Influence of the interference of electron–phonon and electron–impurity scattering on the conductivity of unordered Nb films

N. G. Ptitsina, G. M. Chulkova, and E. M. Gershenzon

Moscow State Pedagogic University, 119882 Moscow, Russia

M. E. Gershenzon

Institute of Radio Engineering and Electronics, Russian Academy of Sciences, 103907 Moscow, Russia

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The temperature dependence of the resistivity of Nb thin films has been studied at $T=4.2$–300 K. It has been shown that quantum interference between electron–phonon and electron–impurity scattering determines the temperature dependence of the resistivity of the films investigated over a broad temperature range. The magnitude of the contribution of the electron-phonon-impurity interference is described satisfactorily by the theory developed by Reĭzer and Sergeev {Zh. Éksp. Teor. Fiz. **92**, 2291 (1987) [Sov. Phys. JETP **65**, 1291 (1987)]}. The interaction constants of electrons with longitudinal and transverse phonons in Nb films have been determined for the first time by comparing the experimental data with the theory. The values of the constants obtained are consistent with the data on the inelastic electron-phonon scattering times in the films investigated. The contribution of the transverse phonons is dominant both in the interference correction to the resistivity and in the electron energy relaxation. © *1995 American Institute of Physics.*

1. INTRODUCTION

Recent theoretical and experimental investigations have shown that the electron–phonon interaction in metals is modified significantly when disorder is enhanced. As a result, impure metals exhibit temperature dependences of the electron–phonon scattering time $\tau_{e\text{-}ph}$ (Refs. 1 and 2), as well as temperature dependences of the resistivity (see, for example, Refs. 3 and 4 and the references therein), differing from those of pure metals. The parameter which specifies the character of the electron–phonon interaction is the product of the electron mean free path l and the wave vector q of a thermal phonon: the "dirty" limit $ql \ll 1$ corresponds to the temperature range $T \ll \hbar u/k_B l$ (u is the velocity of the phonons), and the "clean" limit $ql \gg 1$ corresponds to $T \gg \hbar u/k_B l$. As disorder is enhanced, the interaction of electrons with longitudinal phonons should weaken, while the interaction with transverse phonons should become stronger.

The mechanism determining the temperature dependence of the resistivity of impure metals at low temperatures, i.e., the interference between electron–phonon and electron–impurity scattering, has been studied experimentally and theoretically since the sixties (see, for example, Refs. 5–9). The complexity of the theoretical description of this phenomenon is attributable to the large number of electron scattering channels generated by quantum interference. The neglect of some of them results not only in a change in the numerical value of the corresponding contribution to the resistivity $\Delta\rho_{e\text{-}ph\text{-}imp}$, but also in reversal of its sign. Reĭzer and Sergeev took into account all possible channels and used different methods (a linear response method and a method based on the quantum kinetic equation) to obtain identical expressions for $\Delta\rho_{e\text{-}ph\text{-}imp}$, which suggest that the results obtained in Ref. 9 are definitive. As in the preceding studies,[5–8] Reĭzer and Sergeev[9] showed that the contribution $\Delta\rho_{e\text{-}ph\text{-}imp}(T)$ is proportional to the residual resistivity ρ_0 and depends quadratically on the temperature at $T \ll \Theta_D$ (Θ_D is the Debye temperature). One qualitatively new result of the work in Ref. 9 was the proof that the most significant contribution to $\Delta\rho_{e\text{-}ph\text{-}imp}$ is made by the interaction of electrons with *transverse* phonons. We recall that according to the traditional theory of the electron–phonon interaction in *pure* metals, electrons do not interact with transverse phonons in the simple isotropic model. The systematic treatment of electron–phonon-impurity interference in *impure* metals in Ref. 9 leads to a result which is paradoxical at first glance: the contributions to the resistivity due to the interactions with transverse and longitudinal phonons have different signs, the contribution to the resistivity being *negative* in the latter case (in contrast to the erroneous results of several previous theoretical studies). These features of the electron–phonon interaction in impure metals are associated with the character of the shielding of the transverse electromagnetic fields and the quantum nature of the interference. The total correction $\Delta\rho_{e\text{-}ph\text{-}imp}$ is positive, because the propagation velocity of longitudinal phonons is always greater than the velocity for transverse phonons.

Dependences of the $\Delta\rho(T) \propto T^2$ type have been observed experimentally at low temperatures for metallic samples of various degrees of purity (see, for example, Refs. 3 and 4). For high-purity metals, such dependences, which are generally observed at $T<1$ K, are attributed to electron–electron scattering in the presence of umklapp scattering processes.[3] For impure metals the upper boundary of the temperature range in which $\Delta\rho(T) \propto T^2$ is significantly higher. In the latter case the observed contribution $\Delta\rho(T)$ is proportional to

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TABLE I. Parameters of the samples.

No.	d, Å	l, Å	ρ_0 (Ω·cm)	T_c, K
1	100	12	$3.2 \cdot 10^{-5}$	5
2	140	28	$1.4 \cdot 10^{-5}$	6.7

the residual resistivity ρ_0, indicating that it is due to electron–phonon–impurity interference. After the appearance of the Reĭzer–Sergeev theory, its implications were tested in Ref. 4, where the temperature dependence of the resistivity of thin gold films was measured. It was shown that this dependence is described by the sum of the electron–phonon scattering $\Delta\rho_{\text{e-ph}}$ and the electron–phonon–impurity interference $\Delta\rho_{\text{e-ph-imp}}$, the dominant contribution at low temperatures, $\Delta\rho_{\text{e-ph-imp}}$, being in quantitative agreement with the theory in Ref. 9. It was also shown in Ref. 4 that the data in the previous investigations, in which alkali-metal melts were studied (see, for example, Ref. 3), are in satisfactory agreement with the Reĭzer–Sergeev theory.

It should be noted that a quantitative comparison with the theory in Ref. 9, as well as, on the other hand, with the classical Bloch–Grüneisen theory for electron–phonon scattering, encounters difficulties due to the simplified model of metals used in these theories. Consequently, it is not clear *a priori* that agreement should be equally good for other metals, especially transition metals. Therefore, in the present work the experiments devised to test the theory in Ref. 9 were continued using thin films of niobium, which is characterized by an intricate Fermi surface and a strong electron–phonon interaction. The relatively high (in comparison with gold) Debye temperature (Θ_D=275 K) and the strong electron–phonon interaction in niobium allow us to hope to expand the temperature range in which the interference contribution predominates in the temperature dependence of the resistivity. This situation facilitates, in particular, the problem of isolating the contribution of interest to us against the background of quantum corrections to the resistivity, which dominate at low temperatures.[4] In addition, there is one more reason why Nb films were selected. We previously studied electron–phonon relaxation at various values of ql in similar Nb thin films.[1] It was shown as a result that electronic parameters such as the density of electron states ν_0 and the Fermi velocity v_F do not vary significantly in the films investigated as the electron mean free path varies. This finding is also important for the questions discussed in the present paper.

2. EXPERIMENTAL RESULTS

The temperature dependence of the resistivity was measured in the T=4–300 K range for a large series of Nb films with various values of the electron mean free path, which were deposited by electron-beam evaporation on sapphire substrates. We note that experimental isolation of the interference contribution requires the variation of l over a broad range. The measurement data are presented for the two samples which differ most strongly with respect to the value of l (their parameters are presented in Table I). The geometry of the samples was such that the total resistance R was equal

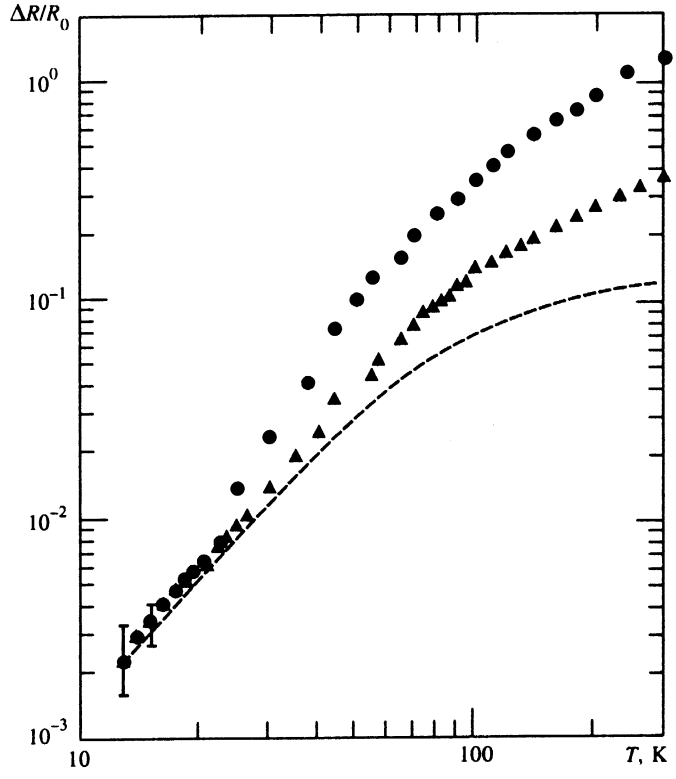

FIG. 1. Temperature dependence of the relative resistivity $\Delta R(T)/R_0 \equiv [R(T)-R_0]/R_0$ for samples 1 (▲) and 2 (●). The dashed curve is a plot of the temperature dependence of $\Delta\rho_{\text{e-ph-imp}}/\rho_0$ calculated from Eq. (1) with $\Delta\rho/(\rho_0 T^2)=1.5\times10^{-5}$ K^{-2}. The error in the measurement of ΔR corresponds to the error in the determination of R_0.

to 10^4 to 10^6 Ω. This made it possible to perform measurements with high relative accuracy $\Delta R/R \approx 10^{-6}$ at low currents that do not heat the sample. The sample, thermometer, and heating element were placed in an evacuated hood, in which the long-term stability of the temperature was at worst 0.5 K at $T<40$ K and no poorer than 1–2 K at $T>40$ K. To increase the accuracy of the measurements, the load resistance in the constant-current source was also placed in a thermally stabilized, low-temperature chamber.

The temperature-dependence contribution to the resistance $\Delta R(T)$, which is equal to the difference between the total resistance R and the residual resistance R_0, will be discussed below. The resistance corresponding to the plateau of $R(T)$ immediately before the superconducting transition, was taken as the value of R_0. The plots of $\Delta R(T)/R_0 \equiv \Delta\rho(T)/\rho_0$ for samples 1 and 2 are shown in Fig. 1. The plots for all the samples investigated coincide at low temperatures: $\Delta\rho(T)$ is proportional to T^2, and the ratio $\Delta\rho(T)/\rho_0$ does not depend on the residual resistivity. The values of $\Delta\rho(T)/(\rho_0 \cdot T^2)$ are equal to 1.5×10^{-5} K^{-2}. As room temperature is approached, the values of $\Delta\rho(T)/\rho_0$ obtained for different films begin to differ significantly, a deviation from the $\Delta\rho(T)/\rho_0 \propto T^2$ dependence observed at lower temperatures for films with large values of l.

3. DISCUSSION OF RESULTS

The intense impurity scattering in all the samples investigated with a resistivity $\rho_0 > 10^{-5}$ Ω·cm completely sup-

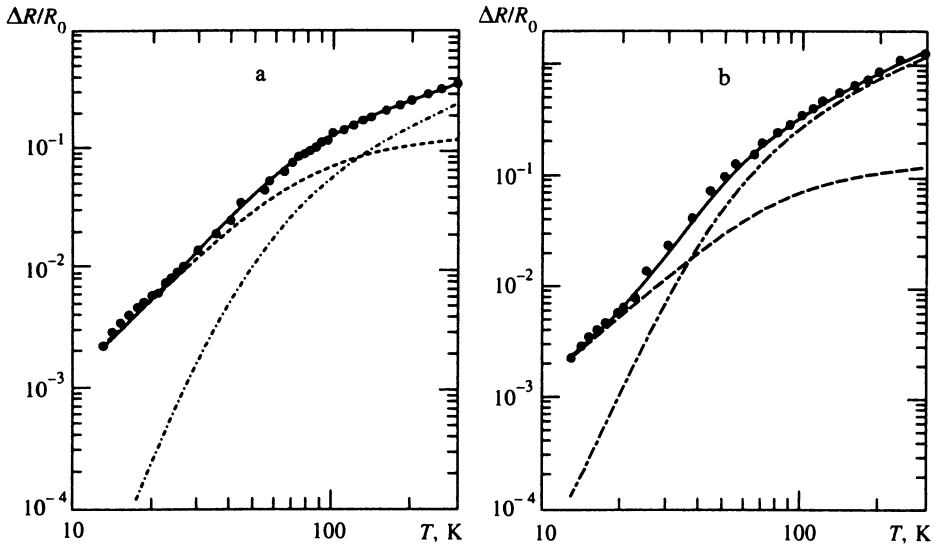

FIG. 2. Temperature dependence of the relative resistivity $\Delta R(T)/R_0$ for samples 1 (a) and 2 (b). The dashed curves represent the dependence of $\Delta\rho_{e\text{-}ph\text{-}imp}/\rho_0$ in Fig. 1, the dot-dashed curves describe the temperature dependence of the Bloch–Grüneisen contribution $\Delta\rho_{e\text{-}ph}(T)/\rho_0$, and the solid curves are plots of the sum of these two contributions.

presses the effects associated with the anisotropy of the electron–phonon interaction for different parts of the Fermi surface.[10] As we have already noted, in this range of values of ρ the deviation from Mathiessen's rule (the appearance of an additional contribution to the resistivity along with ρ_0 and $\Delta\rho_{e\text{-}ph}\propto T^5$) is due to interference between electron–impurity and the electron–phonon scattering. We stress that the large absolute values of this contribution and, evenly more importantly, its proportionality to the residual resistivity ρ_0 permit disregarding the other scattering processes which also produce a quadratic temperature dependence of the resistivity (see, for example, Ref. 4).

Let us compare the experimental values of $\Delta\rho(T)/\rho_0$ with the results of the theory. We stress that the data analyzed below were obtained over a temperature range in which the clean limit condition $ql>1$ holds. When this condition is satisfied and it is assumed that $\rho_0\gg\Delta\rho(T)$, the correction to the resistivity $\Delta\rho_{e\text{-}ph\text{-}imp}$ associated with the interference of electron–photon and electron–impurity interactions is described by the expression[9]

$$\frac{\Delta\rho_{e\text{-}ph\text{-}imp}}{\rho_0}=\left[1-\frac{\pi^2}{16}-2\left(\frac{u_l}{u_t}\right)^3\right]\frac{2\beta(k_BT)^2}{\varepsilon_F p_F u_l}\times\int_0^{\Theta_D/T}\left[\frac{2}{e^x-1}-\frac{2xe^x}{(e^x-1)^2}\right]x\,dx, \qquad (1)$$

where the interaction constant of electrons with longitudinal phonons is

$$\beta=\left(\frac{2}{3}\varepsilon_F\right)^2\frac{\nu_0}{2\tilde{\rho}u_l^2}, \qquad (2)$$

ε_F is the Fermi energy, p_F is the Fermi momentum, k_B is the Boltzmann constant, ρ is the density of the pure metal, ν_0 is the density of electron states, and u_l and u_t are the propagation velocities of longitudinal and transverse phonons, respectively. When $T\leq\Theta_D/10$, the integral in (1) tends to $(-\pi^2/3)$, and dependence (1) is simplified:

$$\frac{\Delta\rho_{e\text{-}ph\text{-}imp}}{\rho_0}=\left[2\left(\frac{u_l}{u_t}\right)^3+\frac{\pi^2}{16}-1\right]\frac{2\pi^2\beta}{3\varepsilon_F p_F u_l}(k_BT)^2. \qquad (3)$$

According to Ref. 11, the temperature dependence of the contribution $\Delta\rho_{e\text{-}ph}$ to the resistivity of an impure metal is described by the Bloch–Grüneisen law, just like the resistivity of a pure metal, which is governed by electron–phonon scattering:[12]

$$\frac{\Delta\rho_{e\text{-}ph}}{\rho_0}=\frac{C}{\rho_0}\left(\frac{T}{\Theta_D}\right)^5\int_0^{\Theta_D/T}\frac{x^5}{(e^x-1)(1-e^{-x})}\,dx. \qquad (4)$$

Since the contribution of electron–phonon interaction $\Delta\rho_{e\text{-}ph}(T)$ does not depend on the residual resistivity, the values of $\Delta\rho_{e\text{-}ph}$ normalized to ρ_0 should be significantly dependent on the mean free path in the temperature range where the electron–phonon scattering dominates the temperature dependence of the resistivity. This accounts for the differences in the plots of $\Delta\rho(T)/\rho_0$ observed at high temperatures for samples with different values of l (Fig. 1).

The Bloch–Grüneisen theory was devised for an isotropic Fermi surface and a simplified phonon spectrum, and it does not take into account umklapp scattering processes or the presence of two types of carriers in transition metals. As a result, the absolute values of $\Delta\rho_{e\text{-}ph}$ calculated under this theory can, in principle, differ significantly from the experimental values of $\Delta\rho_{e\text{-}ph}(T)$ (Ref. 12). Therefore, we used the following technique to isolate the contribution of the electron–phonon interaction. The temperature dependence of $\Delta\rho_{e\text{-}ph\text{-}imp}/\rho_0$ defined by (1) was appended to the experimental values of $\Delta\rho(T)/\rho_0$ at low temperatures, at which $\Delta\rho_{e\text{-}ph}(T)$ may be neglected owing to its stronger dependence on the temperature ($\propto T^5$) [the dashed curves in Fig. 2 are plots of (1)]. The value of $\Delta\rho_{e\text{-}ph\text{-}imp}(300\text{ K})/\rho_0$ extrapolated to $T=300$ K according to (1) was subtracted from the experimental value of $\Delta\rho(300\text{ K})/\rho_0$, and the difference was identified with $\Delta\rho_{e\text{-}ph}(300\text{ K})/\rho_0$. The coefficient C in Eq. (4) was thus determined. We stress that the value of $\Delta\rho_{e\text{-}ph}$ thus determined at $T=300$ K coincides with the conductivity of pure

niobium at room temperature ($\approx 1.4\times10^{-5}$ Ω·cm). The dot-dash curves in Figs. 2a and 2b are plots of the Bloch–Grüneisen contribution $\Delta\rho_{\text{e-ph}}(T)/\rho_0$ constructed over the entire temperature range according to (4), and the solid curves are theoretical plots of $\Delta\rho(T)/\rho_0$, which is equal to the sum of the Bloch–Grüneisen and Reĭzer–Sergeev contributions. Good agreement between the experimental plots of $\Delta\rho(T)/\rho_0$ and the theoretical plots thus obtained was observed for all the Nb films studied. As is seen from Fig. 2a, the contribution of the interference scattering mechanism to the resistivity is significant for sample 1, which has a small value of l, even at room temperature. At the same time, $\Delta\rho_{\text{e-ph}}$ becomes decisive for sample 2, which has a large value of l, by $T>35$ K. The interference contribution may be neglected for still purer Nb films ($l>30$ Å), the temperature dependence of their resistivity being described satisfactorily by the Bloch–Grüneisen contribution over the entire range of $T>T_c$.

We begin the quantitative comparison of the experimental and theoretical values of $\Delta\rho_{\text{e-ph-imp}}$ with a discussion of the parameters of the Nb films investigated. As we have already noted, we used Nb films with a resistivity $\rho_0=1.3\times10^{-5}$ to 3.5×10^{-5} Ω·cm, which are similar to the films previously investigated in Ref. 1. The resistivity of these films is inversely proportional to the diffusion coefficient D, which was determined in Ref. 1 in the standard manner from the temperature dependence of the upper critical field. The electron density of states $\nu_0=(e^2\rho D)^{-1}$ corresponding to the experimental values of ρ and D for these films is independent of the degree of disorder and equals $\nu_0=1.6\times10^{23}$ eV^{-1}·cm^{-3}. This value coincides with the data obtained for the film samples in Ref. 13 and is approximately 80% greater than the value of ν_0 determined from the electronic specific heat for solid niobium.[14] The uncertainty in the energy of the electron states $\hbar/\tau$ ($\tau=l/v_F$ is the electron momentum relaxation time) amounts to 0.2 eV, which is significantly less than the characteristic scales for the variation of the density of electron states (~1 eV); therefore, electron–impurity scattering cannot have a significant influence on ν_0 or on the value of the Fermi energy ε_F. The value $\varepsilon_F=5.3$ eV, which corresponds to pure niobium,[14] will be used henceforth. On the other hand, electron–impurity scattering causes considerable isotropization of the Fermi surface, to which we shall assign the value of the electron momentum following from the model of free electrons $p_F=1.2\times10^{-19}$ g·cm·s^{-1} (Ref. 14). In addition, we shall use the experimental value of the Fermi velocity averaged over the Fermi surface $v_F=2.7\times10^7$ cm/s (Ref. 15), the velocities of longitudinal and transverse sound $u_l=5.1\cdot10^5$ cm/s and $u_t=1.7\times10^5$ cm/sec, and the density of the pure metal ρ(Nb) $=8.4$ g/cm^3 (Ref. 14) in the calculations.

Let us examine whether $ql>1$ for the films investigated. The experimental values $\rho_0=1.3\times10^{-5}$ to 3.5×10^{-5} Ω·cm correspond to values of the electron mean free path $l=10$–30 Å. For example, for fairly "dirty" sample 1, $ql>1$ for transverse phonons at $T>10$ K and for longitudinal phonons at $T>30$ K. Since the contribution of the transverse photons to $\Delta\rho_{\text{e-ph-imp}}$ is dominant, an interference contribution to the resistivity quadratic in the temperature is observed experimentally down to the lowest temperatures used, i.e., $T\approx 2T_c$.

The experimental value of the interaction constant with longitudinal phonons β calculated from the value $\Delta\rho_{\text{e-ph-imp}}/(\rho_0T^2)=1.5\times10^{-5}$ K^{-2} obtained from (3) using the values of the parameters given above is 1.2, which is approximately 60% greater than the value of β calculated according to (2). Such agreement should be considered satisfactory in view of the uncertainty in some of the parameters of Nb. Since the main contribution to $\Delta\rho_{\text{e-ph-imp}}$ is made by the transverse phonons, we can omit the contribution of the longitudinal phonons in a first approximation and write expression (3) in the form

$$\frac{\Delta\rho_{\text{e-ph-imp}}}{\rho_0}\approx\frac{4\pi^2\beta_t}{3\varepsilon_F p_F u_t}(k_BT)^2, \tag{5}$$

where $\beta_t=\beta(u_l/u_t)^2$ is the interaction constant of electrons with transverse phonons. Substituting the experimental data and the parameters of Nb into (5), we obtain a value of 10.8 for β_t.

We stress that hitherto there have been practically no data on the determination of β_t. Indirect information on the interaction with transverse phonons can be obtained from data on electron energy relaxation in impure metals in particular by studying the phase relaxation time τ_φ of the electronic wave function. The values $\tau_{\text{e-ph}}=2\times10^{-12}$ to 3×10^{-12} s were obtained in Refs. 16–18 for Nb films with a mean free path $l=10$–15 Å at $T=20$ K. (In the ultrathin Nb films investigated in those studies the electron–phonon interaction makes the main contribution to the phase relaxation rate only at $T>10$ K, while at lower temperatures the dominant phase relaxation mechanism is based on electron–electron collisions). Since these values of l correspond to q_ll and q_tl of the order of unity at $T=20$ K, the expressions obtained in the "clean" case can be used to evaluate the energy relaxation time $\tau_{\text{e-ph}}$. For the interaction of electrons with longitudinal phonons we can write[19]

$$(\tau_{\text{e-ph},l})^{-1}=\frac{7}{2}\pi\zeta(3)\beta\frac{(k_BT)^3}{h(p_Fu_l)^2}. \tag{6}$$

Plugging in the data used above for Nb, including the derived value $\beta=1.2$, we obtain $(\tau_{\text{e-ph},l})^{-1}=8\times10^{10}$ s^{-1}, which is significantly lower than the observed relaxation rate. Thus, the interaction with longitudinal phonons alone is not capable of accounting for the experimental results. The relaxation rate on transverse phonons has the form[19]

$$(\tau_{\text{e-ph},t})^{-1}=\frac{3\pi^2}{2}\beta_t\frac{(k_BT)^2}{(p_Fu_t)(p_Fl)}. \tag{7}$$

Using $\beta_t=10.8$, we find a relaxation rate equal to 3.6×10^{11} s^{-1}, which is in good agreement with the experimentally determined phase relaxation time of the wave function. We note, however, that the dependence $\tau_{\text{e-ph}}\propto l$, which follows from (7), has not yet been confirmed experimentally in the temperature range where $ql>1$. At lower temperatures, at which $ql\ll1$, the dependence $\tau_{\text{e-ph}}\propto1/l$, which is characteristic of the "dirty" limit,[1,19] was observed.

4. CONCLUSIONS

It has been established experimentally that the temperature dependence of the resistivity of thin films of a transition metal such as niobium, as well as the previously investigated univalent metals, is satisfactorily described by the theory of electron–phonon-impurity interference in Ref. 9 over a broad temperature range. The interaction constants of electrons with longitudinal and transverse phonons in Nb films have been determined for the first time by comparing the experimental data with theory. The values of the constants obtained are consistent with data on the inelastic electron–phonon scattering times in the films investigated. The contribution of transverse phonons is dominant both in the interference correction to the resistivity and in electron energy relaxation.

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