

Interrelation of resistivity and inelastic electron-phonon scattering rate in impure NbC films

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A complex study of the **electron-phonon interaction** in thin NbC films with **electron mean free path** $l=2-13$ nm gives strong evidence that **electron scattering** is significantly modified due to the interference between electron-phonon and elastic electron scattering from impurities. The interference T^2 term, which is proportional to the residual resistivity, dominates over the Bloch-Grüneisen contribution to resistivity at low temperatures up to 60 K. The electron energy relaxation rate is directly measured via the relaxation of hot electrons heated by modulated electromagnetic radiation. In the temperature range 1.5–10 K the relaxation rate shows a weak dependence on the electron mean free path and strong temperature dependence $\sim T^n$, with the exponent $n=2.5-3$. This behavior is explained well by the theory of the electron-phonon-impurity interference taking into account the electron coupling with transverse phonons determined from the resistivity data.
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INTRODUCTION

The study of the electron-phonon scattering in disordered conductors, such as impure metals, metallic glasses, and ultrathin films, has attracted much attention in recent years. The inelastic electron-phonon scattering time τ_{e-ph} was investigated in experiments with hot electrons (the electron cooling time), in different nonequilibrium phenomena in superconductors (the quasiparticle recombination time, the relaxation times of phase, and module of the order parameter), and in weak localization experiments (the electron dephasing time). However, in spite of numerous experimental results and theoretical works, the modification of the electron-phonon interaction due to the elastic electron scattering from impurities, defects, or boundaries is still poorly understood.

In clean films (three dimensional with respect to the typical phonon wavelength) a cubic dependence $\tau_{e-ph}^{-1} \sim T^3$ has been typically observed.¹⁻³ In impure films a variety of temperature dependencies from $T^{-1.5}$ to T^{-4} has been obtained for the electron-phonon relaxation time (see, e.g., Refs. 4–6, and references therein). The disorder dependence of τ_{e-ph} has also been measured by several groups, which have obtained inconsistent results. Peters and Bergmann⁷ obtained a very weak dependence of τ_{e-ph} on the electron mean free path l in disordered Au, Ag, and Mg films. Gordon and Goldman⁸ observed a dependence close to $\tau_{e-ph}^{-1} \sim l^{-0.6}$ in Al films. Gershenzon *et al.*⁹ found that the relaxation rate decreases with the mean free path in Nb films, $\tau_{e-ph}^{-1} \sim l$. Recently, Lin and Wu¹⁰ obtained an opposite result in crystalline disordered TiAl alloys, $\tau_{e-ph}^{-1} \sim l^{-1}$.

For years a purposeful study of the interference between electron-phonon and elastic electron scattering has been hampered by the controversial theoretical conclusions. Start-

ing with the same basic model, where the elastic electron scattering is due to the Gauss δ -correlated impurity potential, theoreticians obtained different results. Using transformation to a frame of reference moving with the ions, Schmid¹¹ has found that in presence of strong disorder, $ql \ll 1$ (q is the wave vector of thermal phonon), the electron-phonon interaction should weaken, and the energy relaxation rate τ_{e-ph}^{-1} is of the order of $(ql)\tau_0^{-1}$, where $\tau_0^{-1} \sim T^3$ is the relaxation rate in the pure material. This conclusion was disputed in a number of papers where the relaxation rate $\tau_{e-ph}^{-1} \sim (ql)^{-1}\tau_0^{-1}$ has been found solely due to the electron scattering from vibrating impurities. The disagreement between the two concepts was finally settled by Reizer and Sergeev.¹² The authors demonstrated that a correct kinetic approach to the interference of scattering processes leads to the Schmid's result in any frame of reference. Theory^{12,13} predicts that in the limit $ql \ll 1$, the total contribution of longitudinal and transverse phonons to the inelastic electron scattering rate τ_{e-ph}^{-1} is smaller than the scattering rate τ_0^{-1} in a pure material, where the interaction of electrons with longitudinal phonons is only important.

In a wide intermediate interval $ql \sim 1$ the situation is very nontrivial. The inelastic electron scattering due to longitudinal phonons weakens, while the scattering due to transverse phonons strengthens with a disorder enhancement. Since the velocity of longitudinal sound u_l is usually two to three times greater than the velocity of transverse sound u_t , a crossover from the "impure" limit ($ql \ll 1$) to the "pure" limit ($ql \gg 1$) for transverse phonons occurs at lower temperature than for longitudinal phonons ($q_{t,l} = k_B T / \hbar u_{t,l}$). Therefore, the character of temperature and disorder dependencies of τ_{e-ph} may strongly depend on the ratio u_l/u_t .

Although most of the data obtained correspond to the case

$ql \sim 1$, the comparison of experimental data with theoretical formulas beyond their asymptotic form for $ql \ll 1$, to the best of our knowledge, is still absent. The reliable calculations of τ_{e-ph} are complicated by a lack of information about the phonon and electron energy spectra in films. The sound velocities u_l and u_t may be strongly modified in thin films by defects and acoustic mismatch between film and substrate.

The interference between electron-phonon and electron-impurity interactions manifests itself also in the temperature dependence of resistivity. The corresponding corrections to the resistivity were calculated in a number of theoretical papers. The processes of electron scattering on vibrating impurities were taken into account in Ref. 14. All interference processes were considered in Ref. 15. It was shown that the interaction of electrons with transverse phonons makes a major contribution to the interference correction. The effect of longitudinal phonons, as well as that of the transverse ones, is proportional to T^2 and to the residual resistivity. However, it turns out to be negative and much smaller than the positive contribution of transverse phonons. The interference correction was experimentally observed in alkali metals at very low temperatures.¹⁴ Recently, it was found in thin Au films¹⁶ with $l \sim 10$ nm in the temperature interval up to 15 K, and in Nb films¹⁷ with $l \sim 1$ nm up to 120 K.

One more factor which can effect the strength of the electron-phonon interaction and its temperature dependence is a modification of phonon spectrum in thin film. The role on the phonon dimensionality has been studied both theoretically^{18,19} and experimentally.^{20,21} No strong experimental evidence of the reduced phonon dimensionality effect has been obtained so far. It is stated, in general, that a good coupling between the substrate and film phonons will eliminate the low dimensionality effect.²² Since the acoustic impedances of the film and substrate materials which were used in this work are close, we believe that the above reasoning applies to us, therefore only interaction with three-dimensional (3D) phonons will be considered in the following sections.

In the present paper we study the inelastic electron scattering rate and the interference correction to the resistivity of niobium carbide (NbC) thin films with the electron mean free path l ranging from 2 to 13 nm. The data on the Debye temperature and kinetic constants of electron-phonon interaction obtained in the resistivity measurements are used for calculations of the relaxation rate in the intermediate region $ql \sim 1$. The comparison is made between the theoretical and experimental values of the electron-phonon energy relaxation time. The latter was determined as a cooling time of the electron subsystem perturbed by the amplitude modulated electromagnetic radiation.

THEORY

The electron scattering from longitudinal phonons in impure metal gives a rise to the temperature-dependent resistivity,²³ which is given by exactly the same equation, as the well-known Bloch-Grüneisen term in pure metal:

$$\frac{\Delta \rho_{e-ph}}{\rho_0} = \frac{1}{2} \frac{\pi \beta_l \tau}{\hbar (p_F u_l)^4} (k_B T)^5 \int_0^{\Theta_D/T} \frac{x^5 dx}{(e^x - 1)(1 - e^{-x})}. \quad (1)$$

Here τ is the elastic electron scattering time due to the interaction with impurities, defects, or boundaries, p_F is the Fermi momentum, Θ_D is the Debye temperature, β_l is the kinetic constant responsible for the interaction of electrons with longitudinal phonon, and u_l is the sound velocity of longitudinal phonons. The interference between electron-phonon and electron-impurity scatterings results in a violation of the Matthiessen rule. The interference correction to the resistivity of impure metal for $ql > 1$ is given by¹⁵

$$\frac{\rho_{int}}{\rho_0} = \left[2 \left(\frac{u_l}{u_t} \right) \beta_t - \left(1 - \frac{\pi^2}{16} \right) \beta_l \right] \frac{2 \pi^2 k_B^2}{\varepsilon_F p_F u_l} T^2 \times \int_0^{\Theta_D/T} \frac{e^x (x-1) + 1}{(e^x - 1)^2} x dx, \quad (2)$$

where β_t is the constant of electron coupling with transverse phonons and ε_F is the Fermi energy. The first term in the square bracket corresponds to the interaction with transverse phonons. It dominates significantly over the second term, which is due to the interaction of electron with longitudinal phonons. In the "jellium" model the ratio between the constants is $\beta_t/\beta_l = (u_l/u_t)^2$. Using the data on elastic constants of NbC lattice from Ref. 24 the sound velocities for bulk NbC $u_l = 9.0 \times 10^5$ cm/s and $u_t = 4.4 \times 10^5$ cm/s were calculated, i.e., $\beta_t/\beta_l \approx 4$. One can obtain from Eq. (2), that the contribution of longitudinal phonons is smaller than 3%. Note, that in any material the minimum value of the ratio u_l/u_t is $\sqrt{2}$, and, therefore, the effect of transverse phonons always significantly prevails over the effect of longitudinal ones. Ignoring the contribution of longitudinal phonons, Eq. (2) at $T < \Theta_D$ can be written as

$$\frac{\rho_{int}}{\rho_0} = B T^2 \approx \frac{4 \pi^2 \beta_t}{3 \varepsilon_F p_F u_l} (k_B T)^2. \quad (3)$$

The constants of electron coupling with transverse phonons B and β_t can be determined from the temperature dependence of resistivity.

According to theory^{12,13} the inelastic electron scattering rate of an electron at the Fermi surface ($\varepsilon = 0$) due to the interaction with longitudinal phonons is

$$\tau_{e-l}^{-1} \text{ph}(0) = \frac{7 \pi \zeta(3)}{2} \frac{\beta_l (k_B T)^3}{\hbar (p_F u_l)^2} F_l(q_l l), \quad (4)$$

$$F_l(z) = \frac{2}{7 \zeta(3)} \int_0^{k_B \Theta_D / \hbar u_l z} dx \Phi_l(xz) [N(x) + f(x)] x^2, \quad (5)$$

$$\Phi_l(x) = \frac{2}{\pi} \left(\frac{x \arctan(x)}{x - \arctan(x)} - \frac{3}{x} \right).$$

$N(x)$ and $f(x)$ are the Bose and Fermi functions, $\Phi_l(x)$ is the Pippard function, and $\zeta(n)$ is the Riemann zeta function. The contribution of transverse phonons, in turn, is given by

$$\tau_{e-t}^{-1} \text{ph}(0) = \frac{3 \pi^2 \beta_t (k_B T)^2}{(p_F u_l) (p_F l)} F_t(q_t l), \quad (6)$$

$$F_t(z) = \frac{4}{\pi^2} \int_0^{k_B \Theta_D / \hbar u_t z} dx \Phi_t(xz) [N(x) + f(x)] x,$$

$$\Phi_t(x) = \frac{2x^3 + 3x - 3(x^2 + 1)\arctan(x)}{2x^3}. \quad (7)$$

The following asymptotics are valid for the F functions at $T \ll \theta_D$: $F_{l,t}(q_{l,t}, l) = 1$ if $q_{l,t} l \gg 1$ and $F_l(q_l l) = 2\pi^3 q_l l / 35\zeta(3)$, $F_t(q_t, l) = (\pi q_t l)^2 / 10$ in the opposite case $q_{l,t} l \ll 1$.

The calculations using Eqs. (4)–(7) (see the following sections) show that for our films with the electron mean free path $l = 2$ –13 nm, the inelastic interaction of electrons with transverse phonons dominates (i.e., $\tau_{e-t \text{ ph}} \ll \tau_{e-l \text{ ph}}$) up to 60 K. Because the interaction with transverse phonons plays a key role in both phenomena, one can find the corresponding constant B from the resistivity data at $ql > 1$ [Eq. (3)] and then use it to calculate the electron inelastic scattering rate at arbitrary values of ql including the intermediate region $ql \sim 1$. In a formal way, by extracting the coupling constant from Eq. (3) and substituting it into Eq. (6), we find

$$\tau_{e-t \text{ ph}}^{-1} = \frac{9}{8} BT^2 F_t(q, l) \tau^{-1}. \quad (8)$$

The remarkable feature of this approach is that the poorly known Fermi momentum and energy are not needed for calculation of the relaxation rate.

PARAMETERS OF SAMPLES AND CHARACTERIZATION

Thin NbC films were prepared by laser ablation of a niobium target in hydrocarbon atmosphere onto polished sapphire substrates. The fabrication details have been given elsewhere.²⁵ The experimental samples were several parallel strips 2 μm wide and 40 μm long connected to contact pads and were prepared by photolithography and reactive ion etching techniques. The electron mean free path was controllably varied by a bombardment of initially pure films ($l \approx 13$ nm) with 200 keV N_2^+ ions. The ion dose ranged between 20 and 240 μC . Note, since the ion energy was very high, then the procedure did not give a rise to the triple NbCN compound for 20 nm thick films.

In this paper we present the experimental results obtained for one typical sample. The sample was exposed to four rounds of the ion bombardment. In the following, the notation sample No. 1 means the original pure sample, whereas samples Nos. 2–4 are the sequential states of sample No. 1 after ascending ion exposure. After each exposure the critical temperature of the superconducting transition T_c , residual resistance R_0 , and temperature dependence of the perpendicular critical magnetic field $H_{c2}(T)$ were measured.

In the course of measurements an experimental sample was mounted on a copper holder along with a heater and a semiconductor thermometer. The thermometer had a resolution of 1 K at $T > 100$ K and of 0.01 K at $T \leq 4.2$ K. A multimode optical fiber was attached to the holder to illuminate the sample through a transparent substrate. The holder was attached to the output of a millimeter wave multimode waveguide made from stainless steel. All the components were assembled in a vacuum jacket immersed in liquid He bath. The control of the operating temperature was done either by He vapor pumping in the range 1.6–4.2 K (no vacuum jacket was used for this range) or by the heater for

TABLE I. Parameters of samples.

Sample	ρ_0 ($\mu\Omega$ cm)	$-dH_{c2}/dT$ (kOe/K)	T_c (K)	D (cm^2/s)	τ (fs)	l (nm)
1	16	2.2	12	9.5	59	13.0
2	54	5.0	10	3.0	19	4.1
3	68	6.2	9.5	2.3	14	3.1
4	84	6.8	8.8	2.1	13	2.9
5	98	9.7	8.0	1.4	8.7	1.9

$T > 4.2$ K. A uniform magnetic field perpendicular to the sample surface was created by a superconducting solenoid mounted in the same bath cryostat.

The diffusion coefficient D was determined in the framework of the Ginzburg-Landau theory using T_c , R_0 , and $H_{c2}(T)$ and the approximation of Ref. 26. The theory for strongly coupled superconductors [in NbC the coupling constant $\lambda \approx 0.9$ (Ref. 27)] suggests the following expression for the diffusion constant:

$$D = \frac{4\pi k_{BC}}{7\zeta(3)e} \left(\frac{T_c}{T_{c0}} \frac{dH_{c2}^0}{dT} - \eta^{-1} \frac{dH_{c2}}{dT} \right)^{-1}. \quad (9)$$

Here H_{c2}^0 and T_{c0} are the critical magnetic field and temperature in ideally pure material. The value of $dH_{c2}^0/dT = 0.8$ kOe/K and Fermi velocity $v_F = 2.2 \times 10^7$ cm/s was obtained in Ref. 28 for thicker and purer NbC films. The critical temperature of pure material T_c^0 was assumed to be 12 K, $\eta = 1.05$.²⁸ Experimental values of the diffusivity are presented in Table I. Then the elastic relaxation times and mean free paths were found, respectively, as $\tau = 3D/v_F^2$ and $l = 3D/v_F$.

INTERFERENCE CORRECTION TO RESISTIVITY

The temperature dependence of the resistance $R(T)$ was measured over the range 10–300 K with a highly precise digital multimeter with an automatic compensation of the thermal-electrical emf. For the samples with $R \sim 10^2$ – 10^4 Ω the relative error of the resistance measurements was of the order of 10^{-6} . The measuring current was typically 10 μA , which is too small to cause any noticeable heating of the samples.

The temperature-dependent resistance in the normal state of NbC films was studied at $T \geq 2T_c$ where the effect of superconducting fluctuations is negligible. Temperature-dependent resistivities $\Delta\rho(T)/\rho_0 = \Delta R(T)/R_0 = [R(T) - R_0]/R_0$ are plotted in Fig. 1. The dependencies $\Delta\rho(T)/\rho_0$ for the exposed samples show similar behavior at low temperatures, namely, $\Delta\rho(T)$ grows proportional to T^2 , and the ratio $\Delta\rho(T)/\rho_0$ does not depend on the residual resistivity. According to Eq. (2), the proportionality of the resistivity to T^2 and also to the residual resistivity unambiguously evidences in favor of its interference origin. Deviations from T^2 law at higher temperatures are associated with the temperature dependence of the interference term (the asymptotic T^2 form is valid only for $T \leq 0.3\theta_D$) and also with the Bloch-Grüneisen term [Eq. (1)].

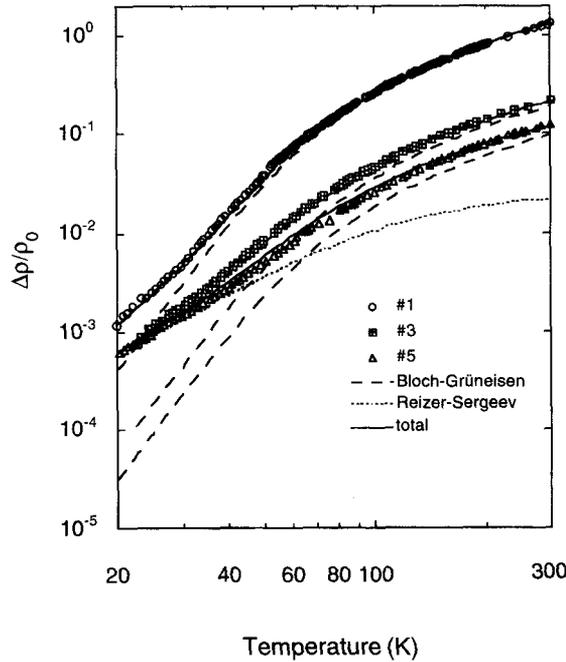


FIG. 1. Temperature dependencies of resistivity for sample Nos. 1, 3, and 5. The dashed lines are the Bloch-Grüneisen contribution [Eq. (1)], the dotted line is the interference contribution [Eq. (2)], the solid lines are the total of the two above.

In order to extract the interference contribution $\Delta\rho_{\text{int}}/\rho_0$, we used the following procedure. Since in the impure samples the Bloch-Grüneisen contribution is negligible at low temperatures, we associated the T^2 term proportional to the residual resistivity with the interference contribution. In the entire temperature interval the measured temperature-dependent resistivity was fitted by a sum of the interference and the Bloch-Grüneisen terms, given by Eqs. (2) and (1), respectively. As seen from Fig. 1, the relative interference contribution $\Delta\rho_{\text{int}}/\rho_0$ remains unchanged after the first exposure to the ion beam. It allows one to conclude that the electron parameters also do not change between the bombardment cycles. The Debye temperature Θ_D obtained from the fitting using Eq. (1) turns out to be 370 K for all bombarded samples (Nos. 2–5). It agrees well with the previously reported data on NbC crystal.²⁹ Comparing the separated interference contribution with Eq. (3) we determined the constant $B = 1.46 \times 10^{-6} \text{ K}^{-2}$. Note, that in the pure sample (No. 1) the T^2 term is not well observed at low temperatures. Using the same fitting procedure the interference and Bloch-Grüneisen terms can be separated out (see Fig. 1). The value of the Debye temperature happened to be same as in the exposed samples, but the relative value of the interference contribution was different and $B = 1.95 \times 10^{-6} \text{ K}^{-2}$.

ELECTRON ENERGY RELAXATION RATE

We determined the inelastic electron-phonon energy relaxation rate directly from the measurements of a modulation frequency spectrum of the resistive response in a superconducting sample biased at the middle of superconducting transition, to electromagnetic radiation. The critical temperature has been suppressed by the magnetic field allowing one to perform such measurements from 1.5 K up to the film critical

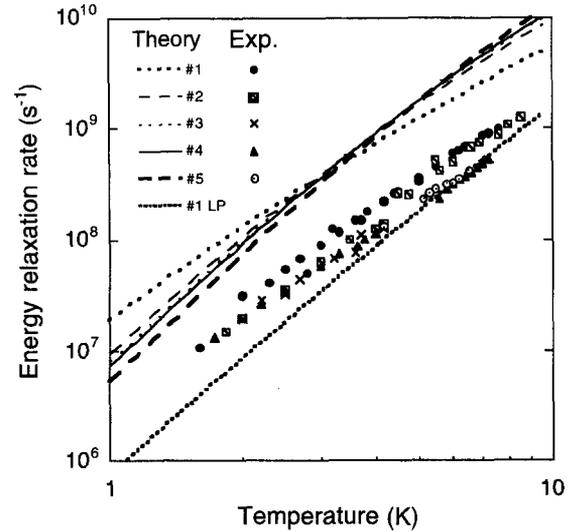


FIG. 2. Energy relaxation rate. LP is the theoretical contribution of longitudinal phonons for sample No. 1.

temperature of 10 K. The radiation power absorbed by electrons raises the electron temperature T_e and the resistance in the resistive state of a sample (see Ref. 9 for details). The frequency dependent voltage $\Delta U(f)$ develops across the sample biased with a small dc current. The relaxation rate of the resistance is determined by the electron energy relaxation rate τ_e . In the course of measurements the modulation frequency f was swept and the voltage $\Delta U(f)$ across the sample was read out. A large temperature steepness of resistance ($\approx 10^3 \text{ } \Omega/\text{K}$) in the resistive state of the NbC films and narrow resolution bandwidth ($\sim 100 \text{ Hz}$) provided high sensitivity of the measurements. It allowed us to use small bias dc current and low radiation power and avoid an overheating of the samples. In the experiment we used either two tunable backward wave oscillators (BWO's) with the wavelength 2.2 mm or a $0.83 \text{ } \mu\text{m}$ semiconductor GaAsAl laser. At frequencies $f = 0.1\text{--}10 \text{ MHz}$ a direct modulation of the voltage across the BWO's was used. At higher frequencies $f = 5\text{--}1500 \text{ MHz}$ a frequency beating of two BWO's was used. The output power of the semiconductor laser was modulated at frequencies $f = 1\text{--}1500 \text{ MHz}$ by means of amplitude modulation of the bias current through the laser diode. The measured $\Delta U(f)$ dependencies were coincidental at 2.2 mm and $0.83 \text{ } \mu\text{m}$. All curves were well fitted by the equation

$$\Delta U(f) = \Delta U(0) [1 + (2\pi\tau_e f)^2]^{-1/2}, \quad (10)$$

and the electron energy relaxation time τ_e was determined as a fitting parameter in Eq. (10). The temperature dependencies of the electron energy relaxation rate for samples Nos. 1–5 are shown in Fig. 2.

Along with the experimental data, Fig. 2 contains the theoretical calculations of the relaxation rate using Eq. (8) with constant B determined from the $R(T)$ measurements. The electron-phonon time given by Eq. (8) relates to a single-electron process. For comparison with the experiment, the calculated electron scattering rate should be averaged over the range of electron states $\sim k_B T$. The relationship between

the inelastic electron-phonon relaxation rate $\tau_{e-\varphi\text{ ph}}^{-1}$ ($\varphi = l, t$) and the energy averaged relaxation rate $\langle \tau_{e-\varphi\text{ ph}}^{-1} \rangle$, is given by¹³

$$\langle \tau_{e-\varphi\text{ ph}}^{-1} \rangle = \frac{3(n+2)\Gamma(n+2)\zeta(n+2)}{2\pi^2(2-2^{-(n-1)})\Gamma(n)\zeta(n)} \tau_{e-\varphi\text{ ph}}^{-1} \quad (11)$$

where $\Gamma(n)$ is the gamma function, n is the exponent in the power dependence $\tau_{e-\varphi\text{ ph}}^{-1} \sim T^n$.

The averaging factor used in Eq. (11) differ from those of Refs. 2 and 30. This is because in the theory which we use, the heat flow from electrons to phonons is expressed rather through the electron-phonon relaxation time taken at the Fermi surface, than through the energy averaged relaxation time.

The energy relaxation rate for all samples was due to the interaction with transverse phonons. Even in pure sample No. 1 the longitudinal phonons play an insignificant role and make a minor contribution only at high temperatures (see Fig. 2).

As seen from Fig. 2, the measured temperature dependence of the energy relaxation rate for all samples correlates well with the theory. The exponent in the theoretical dependence of the relaxation rate changes over the presented temperature range from $n=3.9$ to $n=2.8$ for the most impure sample, and from $n=3.0$ to $n=2.3$ for the pure sample. The investigated temperature interval corresponds to the intermediate region $ql \sim 1$, where the relaxation rate is characterized by a weak dependence on the electron mean free path. In order to observe the better pronounced $\tau_{e-1\text{ ph}}^{-1}(l)$ dependen-

cies, the temperatures below 1 K are required. The five times difference between the experimental and theoretical values of the relaxation rate is not surprising. Experimental data on the sound velocities and Fermi velocity in impure NbC are absent. Also, a real phonon spectrum in thin films is unknown (theory^{12,13} assumes the Debye spectrum of phonons). With given uncertainties the consistency between the two different experimental sets of data is very good.

CONCLUSION

We have demonstrated the importance of the interference between the electron-phonon and the electron-impurity interactions in the temperature-dependent conductivity and in the electron energy relaxation in NbC films. The interference T^2 term, proportional to the residual resistivity, dominates over the Bloch-Grüneisen term up to 60 K. The electron energy relaxation rates calculated using the resistivity data are in good agreement with the results of direct measurements. The results show that in a wide temperature interval the electron energy relaxation is mainly determined by the interaction of electrons with transverse phonons due to the inelastic electron scattering from vibrating impurities. The results suggest that the resistivity in impure systems can be used as a tool for probing of the electron-phonon relaxation time.

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