Evidence of interference between electron-phonon and electron-impurity scattering on the conductivity of thin metal films

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The temperature dependence of the resistivity ρ of thin gold films (thickness $d = 100$–400 Å) has been measured at $T = 30$ mK–300 K. In a wide temperature range below $\Theta_D/15$ (Θ_D is the Debye temperature) $\Delta\rho(T)/\rho$ is proportional to T^2 and does not depend on the mean free path of electrons. Experimental determinations of the dependences $\Delta\rho(T)/\rho$ in this temperature range are in good agreement with the correction to the impurity resistivity of a normal metal due to the quantum interference between the electron-phonon and electron-impurity interactions.

Recently, considerable attention has been devoted to the temperature-dependent contributions to the low-temperature resistivity of impure metals. During the past ten years the effects of weak localization (WL) and electron-electron interaction (EEI) have been studied intensively, providing an accurate description of the temperature dependence of the resistance of an impure metal with $k_F l \gg 1$ (k_F is the Fermi wave vector, l is the mean free path for electrons) at temperatures low enough to neglect all other temperature-dependent corrections to ρ. A much less clear situation exists at higher temperatures, where the scattering processes involving phonons should be taken into account. The correction $\Delta\rho_{e\text{-ph}}$ due to the electron-phonon scattering in an impure metal, derived under the assumption that the basic mechanism for electron momentum relaxation is scattering by impurities, was obtained by Altshuler:[1] for $T \ll \Theta_D$, $\Delta\rho_{e\text{-ph}}/\rho_0 \propto T^5$, as in a pure metal. The problem of interplay between electron-phonon and electron-impurity interactions has a long history. Koshino,[2] Taylor,[3] and Kagan and Zhernov,[4] predicted a correction to the resistivity of impure metals proportional to T^2 due to the inelastic scattering of electrons by vibrating impurities. The existence of this term has been confirmed by experiments on alkali-metal alloys.[5–7] However, the theory[2,4] does not predict accurately the magnitude of the term. All subsequent attempts to bring the theory and experiments into agreement did not improve the situation much.[8,9] This numerical discrepancy is not the main reason for concern. As pointed out by Reizer and Sergeev,[10] all previous theoretical works used the semiclassical approach, which is not appropriate for the description of the quantum interference between electron-impurity and electron-phonon scattering. Reizer and Sergeev[10] have shown that the interplay between these two types of scattering cannot be reduced to the two mechanisms considered previously, namely inelastic electron-impurity scattering and the lowest-order temperature-dependent correction to the zero-point motion of the lattice (the Debye-Waller factor). Several additional temperature-dependent contributions to ρ of the same order of magnitude but of different signs should coexist. Taking into account all channels of quantum interference, Reizer and Sergeev[10] have shown that for the clean limit of electron-phonon interaction ($q_l l \gg 1$, where $q_l = k_B T/\hbar u_l$ is the wave vector of a thermal phonon, propagating with velocity u_l) $\Delta\rho_{e\text{-ph-imp}}/\rho_0$ should be indeed proportional to T^2, but the coefficient of proportionality should depend in a different way on the electron and phonon parameters with respect to the previous calculations. It is worth noting that even for the Reizer-Sergeev calculations it is hard to expect *a priori* perfect agreement with experiment, because this theory was developed in the framework of a "jellium" model.

The main objective of this work is to check experimentally the predictions of the Reizer-Sergeev theory for impure metals with different intensities of impurity scattering. In order to do this it was necessary to measure the dependence of the resistivity $\rho(T)$ of an impure metal and accurately distinguish between the effects produced by electron-phonon-impurity interference and those produced by other scattering processes. For this purpose we have used thin films of noble metals, where, in contrast to the bulk samples of alkali alloys, the mean free path of electrons could be changed simply by varying the film thickness without any effect on other parameters of electron and phonon subsystems. We demonstrate that the predictions of the Reizer-Sergeev theory are in *quantitative* agreement with the results for the resistivity of thin metal films in a wide temperature range.

Gold films are a convenient system for studying the different contributions to resistivity, because the dependence $\rho(T)$ of pure gold is well known, and localization and interaction effects have been intensively studied in thin gold films.[11] To be able to compare the experimental data with the result of Reizer-Sergeev theory quantitatively, one should meet at the same time two requirements: (a) $\hbar u_l/k_B l = T_1 \leq T$ (the same as $q_l l \geq 1$), in order to avoid the Pippard ineffectiveness of electron-phonon scattering that occurs at $ql < 1$; and (b) $T \leq \Theta_D/10$, at higher temperatures electron-phonon collisions become quasielastic.

For gold $\Theta_D = 170$ K,[12] and in order to have a reasonable temperature range for observation of $\Delta\rho_{e\text{-ph-imp}}$, it is

necessary to use films with $l > 100$ Å. The thin gold films used in the experiment were evaporated from a pure Au (99.9995%) source onto glass substrates at room temperature at pressures $P = 10^{-5}$–10^{-6} mbar. The values of the thickness d and the mean free path l calculated from the relation $\rho l = 8.7 \times 10^{-12}$ Ω cm^2 (Ref. 12) were very close to one another for films with $d < 500$ Å. Thicknesses $d = 100$–400 Å were chosen sufficiently large to meet the requirement $q_l l \gg 1$ for $T > 1$ K, and, on the other hand, small enough to provide the resistance per square $R_\square = \rho/d = 1$–10 Ω/□, appropriate for the necessary accuracy of measurements. The films were photolithographically patterned in a meander line shape, the width of a strip forming the meander was 50 μm, and the sample resistance varied from 5 to 50 kΩ depending on the film thickness. The resistance was measured with an accuracy $d\rho/\rho = 10^{-6}$ with an ac resistance bridge. The measuring current was sufficiently small (10 nA) to prevent any overheating of the sample down to 0.1 K. Parameters of two samples with different l values are listed in Table I.

The temperature dependence of the resistivity was measured between 30 mK and 300 K, and the data for $T < 10$ K are shown in Figs. 1(a) and 1(b). The resistance drops with the decrease of T down to $T = T^*$ and then starts to increase again. Below 0.1 K the resistance saturates due to heating caused by the measuring current. We compared the experimental data with the theoretical dependence

$$\rho = \rho_0 + \Delta\rho_{\rm loc} + \Delta\rho_{e\text{-}e} + \Delta\rho_{e\text{-ph}} + \Delta\rho_{e\text{-ph-imp}} , \quad (1)$$

where ρ_0 is the resistance due to elastic scattering of electrons by impurities, $\Delta\rho_{e\text{-}e}$ the correction due to EEI in impure metals, and $\Delta\rho_{\rm loc}$ the WL correction. The $\rho(T)$ dependence at $T < T^*$ is mainly due to localization and interaction effects.

In order to determine the temperature dependence of $\Delta\rho_{\rm loc}$ we studied the magnetoresistance (MR) of the samples in a magnetic field $H = 0$–500 Oe perpendicular to the plane of the film. The MR data were in good agreement with the prediction of the WL theory for two-dimensional films with strong spin-orbit scattering.[11] The temperature dependence of the phase-breaking time τ_φ, extracted from these data, is shown for sample 2 in Fig. 2. For $T < 0.3$ K the temperature dependence of τ_φ becomes very slow, indicating that at these temperatures τ_φ is limited by the processes of spin-spin scattering due to a small concentration of paramagnetic impurities. The large phase-breaking length $L_\varphi = (D\tau_\varphi)^{1/2}$ (D is the diffusion constant), which is 3.5 μm at low temperatures even for our "dirtiest" sample, indicates that the relative concentration of paramagnetic impurities is extremely small (of the order of 10^{-10}). In the interval $T = 0.3$–3 K the $\tau_\varphi(T)$ dependence is mainly due to the electron-electron collisions [$\tau_{e\text{-}e} \propto T^{-1}$ in disordered films two-dimensional with respect to the EEI (Ref. 11)]. At higher temperatures the electron-phonon collisions with $\tau_{e\text{-ph}} \propto T^{-3}$ ($T = 3$–10 K) become the dominant mechanism for phase breaking. The sum of ρ_0, $\Delta\rho_{e\text{-}e}$, and $\Delta\rho_{\rm loc}$, calculated on the basis of the measured $\tau_\varphi(T)$ dependence, is shown in Fig. 1 by the center dashed line (all details of such calculations can be found in Ref. 11). The best fit of the experimental data for both samples at $T < 1$ K was obtained with values of the constant A larger than 1 in the formula for the contribution of EEI $\Delta\rho_{e\text{-}e} = A(e^2/2\pi^2\hbar)R_\square\rho \ln T$. For sample 1, $A = 1.45$, and for sample 2, $A = 1.55$. For EEI only, A should be

TABLE I. Parameters of two samples studied in this work.

Sample	d (Å)	l (Å)	$R_\square$(4 K) (Ω/□)	T_1 (K)
1	339	277	0.92	0.85
2	116	115	6.9	2.05

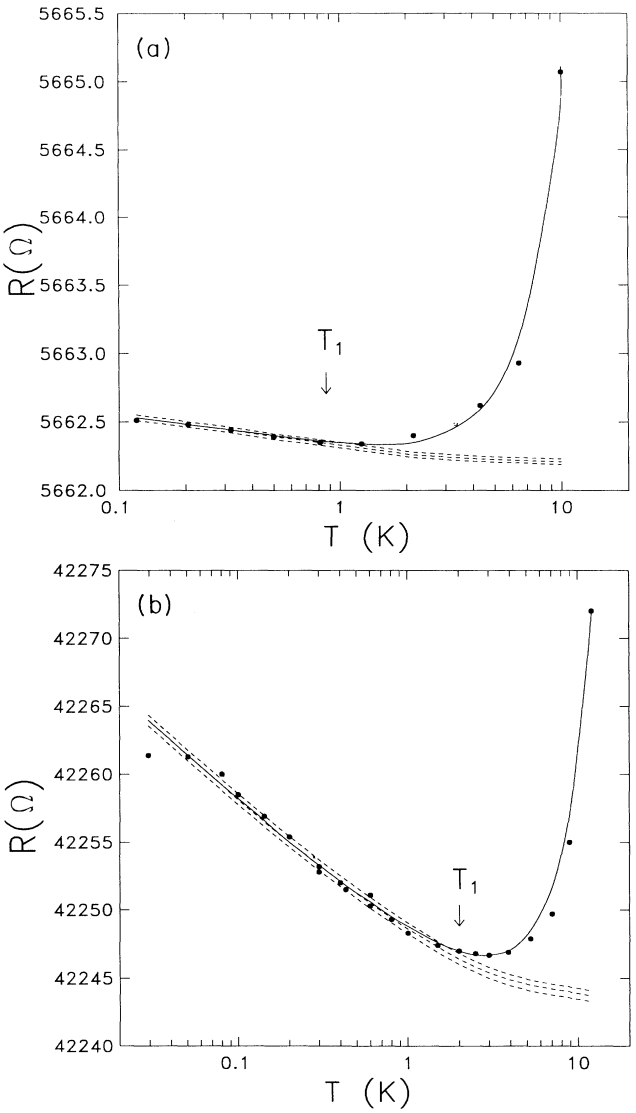

FIG. 1. Temperature dependence of resistivity of (a) sample 1 and (b) sample 2. The center dashed lines represent the best fit of $\Delta\rho_{\rm loc} + \Delta\rho_{e\text{-}e} + \rho_0$, the upper and lower dashed lines represent the uncertainty of the fit, and the solid lines represent $\Delta\rho_{\rm loc} + \Delta\rho_{e\text{-}e} + \Delta\rho_{e\text{-ph-imp}} + \rho_{e\text{-ph}} + \rho_0$.

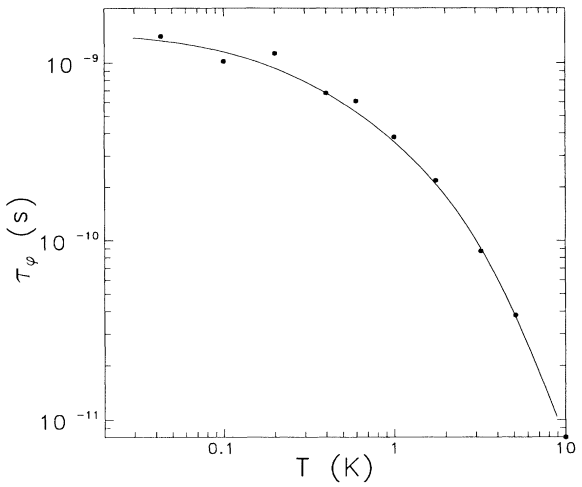

FIG. 2. Temperature dependence of τ_φ for sample 2. The solid line is $\tau_\varphi^{-1}=6.7\times10^8+2\times10^9\ T+1.3\times10^8\ T^3\ s^{-1}$ for T in K.

close to 1, and the difference could be ascribed to the Kondo effect in the samples containing a small concentration of paramagnetic impurities with a low Kondo temperature. The uncertainty in the determination of the localization and interaction corrections is represented by the upper and lower dashed lines in Figs. 1(a) and 1(b).

Now we concentrate on the temperature range $T > T^*$, where scattering processes involving phonons become dominant. We subtract the sum $\rho_0+\Delta\rho_{\rm loc}+\Delta\rho_{e\text{-}e}$ from $\rho(T)$ to get $\delta\rho(T)$ as shown in Figs. 3(a) and 3(b). The error bars shown in Figs. 3(a) and 3(b) represent the error introduced by the variation of the localization and interaction corrections between the lower and upper dashed lines in Figs. 1(a) and 1(b).

In the temperature range $T \geq T_1$ (T_1 values are listed in Table I) the phonon spectrum in the films could be considered as three dimensional ($qd \sim ql \gg 1$). For $T \ll \Theta_D$ the correction to the impurity resistivity due to the interference between electron-phonon and electron-impurity interactions is[10]

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

where u_l and u_t are the sound velocities for longitudinal and transverse phonons, p_F the Fermi momentum, ε_F the Fermi energy, and $\beta=0.5$ in the "jellium" model. Equation (2) is the result of consideration of all quantum electron-phonon-impurity interference effects. The contributions to $\Delta\rho_{e\text{-ph-imp}}$ due to the interaction of electrons with longitudinal and transverse phonons have different signs because of the quantum nature of interference and peculiarities in the screening of transverse electromagnetic fields in metals.[10] We note that the sign of $\Delta\rho_{e\text{-ph-imp}}$ for longitudinal phonons is negative in contrast to the previous calculations,[2–4,8,9] while for transverse phonons it is positive. The total correction is positive and results in an increase of the resistance with temperature because u_l is always larger than u_t. At higher temperatures ($T \geq \Theta_D/10$) if the electron-impurity scattering is still the main scattering process, $\Delta\rho_{e\text{-ph-imp}}$ would be given by[10]

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

At $T \leq \Theta_D/10$ the integral in Eq. (3) tends to $-\pi^2/3$, and Eq. (3) coincides with Eq. (2). The theoretical dependence of Eq. (3) is shown in Fig. 3 as the dashed line.

Figure 3 shows that the theory is within the experimental error in the temperature range $T_1 \leq T \leq \Theta_D/15$. This agreement was obtained with *no* fit parameters for all samples. The ratio $\delta\rho(T)/\rho$ at these temperatures does not depend on l in agreement with Eqs. (2) and (3).

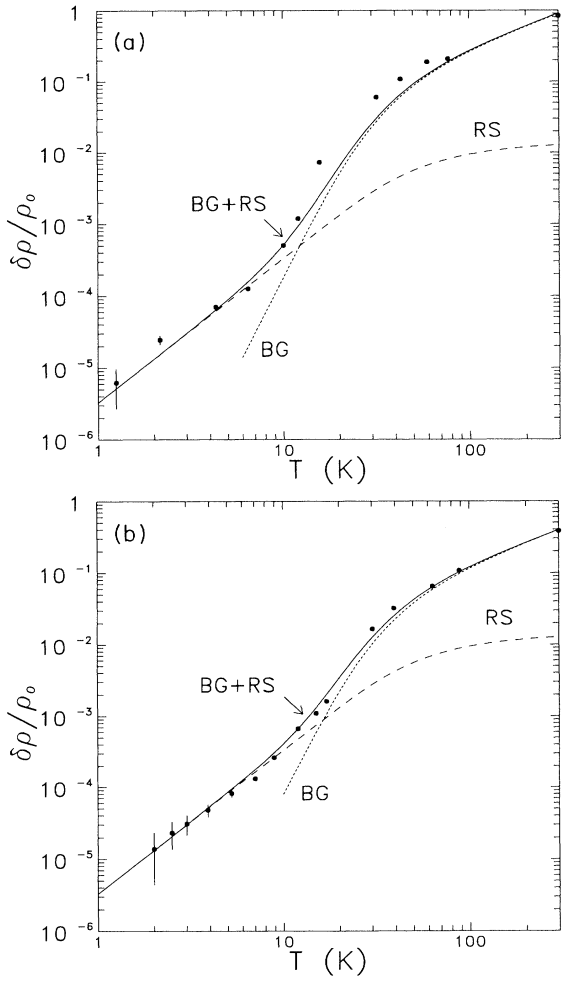

FIG. 3. Dependences of $\delta\rho/\rho_0=[\rho(T)-\rho_0-\Delta\rho_{\rm loc}(T)-\Delta\rho_{e\text{-}e}(T)]/\rho_0$ for (a) sample 1 and (b) sample 2. The dashed lines (RS) are $\Delta\rho_{e\text{-ph-imp}}(T)/\rho_0$ from Eq. (3), obtained by Reizer and Sergeev. The dotted lines represent the BG contribution to the resistivity, and the solid lines are the sum of the RS and BG terms. The error bars indicate the uncertainty of the procedure of subtraction of the WL and EEI contributions (see text).

This indicates that possible modifications of the Reizer-Sergeev result because of "real metal" effects (nonspherical Fermi surface, nonparabolic phonon spectrum, etc.), at least for a noble metal, are within the accuracy of the separation of $\Delta\rho_{e\text{-ph-imp}}$ from $\rho(T)$ in our experiment.

We can rule out confidently a significant contribution from "pure" electron-phonon interaction to $\rho(T)$ at temperatures below 10 K. Several studies on *clean* bulk gold samples showed that there are deviations from Bloch's law in that temperature range, yielding $\rho(T) \propto T^k$, k varying between 4 and 4.88.[13–18] The power law of the effect we observe (T^2) is quite different and completely obscures any such deviations of Bloch's law.

Another effect that we can safely rule out is a possible T^2 term due to finite-size effects[19,20] observed in clean alkali-metal samples. Those are classical size effects that apply only to samples with large mean free paths ($l \geq 50$ μm in Ref. 20, for example). An estimate of the contribution to the resistivity from electron-electron scattering due to umklapp processes and surface scattering of electrons,[21,22] which are also proportional to T^2, gives for our "dirty" films a value of $\delta\rho/\rho_0$ one to two orders of magnitude smaller than the predictions of Eqs. (2) and (3).

Electron-phonon scattering becomes dominant for $T \geq 20$ K, where the result of Ref. 1 is no longer valid since it was derived under the assumption that $\Delta\rho_{e\text{-ph}} < \rho_0$. In order to compare our data with the theory at high temperatures we fixed the temperature dependence of the Bloch-Grüneisen term $\rho_{e\text{-ph}}$ to our experimental data $\delta\rho(T) = \rho - \rho_0$ at room temperature. It is clear from Figs. 3(a) and 3(b) that for $T \geq 20$ K the temperature dependence of the resistivity is mainly due to electron-phonon scattering. In spite of the small values of the mean free path in our samples, for $T \geq 20$ K the values of the contribution $\delta\rho = \rho - \rho_0$ due to electron-phonon scattering are in reasonable agreement with the measurements of the resistance of ultrapure bulk gold.[13,23]

We conclude that for the samples studied, three temperature regions with three different dominant contributions to the temperature dependence of ρ can be distinguished. At low temperatures ($T < T^* = 1$–4 K depending on $R_\square$) the dependence $\rho(T)$ is mainly due to the WL and EEI effects. At $T^* \leq T \leq \Theta_D/15$, $\Delta\rho_{e\text{-ph-imp}}$ becomes the dominant temperature-dependent contribution to ρ, and for $T > \Theta_D/15$ the temperature dependence of ρ is determined by $\rho_{e\text{-ph}}$.

The minimum of ρ occurs at T^*, which can be estimated from $(d/dT)[\Delta\rho_{\rm loc}/\rho_0 + \Delta\rho_{e\text{-}e}/\rho_0 + \Delta\rho_{e\text{-ph-imp}}/\rho_0] = 0$. For a rough estimate of T^* in two-dimensional films (with respect to WL and EEI) one can use $(\Delta\rho_{\rm loc} + \Delta\rho_{e\text{-}e})/\rho_0 = A\,(e^2/2\pi^2\hbar) R_\square \ln T$ with $A = 1$. This estimate becomes more accurate in a film if the magnetic field is strong enough for suppression of the temperature dependence of the WL correction. In this case the solution of this equation can be written as

$$T^* = \left[\frac{e^2}{2\pi^2\hbar} \frac{1}{C} \frac{\varepsilon_F p_F u_l}{k_B^2} R_\square \right]^{1/2}, \tag{4}$$

where $C = (2\pi^2/3)[2(u_l/u_t)^3 + (\pi^2/16) - 1]$. For gold $T^* = (1.84 R_\square)^{1/2}$ (K $\Omega^{-1/2}$). The values of T^* calculated from Eq. (4) are in good agreement with T^* values obtained not only in this experiment, but also in previous "weak localization" experiments with much "dirtier" films with $T^* = 10$–30 K (see, for example, Ref. 11). The inequality $q_l l \gg 1$ is still valid for small l at such high T^*, but the temperature interval, where $\Delta\rho_{e\text{-ph-imp}}$ dominates, in the temperature dependence of ρ becomes negligibly small for such "dirty" films.

We would like to emphasize that the correction $\Delta\rho_{e\text{-ph-imp}}$ cannot be considered as a result of some inelastic electron scattering. The inelastic electron-phonon collisions with a rate $\tau_{e\text{-ph}}^{-1} \propto T^3$ ($q_l l > 1$) are still ineffective in momentum relaxation in the temperature range $T \leq \Theta_D/10$. The situation is similar to the electron-electron interaction in impure metals, where, for example, in the two-dimensional case $\tau_{ee} \propto T^{-1}$ in contrast to $\Delta\rho_{e\text{-}e} \propto \ln T$.[11]

It is possible to compare the previous experimental data on the observation of the dependence $\Delta\rho \propto T^2$ in alkali alloys with Eq. (2). In Refs. 5–7 the coefficient B in the dependence $\Delta\rho/\rho = BT^2$ was measured for $K_{1-x}Rb_x$ ($x = 0.005$–0.1) and $K_{1-x}Na_x$ ($x = 0.005$–0.02). The mean free path was varied by changing the composition of the alloys, making the parameters of the electrons and phonon subsystems difficult to ascertain. However, for small x one can use the parameters of pure K: $u_l = 2.08 \times 10^3$ m/s and $u_t = 1.48 \times 10^3$ m/s, calculated from the elastic constants.[24] Equation (2) predicts $B = 7.0 \times 10^{-6}$ K^{-2}, in excellent agreement with $B = 7 \times 10^{-6}$ K^{-2} obtained for K:Na. The larger experimental B value for K:Rb [$B = 8.5 \times 10^{-6}$ K^{-2} (Ref. 5)] could be ascribed to the larger concentration of Rb which has a smaller Debye temperature. We see that the Reizer-Sergeev theory is in agreement with the results of the studies on alkali metals as well as with our results in thin gold films.

To summarize, we have demonstrated that the temperature dependence of resistivity of thin gold films is proportional to T^2 in a wide temperature range. This temperature-dependent correction to the impurity resistivity is in quantitative agreement with the result of the Reizer-Sergeev theory of quantum interference between electron-phonon and electron-impurity interactions. For samples with a small electron mean free path $\Delta\rho_{e\text{-ph-imp}}$ is dominant in the wide T range from $\Theta_D/10$ down to a temperature T^* where WL and EEI effects become significant. T^* depends on the dimensionality of the sample with respect to localization and interaction effects. For three-dimensional samples T^* should be considerably smaller than for two-dimensional samples for the same l value.

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