Hot-electron effect in palladium thin films

Andrea Vinante,[1,2,*] Paolo Falferi,[1,2] Renato Mezzena,[2,3] and Michael Mück[4]
[1]*Istituto di Fotonica e Nanotecnologie, CNR-ITC, Povo, I-38050 Trento, Italy*
[2]*INFN, Gruppo Collegato di Trento, Sezione di Padova, Povo, I-38050 Trento, Italy*
[3]*Dipartimento di Fisica, Università di Trento, Povo, I-38050 Trento, Italy*
[4]*Institute of Applied Physics, University of Giessen, D-35392 Giessen, Germany*
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We have measured the hot-electron effect in sputtered palladium thin films at ultracryogenic temperatures. The temperature of the Joule-heated electron gas has been measured using a SQUID-based noise thermometer. Results are consistent with a T^3 dependence of the electron-phonon scattering rate, as reported for other materials. This dependence is in contrast with some theoretical predictions for the electron-phonon scattering rate in metals in the dirty limit and with recent experimental results on other metals.

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I. INTRODUCTION

It is well known that the conduction electrons in a metal at low temperature become very weakly coupled to the lattice phonons. The so-called hot-electron effect arises when power is directly dissipated in the electrons, leading to an overheating of the electron subsystem with respect to the phonon subsystem. This effect plays an important role in several applications, such as the dc superconducting quantum interference device (dc SQUID), where it sets an upper limit to the energy resolution,[1] or the normal metal hot-electron bolometer (NHEB), where the electron gas is used as thermal sensing element.[2]

Most experimental results on the hot-electron effect are qualitatively explained by a simple model of electron-phonon (*e*-*p*) interaction, based on a clean three-dimensional (3D) free-electron model.[1,3] It leads to an *e*-*p* scattering rate proportional to T^3, which has been indeed observed in several systems using different techniques.[1,4,5] However, a different temperature dependence is predicted by more specific models. In particular, in the dirty limit $ql<1$, where q is the wave vector of the dominant thermal phonons and l the electron mean free path, a scattering rate proportional either to T^2 or T^4 is predicted.[6] The T^4 dependence, which should be related with vibrating disorder, is only seldom observed. It has been conjectured that this dependence can be observed only in films made of pure noble metals in the dirty limit.[7]

In this paper, we report on the measurement of the *e*-*p* scattering rate in pure palladium films in the dirty limit. The measurement technique is similar to that described by Wellstood *et al.*[1] A constant power is dissipated in the film by Joule heating, and a SQUID-based noise thermometer directly measures the steady-state electron temperature. As pointed out by other authors,[4,7] techniques based on the measurement of the electron temperature provide direct information on the energy-loss rate of the hot electrons. In contrast, other techniques, based on weak localization effects, provide information on the electron dephasing rate, which may not be uniquely related with the *e*-*p* interaction. The *e*-*p* scattering rate inferred from our data is in good agreement with the classical T^3 dependence and is thus in contrast with the prediction for dirty metal films.

II. THEORETICAL MODEL

A standard model of the hot-electron effect in metallic thin films is described in several earlier articles.[1,3,5] We briefly summarize the main concepts. We assume that the phonon and the electron subsystems in the film are in thermal equilibrium at different temperatures, respectively, T_p and T_e. A constant power P dissipated in the electron gas will be transferred to the phonons through a thermal resistance R_{ep}, and finally to the thermal bath, at temperature T_0, through a thermal resistance R_K. The resistance R_K is usually thought to be a Kapitza contact resistance between the film and the liquid helium (if the sample is immersed in liquid helium) or between the film and the substrate (if the sample is in vacuum). In the latter case, additional thermal resistance could exist in the interface between the substrate and the chip holder or through the holder.

In the further assumptions of a clean 3D free-electron gas with spherical Fermi surface and a deformation-potential *e*-*p* interaction, the following relation between power P and film temperatures T_e and T_p is found:[3]

$$P=\Sigma\Omega(T_e^n-T_p^n), \tag{1}$$

where $n=5$, Ω is the volume of the film, and Σ is a material parameter given by

$$\Sigma=\frac{8\zeta(5)k_B^5E_F^2N(E_F)}{3\pi\hbar^4\rho v_F v_s^4}, \tag{2}$$

where ζ is the Riemann zeta function, ρ is the mass density, v_s the longitudinal sound velocity, E_F and v_F the Fermi energy and velocity, and $N(E_F)$ the density of electronic states per unit volume.

Other models predict the same qualitative behavior described by Eq. (1), but with different exponents n and prefactors Σ. In general, the exponent n is strictly related to the temperature dependence of the energy-relaxation rate $1/\tau_{ep}$. In particular, if the latter is given by $1/\tau_{ep}=\alpha T^p$, with α and p as constants, then the relation $n=p+2$ must hold.[5] Furthermore, if we introduce for convenience the electronic heat capacity per unit volume $C=\gamma T$, with γ constant, then the following expression for Σ can be derived:[5]

$$\Sigma = \frac{1}{p+2}\alpha\gamma. \tag{3}$$

The clean 3D model predicts a scattering rate $1/\tau_{ep} \propto T^3$, leading to $n=5$ in Eq. (1). In contrast, for a metal in the dirty limit[6] $ql < 1$, it is predicted that $1/\tau_{ep} \propto T^4 l$ or $1/\tau_{ep} \propto T^2/l$, leading to $n=6$ or $n=4$.

Experimentally, testing Eq. (1) requires measuring simultaneously both T_e and T_p. Instead, in our experiment the measured quantities are T_e and T_0. If the thermal resistance R_K is negligible with respect to the electron-phonon thermal resistance R_{ep}, we may approximate $T_p \simeq T_0$. This leads to the following approximation:

$$P = \Sigma\Omega(T_e^n - T_p^n) \cong \Sigma\Omega(T_e^n - T_0^n). \tag{4}$$

In the opposite case, when R_K is dominant, the overheating of the electrons with respect to the phonons is negligible and we approximate $T_p \simeq T_e$. Assuming a dominant thermal resistance $R_K = AT^{-m}$, with A and m constants, leads to the expressions

$$P = \frac{1}{A(m+1)}(T_p^{m+1} - T_0^{m+1}) \cong \frac{1}{A(m+1)}(T_e^{m+1} - T_0^{m+1}). \tag{5}$$

Equations (4) and (5) differ substantially only in the characteristic exponent. In particular, for a Kapitza thermal resistance ($m=3$) or typical thermal resistances of cryogenic insulating materials ($m \approx 1-2$), the expected exponent is $m+1 \approx 2-4$. Finally, if R_K and R_{ep} are comparable, one would expect a transition between the two regimes by changing the temperature.

III. EXPERIMENTAL DETAILS

Our measurements were done on palladium thin-film resistors which we normally use to shunt the Josephson junctions of thin-film niobium dc SQUIDs. A resistor consists of two sections, each one intended for shunting one Josephson junction. Each section has a length of 25 μm, a width of 15 μm, and a thickness of 25 nm. The palladium resistor was prepared on an oxidized silicon substrate by rf-magnetron sputtering in an argon atmosphere of 6 mTorr, with a sputtering rate of 12 nm/min. The resistor was connected to niobium traces on the substrate, to which thin aluminum wires were bonded for electrical contact.

The measured resistance at $T=4.18$ K is $R_s=8.72\ \Omega$, leading to a nominal resistivity $\rho=2.6\times10^{-7}\ \Omega$ m. The corresponding elastic mean free path is estimated as $l \approx 3.3$ nm using palladium electronic parameters reported in literature.[8] Thus, l is not substantially limited by the film thickness. The wave vector of the dominant thermal phonon $q=2k_BT/\hbar v_s$ is about 3×10^6 m^{-1} at the lowest operating bath temperature[9] ($T_0=59$ mK), leading to $ql \approx 0.01$.

We have also fabricated similar resistors with a large cooling fin attached to each section of them. The fins were also made of palladium, with a thickness of 100 nm. The area of the fin attached on the first section is 1.78 mm^2 and that of the other fin is 2.06 mm^2. Thus, the overall volume of

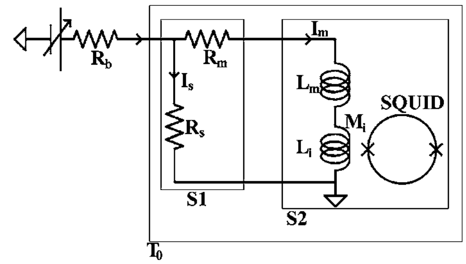

FIG. 1. Schematic of the measurement circuit. R_s and R_m are, respectively, the film resistor and the auxiliary SMD resistor and L_m is a filtering inductance. The readout SQUID, with input coil inductance L_i and mutual inductance M_i, measures the low-frequency current noise generated in the R_m-R_s loop. S1 and S2 are the superconducting niobium shields which contain, respectively, the resistors holder and the SQUID holder. Both shields are enclosed in a copper box at temperature T_0. A current $I=I_s+I_m$ can be imposed by means of a battery supply through a resistor R_b.

the cooling fins is 3.8×10^{-13} m^3, and that of the resistor alone is 1.9×10^{-17} m^3. The purpose of the fins is to increase the effective volume available for the *e-p* interaction, reducing the overheating due to the hot-electron effect.[1] Moreover, this allows us to check that the *e-p* interaction is really the dominant thermal resistance mechanism.

The measurement configuration is shown in Fig. 1. The film resistor is placed in parallel to a surface mount device (SMD) resistor $R_m=2.21\ \Omega$. The current noise of the R_m-R_s loop is measured by a commercial dc SQUID with input mutual inductance $M_i=10.71$ nH, operated in standard flux-locked loop configuration. Both resistors are mounted on a fiberglass holder, inserted in a niobium shield. The film resistor chip is attached to the holder by means of quick-set epoxy glue. The readout dc SQUID is inserted in a second niobium shield. The connection between resistors and SQUID is provided by a twisted pair of copper wire, of negligible resistance, placed in a lead tube. An inductance $L_m=15\ \mu$H, of negligible resistance, is placed in series with the SQUID input coil $L_i=1.6\ \mu$H to act as radiofrequency (rf) filter. Both SQUID and resistor holders are placed inside a copper box,[10] in good thermal contact with the mixing chamber of a dilution refrigerator. The box is filled with 1 atm of ^{4}He gas at room temperature to improve thermalization of the internal elements. The box temperature T_0 is monitored by a calibrated germanium resistor. The wiring from room temperature is progressively cooled by means of heat sink copper bobbins thermally anchored on the three main stages of the dilution refrigerator (1 K pot, still and mixing chamber).

The resistors R_s and R_m are biased in parallel with a constant current I from a battery supply through a room-temperature resistor $R_b=100$ kΩ. The total powers dissipated in the resistors R_s and R_m are calculated as $P_s=I^2R_sR_m^2/(R_m+R_s)^2$ and $P_m=I^2R_mR_s^2/(R_m+R_s)^2$.

The low-frequency spectral density of the current noise, measured by the SQUID, is given by

$$S_I = 4k_B\frac{T_sR_s + T_mR_m}{(R_s+R_m)^2} + S_{I0}, \tag{6}$$

where T_s and T_m are, respectively, the electron temperature of R_s and R_m. The first term in the right-hand side of Eq. (6)

represents the thermal noise of the resistors. S_{I0} represents the sum of all other noise terms, which include the thermal noise from R_b, the noise from the current supply, and the noise of the room-temperature SQUID electronics. All these terms are expected to be independent of temperature. We note that the intrinsic noise of the dc SQUID amplifier does depend on the bath temperature, but it is estimated to be much smaller than the noise from the resistors, and is thus negligible at the operating temperature. If S_{I0} and the R_m thermal noise are known, a measurement of the total noise S_I allows us to calculate the temperature of the electrons in the film resistor T_s. The circuit is thus configured as a noise thermometer.

For the calculation mentioned above, an accurate estimation of R_s and R_m is also required. We estimated the resistances at $T=4.18$ K by means of separate thermal noise measurements, for which the resistors were directly attached to the input coil of the SQUID. The resistances are then assumed to be independent of temperature down to 60 mK. In order to prove this assumption, we imposed a constant current I in the resistors at different temperatures (4.2 K, 600 mK, and 59 mK) and measured with the SQUID the fraction of current I_m flowing in the R_m branch. We found that the measured ratio $R_m/R_s=(I-I_m)/I_m$ is indeed independent of temperature within less than 0.5% in the explored temperature range. In principle, the values of R_m and R_s could change with temperature by the same relative amount, leaving their ratio unchanged. However, this eventuality is very unlikely, given the completely different nature of the two resistors. A weak increase of resistance of the palladium film with decreasing temperature is indeed predicted by electron localization theory,[8] but it has been estimated to be negligible for our case.

The noise measurements were performed in the frequency range of 500–1000 Hz, in which the frequency response of the flux-locked SQUID is flat and its $1/f$ noise is negligible. Moreover, the measurement frequency is well below the cutoff frequency $f_c \cong 100$ kHz of the RL circuit, formed by the series of the resistors R_s and R_m and the inductances L_m and L_i. A first set of noise measurements was performed as a function of temperature at zero bias current, as shown in Fig. 2, with the purpose of verifying the linear dependence of the noise on the thermodynamic temperature and calibrating the noise thermometer. The noise scales linearly with T as expected above 100 mK. The observed deviation from a linear behavior below 100 mK is attributed to residual power which overheats the film resistor even at zero bias current, as discussed in the next section. For this reason, the linear fit is restricted to the linear region above 100 mK. The constant term S_{I0} is then estimated as the intercept of a linear fit to the data.

The thermal noise of the SMD resistor R_m can be estimated, to a first approximation, by setting $T_m=T_0$, that is, assuming a negligible overheating of the SMD resistor. To check this hypothesis, we have performed separate tests, both in vacuum at $T=60$ mK and in a liquid helium bath at $T=1.2$ K, in which R_s was replaced with a SMD resistor identical to R_m. In these tests, we have observed a not negligible overheating of the SMD resistors, which follows the standard behavior expressed by Eq. (1), with $n=3.4\pm0.2$.

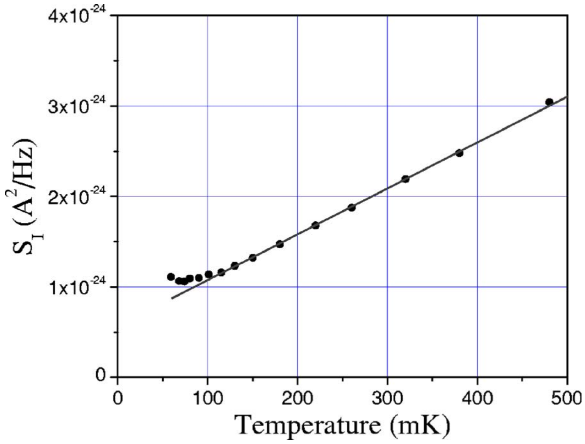

FIG. 2. (Color online) Total noise measured by the readout SQUID as a function of the bath temperature at zero bias current. The deviation from a linear dependence below 100 mK is attributed to excess power dissipated in the resistor. The linear fit of the data above 100 mK is used to calibrate the noise thermometer, that is, to convert the noise to electron temperature.

During the main experimental tests with the film resistor R_s, the temperature T_m has always been calculated by taking into account the overheating of R_m. The corresponding noise contribution is lower than 5% of the total noise for the measurements at $T=59$ mK and lower than 10% for a measurement performed $T=1.2$ K.

The knowledge of both S_{I0} and T_m allows us to measure the electron temperature T_s of the film resistor as a function of the measured total noise S_I at different bias power levels. For the data analysis, we fit the T_s vs P_s experimental data with the function

$$T_s = \left(\frac{P_s + P_0}{\Sigma\Omega} + T_0^n \right)^{1/n}, \quad (7)$$

where T_0 is the bath temperature, as measured by the thermometer, and P_0, n, and $\Sigma\Omega$ are the fitting parameters. Equation (7) is another form of Eq. (4), with $T_e \equiv T_s$ and the total power P expressed by P_s+P_0. P_s is the power dissipated in the film resistor by the bias current, and P_0 is a constant positive term, which accounts for excess power dissipated in the resistor. For example, this excess power could be produced by rf interference, generating rf currents in the wiring.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

Measurements of the electron temperature as a function of the dissipated power at the lowest operating bath temperature $T_0=59$ mK are shown in Fig. 3, both for a simple resistor without cooling fins and for a resistor with the cooling fins. The data relative to the simple resistor are in good agreement with Eq. (7). The fit yields $P_0=1.4\pm0.2$ pW, $n=5.01\pm0.05$, and $\Sigma\Omega=(2.2\pm0.7)\times10^{-8}$ W/K^5. In contrast, the data relative to the resistor with the cooling fins are not in good agreement with Eq. (7). In particular, the slope is found to be slightly variable from about $n=2.5$ at low power to about $n=3.5$ at high power. The excess power term $P_0 \simeq 1$ pW is

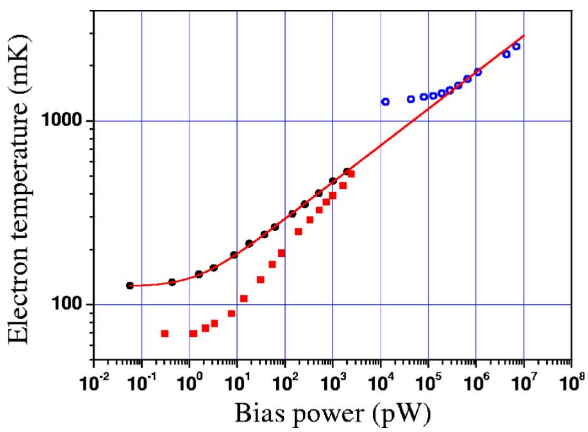

FIG. 3. (Color online) Electron temperature as a function of the power dissipated in the palladium film resistor. Curves are shown for a standard resistor without cooling fins (full circles) and for a resistor with the cooling fins (squares) at T_0=59 mK. Measurements on the standard resistor in pumped liquid helium bath (T_0=1.2 K) are also shown (hollow circles). The solid line is the best fit with Eq. (7) on the data relative to standard resistor at T_0=59 mK.

instead comparable to the one measured without cooling fins.

We briefly discuss the term P_0. The magnitude of the measured excess power is comparable to what one expects from rf interference in a typical laboratory environment, especially as we did not use any rf or microwave filters in our wiring. Clearly, using suitable filters will be required if one wishes to explore regions at lower power and temperature. It is less important if one wishes to operate devices, such as dc SQUIDs, for which the intrinsic power dissipation is significantly larger.

Let us discuss the observed exponent. We found n very close to 5 for resistors without cooling fins. This cannot be explained by either a contact thermal resistance (for instance, between the film and the substrate or between the substrate and the fiberglass board), or by a series thermal resistance (for instance the fiberglass holder), for which we would expect, respectively, n=4 and n=2–3. Moreover, a naive estimation of the total thermal resistance between the chip and the copper box gives a value at least 2 orders of magnitude smaller than that observed. Therefore, we believe that the overheating is caused by the hot-electron effect.

A further support to this conclusion is given by the additional data, also shown in Fig. 3, measured at much higher-power levels. The measurements were performed by immersing the sample in a superfluid helium bath at T_0=1.2 K, so as to substantially reduce the contact resistance R_K. In spite of this, the overheating of the resistor, above the low power knee, is very close to the extrapolation of the fit to the T_0=59 mK data.

Using the estimated volume of the resistors, $\Omega=19\ \mu\mathrm{m}^3$, we infer the material parameter $\Sigma=(1.2\pm0.4)\times10^9\ \mathrm{W/K^5\,m^3}$. This value is comparable to the value obtained in similar experiments on metals in the pure limit[4] and on alloys.[1] The value predicted by the free-electron model Eq. (2), using electronic and elastic parameters of palladium reported in literature,[8,11] is roughly an order of magnitude smaller. Therefore, the simple clean 3D free-electron model predicts the right exponent n but fails to predict the magnitude of the *e-p* rate. This result is not surprising, as our sample is clearly in the dirty limit.

On the other hand, the exponent n=5 is in contrast with the predictions for a metal in the dirty limit,[6] which would predict n=6 or n=4. The remarkable agreement with n=5 over the whole explored range leads one to exclude that the observed exponent is due to some transitions between the two regimes. However, in a recent work,[7] the exponent n=6 has indeed been observed in samples of Au and Cu in the dirty limit. It has then been conjectured that the expected exponent n=6 should be observed in pure noble metals in the dirty limit, but for some unknown reason it is not observed in alloys. Our measurements show that, at least for palladium, this hypothesis is not verified. We conclude that the conditions that make possible the transition to n=6 have not been clearly identified yet. More generally, as pointed out in another recent article,[3] the exponent n=5 is observed much more frequently than one would expect on the basis of the most accepted models, suggesting a widespread breakdown of the models.

Let us discuss the measurements on the resistors with cooling fins. Cooling fins allow the electrons to diffuse in a larger region, thus increasing the effective volume available for electron-phonon energy relaxation.[1] This significantly reduces the electron temperature at a given power, as shown in Fig. 3. A decrease of the contact resistance R_K because of the increased area would explain a reduction of the electron temperature as well. However, the different slopes of the temperature-power relation could not be explained. On the other hand, the change in slope is qualitatively predicted by the hot-electron effect model, if one takes into account the diffusion of the hot electrons. In fact, only a limited portion of the total volume of the fin is available for the *e-p* thermalization, because the hot electrons generated in the resistor diffuse in the fin only over a distance comparable with the temperature-dependent diffusion length. A naive estimation, based on considering the Brownian motion of the hot electrons, leads to a diffusion length $l_d \simeq (l_{\mathrm{in}} l)^{1/2}$, where $l_{\mathrm{in}} = v_F \tau_{ep} \propto T_e^{-(n-2)}$ is the inelastic mean free path.[1] Thus, l_{in}, l_d, and the effective volume increase with decreasing temperature. A course estimate yields $l_d \sim 1$ mm at T_0=59 mK. The following behavior is thus expected: at high temperatures, the hot electrons generated in the resistor cannot diffuse significantly into the fin, and one has the standard curve with n=5, as expected in the absence of cooling fins. At intermediate temperatures (roughly $T_e < 1$ K in our case), the effective volume increases with decreasing temperature. This is equivalent to an effective reduction of the exponent n in Eq. (7), which in turn depends on the size, shape, and dimensionality of the fins. At sufficiently low temperatures, not achieved in our experiment, the entire volume of the fin will be available, and the slope of the curve is expected to return to n=5, but at a temperature level a factor $(\Omega'/\Omega)^{1/n}$ lower than at high temperature, where Ω and Ω' are, respectively, the volume of the resistor and that of the fin.

We conclude with some considerations on the implications of our experimental results on the optimization of a dc SQUID at ultralow temperatures. The noise of such a device, at least in the white noise region, is expected to scale with the thermodynamic temperature of the electron gas in the thin-film shunt resistors.[12] In an earlier experimental study,[1] Wellstood *et al.* showed that the hot-electron effect sets a fundamental limit to the minimum electron temperature that can be achieved, because a constant power, on the order of 10–1000 pW, is dissipated in the device. However, they also showed that the use of cooling fins can reduce the saturation temperature from about 150 mK to less than 50 mK. For a SQUID with shunt resistors similar to those tested in this experiment, at comparable power dissipation of the order of 10 pW, the saturation temperature would improve less, from about 200 mK to 100 mK.

The higher saturation temperature in our case is to be attributed to the slightly worse characteristic parameters of palladium with respect to the AuCu alloy used by Wellstood *et al.* In particular, the scattering parameter Σ is slightly smaller, and the diffusion length l_d in the cooling fins is significantly smaller, due to the lower elastic mean free path. Therefore, the results of our measurements suggest that the cooling fins must be made from a material with the highest possible conductivity, in order to maximize the diffusion length at a given temperature, and hence the effective volume available for *e*-*p* scattering.

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*Electronic mail: vinante@science.unitn.it

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