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Inelastic phase-coherence time in thin metal films

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We obtain the inelastic phase-coherence time τ_ϕ for thin metal films above helium temperature by considering Coulomb and electron-phonon scattering contributions. In this temperature regime, we find the asymptotic expansion used so far for the electron-phonon contribution to be invalid. The resulting T dependence is *not* a power law, but mimics T^p with $p \simeq 2$ for $T = 4-20$ K. Our calculated disorder dependence is very weak. We interpret the experimental results of Bergmann and collaborators as a superposition of Coulomb and a two-dimensional electron-phonon contribution. We obtain good agreement between theory and experiment for τ_ϕ as a function of both temperature and resistivity.

At low temperature, electrons in disordered conductors can maintain phase coherence over long times. This results in interesting interference phenomena discovered by Abrahams, Anderson, Licciardello, and Ramakrishnan,[1] which are generally referred to as "weak localization." Due to the restricted phase space, they are stronger for lower dimensionality $\mathcal{D}$, and for $\mathcal{D} = 2$ and ideal interference, the resistivity ρ diverges logarithmically with decreasing temperature. In real systems, a number of mechanisms modify this behavior. Magnetic impurities lead to phase breaking, and saturation of ρ. Spin-orbit coupling changes the interference from constructive to destructive, changing the sign of the effect. An external magnetic field allows one to determine the various time scales associated with these processes, and to distinguish weak localization from the competing electron-electron (e-e) interaction effects.[2] This way one can extract the inelastic phase-coherence time τ_ϕ, which determines the limitation of interference due to inelastic scattering processes and acts as a cutoff in the theory. All this has been described in the excellent recent review by Bergmann.[3]

Careful experiments on thin metal films and semiconductor space-charge layers in the last few years[3-5] have impressively verified the weak localization ideas. For most phenomena, quantitative agreement between theory and experiment has been achieved. An important exception is the inelastic phase-coherence time. Theory[6] predicts a linear temperature dependence of the e-e contribution to τ_ϕ. Electron-phonon (e-ph) processes yield a T^4 contribution according to Schmid,[7] while Bergmann[3] argues for a T^2 law. Both e-e and e-ph contributions to τ_ϕ are predicted to depend linearly on the elastic mean free path l. Theoretically $\tau_\phi^{e\text{-}e}$ depends linearly on the film thickness d, and $\tau_\phi^{e\text{-ph}}$ is independent of d. Experimentally, the situation was inconclusive for a long time. This has recently been understood[8,9] as due to problems with film inhomogeneity, insufficient vacuum conditions, and missing or incorrect determination of the spin-orbit scattering.

Even though there is still some experimental dispute about the necessary precautions,[9] experiments agree on a linear T dependence of τ_ϕ below $T \simeq 1$ K. This is generally agreed upon to be due to e-e interaction. Between 4 and 20 K, experiments on relatively *clean* and *thick* aluminum films have been interpreted as superposition of the e-e contribution linear in T, and a T^3 term due to the three-dimensional (3D) clean-limit e-ph interaction.[10] However, experiments on thinner, more disordered, quench condensed films show $\tau_\phi^{-1} \sim T^p$ with p between 1.6 and 2.4 for various metals.[9,11-14] Attempts to explain this behavior with existing theory failed. Furthermore, contrary to theoretical expectations τ_ϕ shows very little dependence on l and d. This unsatisfactory state of affairs, which has been discussed recently by Schmid,[7] and by Peters and Bergmann,[9] is the issue we address in this paper.

In this paper, we resolve this problem[7,9] by showing that it arises mostly from a somewhat careless extraction of numbers from theory (in particular, we find that standard asymptotic expansions are invalid in the actual experimental temperature range), and that a careful theoretical evaluation yields good agreement with experiment.[9]

We start by considering the system parameters. For typical experiments,[3,9] one has $d = 50-300$ Å, $k_F l = 10-50$, $D = 3-15$ cm^2/s, and $T = 4-20$ K. Here k_F denotes the Fermi wave number, and D denotes the diffusion constant. At $T = 10$ K, the experimental τ_ϕ is typically $0.5-2 \times 10^{-11}$ s. These numbers give rise to the following observations: (i) Since $l \ll d$, the films are 3D with respect to elastic scattering; (ii) since $D\tau_\phi \gg d^2$, the films are 2D with respect to inelastic scattering; (iii) the most probable phonon wave number, $q = 2k_B T/\hbar c$, is comparable with $1/d$. Therefore, with respect to phonons, the dimensionality of the films is between 2D and 3D. We argue that for inelastic processes, the effective e-ph coupling is 2D: First, short wavelength phonons do not effectively contribute to inelastic scattering, cf. Eq. (3) below. Second, phonons propagating perpendicular to the film will either penetrate into the substrate or be reflected between the interface and the surface. In the former case they are lost; in the latter case we expect phonon attenuation to be predominantly due to surface and interface scattering rather than due to the thin layer of electrons.

According to an argument by Thouless,[15] τ_ϕ is related to the one-particle inelastic lifetime τ_i. Actually, τ_ϕ is in general different from τ_i, the difference coming from processes with small energy transfers.[16,17] This difference is particularly pronounced for the Coulomb contribution in $\mathcal{D}=2$, where $\tau_i^{e\text{-}e} \sim (T\ln T)^{-1}$,[18] while $\tau_\phi^{e\text{-}e} \sim T^{-1}$ at low temperature. We use the result of Eiler,[17] who obtained $1/T_\phi^{e\text{-}e} = c^{e\text{-}e}Ty$, where y is the solution of the equation

$$y = \frac{1}{4DN_F} - \left(\ln\frac{2}{y} - 1\right). \qquad (1)$$

Here N_F is the density of states per spin at the Fermi level. For our quasi-2D system, we have $D = k_F l/3m$, and $N_F = k_F dm/2\pi^2$. For a jellium model $c^{e\text{-}e}=1$.[17] In a realistic system, one would expect band-structure effects to affect the prefactor.

For the e-ph contribution to inelastic scattering, processes with small energy transfer are suppressed by the e-ph vertex. Consequently, one expects $\tau_\phi^{e\text{-ph}} \simeq \tau_i^{e\text{-ph}}$ in an excellent approximation, and we can confine ourselves to calculating the inelastic lifetime.

$\tau_i^{e\text{-ph}}$ is determined by the Eliashberg function α^2F

$$1/\tau_i^{e\text{-ph}} = 2\pi \int d\omega\, \alpha^2F(\omega)/\sinh(\omega/T). \qquad (2)$$

For α^2F, we repeat the calculation by Keck and Schmid[19] for $\mathcal{D}=2$. This is straightforward, and we only give the result

$$1/\tau_i^{e\text{-ph}} = c^{e\text{-ph}}(2l/m)T^3 \sum_b (d_b/c_b^3) \times \int_0^{\Theta_D/T} dx\, x^2 f_b(xt_b)/\sinh x. \qquad (3)$$

Here $d_b = k_F^3/16\pi\rho_{\text{ion}}c_b$ with ion density ρ_{ion}, and sound velocity c_b for polarization branch b. $t_b = Tl/c_b$, and Θ_D is the Debye temperature. $c^{e\text{-ph}}=1$ for a jellium model and Debye phonons. Again, deviations from these idealized model assumptions are expected to lead to $c^{e\text{-ph}} \neq 1$. The functions f_b are static electronic stress correlation functions. We calculate them by solving Boltzmann's equation in collision time approximation. For $b=L$ (longitudinal) and $b=T$ (transverse) we obtain

$$f_L(x) = 2(1+x^2)^{-1/2} \times \{1 + 2/x^2 - 1/[(1+x^2)^{1/2} - 1]\}, \qquad (4a)$$

$$f_T(x) = 8[x^2/2 - (1+x^2)^{1/2} + 1]/x^4. \qquad (4b)$$

The ion density entering Eq. (3) is that of the quasi-2D system. We determine it by the following argument. The functions $f_{L,T}$ determine not only the electron lifetime, but also the sound attenuation in $\mathcal{D}=2$. As mentioned before, the films appear to be 3D for short-wavelength phonons. Therefore we fix ρ_{ion} by demanding that for short wavelengths the sound attenuation coincide with the corresponding bulk value. This way we get slightly different effective ion densities for the longitudinal and transverse cases, viz.,

$$\rho_{\text{ion}}^L = 9\rho_{\text{ion}}^{\text{bulk}}/4k_F, \quad \rho_{\text{ion}}^T = 3\pi\rho_{\text{ion}}^{\text{bulk}}/4k_F.$$

The total inelastic phase-coherence time is now determined by Eqs. (1) and (3). With $\tau_\phi^{e\text{-ph}} \simeq \tau_i^{e\text{-ph}}$, we have

$$\tau_\phi = [1/\tau_\phi^{e\text{-}e} + 1/\tau_\phi^{e\text{-ph}}]^{-1}. \qquad (5)$$

First we consider the temperature dependence of τ_ϕ. Figures 1(a) and 1(b) show the theoretical result based on Eqs. (1), (3), and (5), together with the experimental data from Refs. 11-13.

We see that in the temperature range considered, the e-ph contribution, Eq. (3), is dominant over the e-e contribution, Eq. (1), if $c^{e\text{-}e}=c^{e\text{-ph}}=1$. For the e-ph contribution, neither of the asymptotic expansions of the functions $f_{L,T}$ in Eqs. (4) is applicable. The resulting T dependence is complicated, and so is that of $\tau_\phi^{e\text{-ph}}$. However, between 5 and 20 K it can be well approximated by a power law with an exponent around 2.5, cf. the dashed lines in Fig. 1. We have adjusted $c^{e\text{-ph}}$ in order to get agreement at high temperatures. The fact that one has to

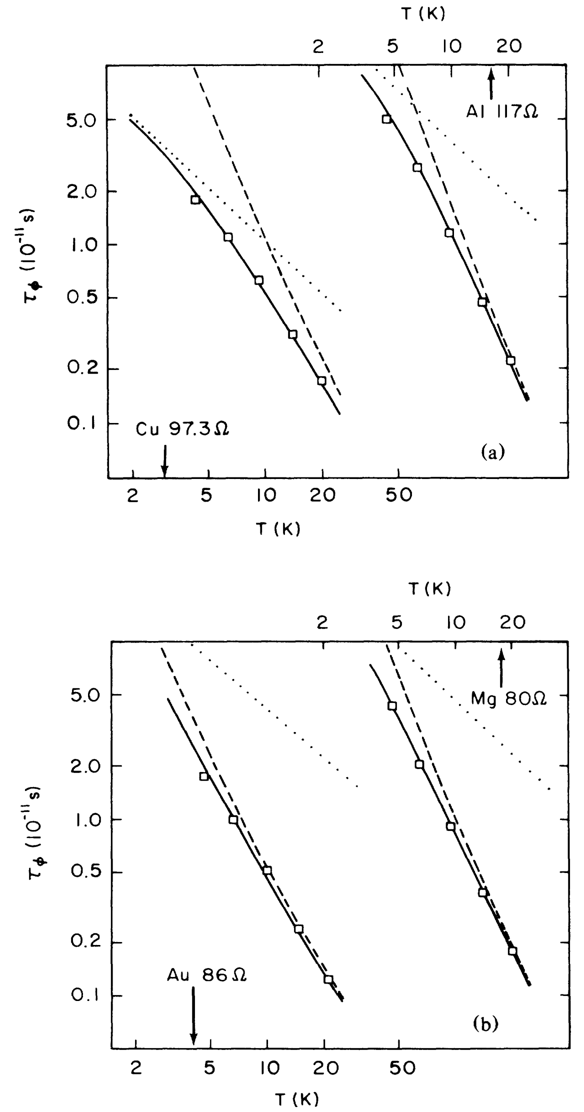

FIG. 1. Experimental data (squares) for τ_ϕ from Refs. 11 (Cu), 12 (Al), and 13 (Au, Mg). Solid lines are theoretical results for τ_ϕ, Eq. (5). Dotted and dashed lines denote theoretical results for $\tau_\phi^{e\text{-}e}$ and $\tau_\phi^{e\text{-ph}}$, respectively. For the fit parameters $c^{e\text{-}e}=4.90$, 1.20, 1.50, and 1.30; $c^{e\text{-ph}}=0.60$, 0.23, 0.55, and 0.32 have been used for Cu, Al, Au, and Mg, respectively. (See text for further explanation).

adjust the prefactor is not very surprising. In the first place, the experiment does not measure τ_ϕ directly, but rather an equivalent magnetic field H_ϕ. Within a free electron model, one has $(H_\phi \tau_\phi)_{\rm FE} = e\rho k_F m/4\pi^2$, where ρ denotes the resistivity of the film. We have used this relation to convert H_ϕ into τ_ϕ. The same free-electron model was used to derive Eq. (3). For real materials, band-structure effects will play a role. They will presumably not change the T dependence much, since this is determined mainly by long-wavelength phonons, but they will certainly change the prefactors, both via Eq. (3) and via $H_\phi \tau_\phi$. In the second place, $\tau_\phi^{e\text{-ph}}$ strongly depends on the sound velocity, like $\tau_\phi^{e\text{-ph}} \sim c^4$. Thus a 20% change in the sound velocity will lead to a change of $\tau_\phi^{e\text{-ph}}$ by a factor of 2. Unfortunately, values for the sound velocity in thin films seem not to be available, and we have used the corresponding bulk values. Under these circumstances the values chosen for $c^{e\text{-ph}}$ appear reasonable.

The deviation of the data from the theoretical $\tau_\phi^{e\text{-ph}}$ at lower temperatures is due to the Coulomb contribution. We have adjusted $c^{e\text{-}e}$ to get the best possible agreement over the whole temperature range. The values needed for Al, Au, and Mg in Fig. 1 are very reasonable. For the data on Cu from Ref. 11, we need a very large $c^{e\text{-}e}$. In this context we note that the temperature dependence found in Ref. 11 was systematically weaker than in later experiments, Refs. 12, 13. For instance, the results on Au in Ref. 11 are different from those in Ref. 13, for the best fit we needed $c^{e\text{-}e} = 5.50$, $c^{e\text{-ph}} = 0.45$. Likewise, it takes $c^{e\text{-}e} = 3.50$, $c^{e\text{-ph}} = 0.40$ to obtain a fit for Ag in Ref. 11 of the same quality as those shown in Fig. 1. We assume that the accuracy of these very difficult experiments improved over the years, and that the much smaller values of $c^{e\text{-}e}$ needed for the later experiments reflect this fact. We have also compared the theory with the results on Mg in Ref. 14. We find agreement as good as in Fig. 1(b) with $c^{e\text{-}e} = 1.90$, $c^{e\text{-ph}} = 0.32$.

Next we consider the disorder dependence of τ_ϕ. Peters and Bergmann[9] have systematically changed the resistivity of their films both by changing the thickness d, and by annealing, which changes the mean free path l. In Figs. 2, we have redrawn their results for Au and Ag (open symbols), together with the theoretical results for the corresponding parameters (full symbols). The Au films of Ref. 9 have been prepared and measured in much the same way as those of Ref. 13. We therefore choose the same values for $c^{e\text{-}e}$ and $c^{e\text{-ph}}$ as for the T dependence in Fig. 1, and compare theory and experiment *without any other fit parameter*. The result is very good. In Fig. 2(b) we show both the data from Ref. 9 (squares and circles), and those from Ref. 11 (triangles). For the latter, we have used $c^{e\text{-}e} = 3.50$, $c^{e\text{-ph}} = 0.40$, which are again identical to the values chosen to fit the temperature dependence. For the former, we have chosen parameters similar to those for Au in Fig. 2(a). It is interesting to see that for different sets of experiments by the same experimentalist on samples prepared in the same way, the experimental H_ϕ scale may differ by a factor of 2. This reassures us that our adjustment of $c^{e\text{-}e}$ and $c^{e\text{-ph}}$ from the low- and high-temperature results, respectively, does not reflect a serious problem.

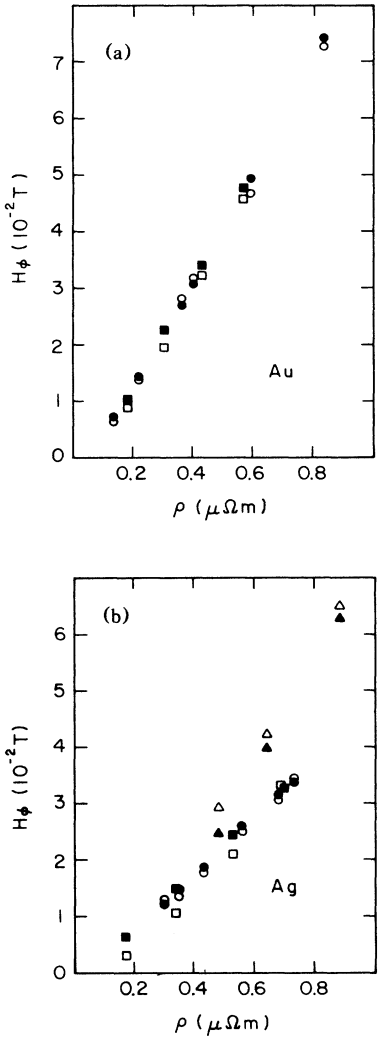

FIG. 2. Resistivity dependence of the inelastic field H_ϕ. Open symbols are data redrawn from Ref. 9 (squares and circles) and Ref. 11 [triangles in Fig. 2(b)]. Squares denote annealed samples; circles and triangles are samples measured *in situ*. Full symbols are theoretical results, Eq. (5), with parameters as given by the experimentalists. Fit parameters $c^{e\text{-}e} = 1.50$, $c^{e\text{-ph}} = 0.55$ for Au and $c^{e\text{-}e} = 3.50$, $c^{e\text{-ph}} = 0.40$ for Ag (Ref. 11), are the same as those used to fit the temperature dependence in Fig. 1. For Ag (Ref. 9), $c^{e\text{-}e} = 1.30$, $c^{e\text{-ph}} = 0.38$ have been used. See text for further explanation.

As can be seen from the figures, the theory reproduces in an excellent way the experimental finding that τ_ϕ is almost independent of both l and d. Again the reason lies in the inapplicability of asymptotic expansions in Eqs. (3). The full solution changes the linear dependence of the asymptotic solution into a very weak one. Agreement between theory and experiment is systematically somewhat worse for annealed samples (squares) than for those where ρ has been changed via changing d. The reason probably is that in the annealed situation the samples could not be kept under ultrahigh vacuum conditions.[9] We made the same comparison with the Mg data of Ref. 9. Agreement was not as good as what we find for Au and

Ag, and for annealed samples it was unsatisfactory. In this context we note that while for Ag the earlier experiments[11] are consistent with the later ones[9] after a simple rescaling, cf. Fig. 2(b), the Mg data of Refs. 13 and 14 cannot be reconciled with those of Ref. 9 by rescaling H_ϕ. Because of the small atomic number of Mg, the spin-orbit scattering time in this case cannot be measured directly. Rather, it had to be determined separately by preevaporating a fraction of a monolayer of Au.[14,9] We suspect that the experiments on Mg might be less reliable than those on the heavier metals due to this difficult procedure.

In conclusion, we have shown that a careful evaluation of the existing theory for electron-electron and electron-phonon scattering explains some hitherto mysterious and unexplained experimental results[9,11-14] for the inelastic phase-coherence time in thin, disordered, quench-condensed films. The crucial points of our analysis are (1) use of a 2D effective *e*-ph coupling; (2) full numerical evaluation of the wave-number integral determining $\tau_i^{e\text{-ph}}$, rather than using high- or low-temperature expansions. We obtain excellent agreement for the temperature dependence of all materials considered, as well as for the resistivity dependence of Au and Ag, while there are problems with the resistivity dependence of Mg. An experimental reinvestigation should clarify the latter point.

We consider the agreement between theory and experiment to be as good as can possibly be expected from a free-electron model. Reliable calculation of the two prefactors, which have been used as fit parameters in the present analysis, would require band-structure considerations as well as more experimental information about phonon properties in thin films.

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