident angles below the critical one are transmitted through the interface. The critical angle is determined by the acoustic properties of the materials on both sides of the boundary. DMM assumes diffusive phonon scattering at the interface and, hence, the phonon transmission probability depends only on the phonon densities of the states and sound velocities on both sides. For

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identical acoustic properties of the two sides, thermal resistance predicted by DMM is about two times larger than that by AMM, as according to AMM, transmission probability is unity, and for DMM transmission probability, it is 50%. For a smaller acoustic difference of the two materials, the resistance differs by less than 30% [21]. In the case of solid-solid boundaries, the mismatch in sound velocities and phonon mode densities is usually small, and the two models give similar predictions.

Here, we present the experimental results showing the crossover between e-ph and boundary-resistance limited thermal relaxation in Cu films at subkelvin temperatures. For Cu film 50 nm in thickness, we find the energy relaxation to be limited by e-ph coupling in the full temperature range explored. By increasing the film thickness to 300 nm, the thermal boundary resistance limits the energy relaxation and we are able to quantify the heat transport between the metal-substrate interface directly from the experiments. By adding a third thin layer of film between the Cu film and the substrate, the thermal boundary resistance is increased fourfold, consistent with the assumption of a series connection of the thermal boundary resistances.

For a heated metal film on a substrate, the energy flow is shown in the thermal model in Fig. 1(a). Within the film, the energy flow rate from electrons to phonons is described by

$$P_{e-ph} = \Sigma V(T_e^n - T_p^n).$$

(1)

Here, $T_e$ and $T_p$ are the electron and phonon temperatures in the film, $V$ is the metal volume, $n = 5$ for three-dimensional (3D) clean normal metals, and $\Sigma$ is the material-specific e-ph coupling constant [22]. The coupling between film phonons and the substrate phonons for a 3D system is characterized by

$$P_{ph-ph} = kA(T_p^n - T_s^n),$$

(2)

where $T_s$ is the substrate phonon temperature, $A$ is the contact area, and $k$ is the interface-material-dependent constant, which can be calculated with DMM as

$$k = \frac{\pi^2 k_B^4}{120 \hbar^3} \left( \frac{1}{c_{2L}^4} + \frac{2}{c_{2T}^4} \right) \left( \frac{1}{c_{1L}^2} + \frac{2}{c_{1T}^2} + \frac{2}{c_{1L}^2 + c_{1T}^2} \right).$$

(3)

Here, $c_{1L}$ and $c_{1T}$ are the speed of longitudinal and transverse sound on the side $x$ of the interface. For small temperature differences, the e-ph thermal coupling resistance is expressed as $R_{e-ph} = 1/5\Sigma VT^4$ and the thermal boundary resistance as $R_{ph-ph} = 1/4kA T^3$. $T_0$ equals the bath temperature of the refrigerator $T_0$ due to the large substrate-bath contact area.

**II. JOSEPHSON JUNCTION THERMOMETER AND MEASUREMENT SETUP**

One of the devices used in the experiments is shown in Figs. 1(b) and 1(c) together with the measurement setup. Cu film (brown) is evaporated on the silicon substrate, with 300 nm silicon oxide on top, by electron beam evaporation. The chamber pressure is kept below $5 \times 10^{-7}$ mbar during the deposition. Before contacting the Cu film with superconducting Al (blue), Ar plasma milling is used to clean the Cu film surface in order to achieve good metal-to-metal contacts between copper and aluminum. The hybrid structures with short channel length behave as a proximity Josephson junction (JJ). Switching current $I_{sw}$ is defined as the bias current when the junction switches from the superconducting state to the resistive state, shown
in the IV curve in Fig. 1(d). The JJ switches back to the superconducting state at a biasing current well below $I_{sw}$, defined as retrapping current $I_r$. The hysteresis of the IV curve originates from the overheating of the electrons after switching to the resistive state. The bath temperature dependence of $I_{sw}$ at zero heating, i.e., in equilibrium, shown in Fig. 1(e), is used as the temperature calibration for the JJ thermometer [23]. The long horizontal Cu wire between the large Cu pad and JJ thermometer is used as the heater to elevate electron temperature in the Cu film. A large pad on top enables us to determine the size of the Cu film with good relative accuracy. Previous work has shown that for our devices with dimensions much smaller than the e-ph relaxation length, the whole film has uniform electron temperature with the power levels applied in our experiments [24]. We verify this by measuring electron temperature with another JJ thermometer located at another end of the Cu pad, shown later in Fig. 3(b). We current-bias the two heater contacts with opposite polarities. Figure 1(f) is the measured $I_{sw}$ as a function of $I_H$ for various bath temperatures from 60 to 340 mK in 20-mK steps from top to bottom. Decrease of $I_{sw}$ while increasing $|I_H|$ indicates heating of the Cu film. The symmetry of the dependence around zero heating suggests no heating current flows to the thermometer in this configuration.

III. RESULTS AND DISCUSSION

For electrons in the copper film, superconducting Al acts as a thermal insulator at sufficiently low temperatures below its critical temperature $T_c \sim 1$ K; the joule power applied $P$ to the film dissipates mainly by e-ph coupling. The ratio of the two series thermal resistances is

$$\gamma = \frac{R_{ph-ph}}{R_{e-ph}} = \frac{5 \Sigma T}{4k}.$$  (4)

Here, $t$ is the thickness of the Cu film. For a thin film at sufficiently low temperatures, we expect $R_{e-ph}$ to dominate over $R_{ph-ph}$, so we have the standard situation usually assumed for thin films, i.e., $P = P_{e-ph} = \Sigma V(T_c^0 - T_0^0)$. In Fig. 2, we plot the experimental results of a sample with 50-nm thin Cu film; a linear dependence versus $T_c^0 - T_0^0$ is clearly seen as expected. From the slope, we obtain the e-ph coupling constant $\Sigma = 2.1 \pm 0.1 \text{nWK}^{-1} \text{m}^{-3}$ with no temperature dependence within the measurement interval from 60 to 250 mK, as shown in the inset of Fig. 2. The measured value of $\Sigma$ is consistent with previous experiments on Cu films [25,26]. Thus, the experiment demonstrates that for the 50-nm Cu film at low temperature, the energy relaxation of electrons is dominated by the e-ph coupling, and the exponent $n = 5$ is consistent with the theory based on the 3D free electron model [22].

Equation (4) suggests that if one changes the film thickness or temperature to the point where $R_{ph-ph}$ becomes equal to $R_{e-ph}$, a crossover from one energy relaxation mechanism to another should take place. The crossover temperature $T_{cr}$ depends on the constants $\Sigma$ and $k$ as $T_{cr} = 4k/5\Sigma t$. For perfect contacts between Cu and the silicon substrate, one finds $k \approx 310 \text{WK}^{-1} \text{m}^{-2}$, calculated with Eq. (3). Hence, the crossover temperature of 0.1 K is expected in films with the thickness of $t \approx 1.2 \mu m$. Recent experiments have suggested that for films evaporated on a silicon substrate, $k$ is smaller than that predicted for perfect contacts [27,28], which makes it possible to observe $T_{cr} \approx 0.1$ K in somewhat thinner films.

In Fig. 3(a), we show the SEM image of a sample with $t = 300$ nm Cu film. Firstly, we deposit 50-nm Cu film (brown) used in the JJ thermometers, heater, and contact pads. Then, we deposit the 300-nm Cu film (purple). Before contacting the two copper films, Ar plasma milling is used to clean the surface of the thin one. The inset of Fig. 3(b) shows the thick film covering the thin film. Electron temperature is measured with two JJ thermometers located at the two ends of the thick Cu film (local, remote) with a distance of 40 $\mu$m to check the uniformity of electron temperature in the thick Cu film while heating. Figure 3(b) shows that the two thermometers show identical temperatures except at the largest applied powers. The small difference at high $P$ originates most likely from the electron diffusion along with the thick Cu film and is negligible for the analysis. The data also suggest that the thermal boundary resistance between the two Cu films is negligible.

We plot $T_c^n - T_0^n$ as a function of $P$ in Fig. 3(c). In contrast to what was seen in Fig. 2, a linear dependence is observed when setting $n = 4$ in the full temperature range and three different bath temperatures explored. For
Comparison, we also show clearly nonlinear dependence for \( n = 5 \) and for the bath temperature 55 mK with the blue dots. From the linear fit of \( n = 4 \) data, we extract the constant \( k \) as a function of temperature, which is shown in the inset of Fig. 3(c). We find \( k \) to be about 60 WK\(^{-1}\)m\(^{-2}\) with a slight increase at high temperatures. The origin of this increase is unclear. The obtained value of \( k \) is consistent with the previous experiments on evaporated metal films [27,28], but it is smaller than the predictions of both AMM and DMM models. This difference may be explained by imperfect interface quality between the Cu film and the substrate.

Previous studies showed that for disordered normal metal films, the exponent \( n \) deviates from 5 depending on the type of disorder [19,29–32]. The observed \( n = 5 \) in 50 nm Cu film indicates the clean limit for the Cu film. Increasing the film thickness reduces the disorder and makes it closer to the 3D clean limit. So, the observed \( n = 4 \) for 300-nm Cu film is not to be ascribed to the film disorder. Instead, it originates from the fact that \( R_{\text{ph-ph}} \) dominates over \( R_{\text{e-ph}} \) for thick film and becomes the bottleneck for heat transport.

As the acoustic mismatch between different materials reduces the phonon transmission, an enhancement of the thermal boundary resistance is expected when adding a third layer of material between the Cu film and the substrate. We fabricate a sample with 3 nm of Ti added between 50-nm Cu film and the substrate. By simply considering a series connection of the two interface resistances [33,34], we find \( k_{\text{Cu-Ti-SiO}_2} = k_{\text{Cu-Ti}}^{-1} + k_{\text{SiO}_2}^{-1} \), where \( k_{\text{Cu-Ti}} \) and \( k_{\text{SiO}_2} \) are the interface-dependent parameters between Cu/Ti and Ti/SiO\(_2\), respectively. In this way, from the DMM model, we estimate \( k_{\text{Cu-Ti-SiO}_2} = 0.49 k_{\text{Cu-SiO}_2} \), where \( k_{\text{Cu-SiO}_2} \) characterizes the Cu/SiO\(_2\) boundary with its measured value shown in the inset of Fig. 3(c).

In Fig. 4(a), we plot measured \( T_e^4 - T_0^4 \) as a function of \( P \). Linear dependence is observed at temperatures above 130 mK, suggesting that \( R_{\text{ph-ph}} \) dominates the energy relaxation in this temperature range. Deviation from the linearity when \( P \) is below around 1 pW, corresponding to electron temperature rise \( \Delta T_e = T_e - T_0 \) below 1–2 mK, may be due to the uncertainty of the electron temperature measurement. Its impact on final conclusion is negligible because of the small \( \Delta T_e \) and the narrow power range observed. From the linear fits, we estimate the constant \( k_{\text{Cu-Ti-SiO}_2} \) to be about 15 WK\(^{-1}\)m\(^{-2}\), which is 25% of the value measured without the intermediate layer. It is less

![FIG. 3. (a) False-color SEM image of a sample with large Cu film (purple) with dimensions 10 \( \mu \)m \( \times \) 40 \( \mu \)m \( \times \) 300 nm. Two JJ thermometers (local, remote) are located at the two ends of the Cu film to check the uniformity of the electron temperature while heating. The inset in (b) is the enlargement of the local JJ thermometer showing the large Cu film covering the thin Cu film (brown). (b) Measured electron temperature by the local and remote thermometer showing the large Cu film covering the thin Cu film while heating. The inset in (b) is the enlargement of the local JJJ thermometer (local, remote) are located at the two ends of the Cu film to check the uniformity of the electron temperature while heating. (c) Measured \( T_e^4 - T_0^4 \) plotted as a function of \( P \) with exponent \( n = 4 \) (triangles, \( T_0 = 55, 100, 150 \) mK, from dark blue to red) and \( n = 5 \) (blue dotted, \( T_0 = 55 \) mK). A linear dependence is observed when plotted with \( n = 4 \); black lines are the linear fits. Experimental data show that for the 300-nm Cu film, thermal boundary resistance limits the energy relaxation process. The derived interface-material-dependent parameter \( k \) is about 60 WK\(^{-1}\)m\(^{-2}\), shown in the inset.](https://example.com/fig3)

![FIG. 4. Measurement results of a sample with a 3-nm Ti layer added between the 50-nm Cu film and the substrate. (a) \( T_e^4 - T_0^4 \) as a function of heating power \( P \) with \( n = 4 \). The observed linear dependence above 130 mK indicates that the thermal boundary resistance limits the energy relaxation. The black line is a guide to the eye. Inset: Derived \( k \) as a function of temperature. (b) \( T_e^4 - T_0^4 \) as a function of heating power \( P \) with \( n = 5 \) at 60 mK. Linear dependence is observed only at temperatures up to about \( T_e = 120 \) mK. At higher temperatures, linear dependence is observed when plotted with \( n = 4 \), shown in (a) with the black dotted line. The red line is a guide to the eye.](https://example.com/fig4)
than 49% expected from the model discussed above. However, considering imperfect interface quality and crudeness of the model, which, for example, ignores the fact that the thermal phonon wavelength is much larger than the thickness of the Ti film, our result is in a good agreement with the theory. With the experimentally measured values of $k$ and $\Sigma$, we estimate the crossover temperature to be $T_{cr} = 124$ mK. At temperatures below $T_{cr}$, $R_{ph-ph}$ should dominate over $R_{ph-ph}$ and the linear dependence of $T_e^4 - T_0^4$ on $P$ is expected. In Fig. 4(b), we show the measurement results at the bath temperature of 60 mK. As expected, a linear dependence is observed at low temperatures, while for $T > T_{cr}$, deviations from it become visible. In contrast, if one plots $T_e^4 - T_0^4$ versus power, the linear dependence is observed at high temperatures $T > T_{cr}$, as shown in Fig. 4(a) with the black line. Thus, an additional 3-nm thin Ti layer between the Cu film and the substrate results in a fourfold increase in the thermal boundary resistance, which allows us to clearly see the crossover between the two energy relaxation mechanisms with changing temperature.

One of the open questions is what the influence of the dimensionality of the film is on the acoustic coupling strength [1,5,20]. It has been shown that even though the phonons in the film are 2D, the strong coupling of phonons in the film and the substrate can broaden its subband structure and make it closer to 3D. For Cu, the dominant phonon wavelength $\lambda$ is about 200 nm at 0.2 K when transverse phonons are considered, and it increases as $\propto T^{-1}$ when lowering the temperature. Assuming weak acoustic coupling and phonons in the film to be 2D, a reduction of the exponent $n$ from 5 is expected [6,8]. The observed $n = 5$ for the 50-nm Cu film suggests phonons in the film are closer to 3D than 2D, though $\lambda$ is much larger than the film thickness. The experimentally observed weaker acoustic coupling strength than what the theory predicts is not significant in making the phonons in the film 2D. Investigations are needed to quantify the effect of the strength of the coupling on the phonon dimensionality.

**IV. CONCLUSION**

In conclusion, we experimentally observe the crossover between the limiting energy relaxation mechanisms in copper films by changing the film thickness and temperature. We demonstrate that an additional Ti layer between the Cu film and the substrate enhances the thermal boundary resistance of the interface fourfold. This result may be useful for hot-electron calorimetry and bolometry since it can help in improving the energy resolution of the detectors [15]. Our experimental results further advance the understanding of energy relaxation mechanisms in mesoscopic devices and of the heat transport through the solid-solid interfaces at low temperatures.

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