PHYSICAL REVIEW B VOLUME 30, NUMBER 5 1 SEPTEMBER 1984

Localization and electron-interaction contributions to the magnetoresistance in three-dimensional metallic granular aluminum

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(Received 30 April 1984)

Magnetoresistance measurements have been made on three-dimensional granular aluminum specimens with room-temperature resistivities ρ_{RT} between 300 and 6000 $\mu\Omega$ cm. Four contributions, from localization and electron-interaction effects, including superconducting fluctuations, have been separated. This leads to values of the inelastic and spin-orbit scattering times $\tau_{in}(T)$ and τ_{so}. The dependences of τ_{in} on T and ρ_{RT} are in qualitative agreement with the theory of Schmid based on electron-electron interactions, but there are discrepancies in the quantitative details. The values of τ_{in} and τ_{so} are of the same order of magnitude as those obtained from measurements on two-dimensional specimens.

The recent interest in the electronic properties of disordered metals has led to a number of studies of the magnetoresistance, both as a check on the theories and as a way to measure the parameters which describe the various microscopic processes such as inelastic and spin-orbit scattering. Investigations of superconducting materials just above their transition temperature have been particularly fruitful because of the amplifying effect of the superconducting fluctuations.

The experiments have concentrated on the two-dimensional (2D) regime because of the special properties emphasized for that case by recent theories. Five experiments have been reported on 2D aluminum films, with essentially similar results, although the interpretations have differed in some details.[1-5]

The experiments which we describe here are, by contrast, on granular aluminum films whose thickness is 1 μm and which are, therefore, in the 3D regime. The scale of the granularity is sufficiently small (about 30 Å) compared to the relevant length scales that from most points of view the films can be considered to be homogeneous.[6]

The specimens are made by electron-beam evaporation onto water-cooled glass substrates. We report here on seven specimens with room-temperature resistivities ρ_{RT} from 290 to 5800 $\mu\Omega$ cm, measured from just above the transition temperature T_c to 30 K, in fields up to 9 T. Their characteristics are shown in Table I.

Near T_c the resistance first rises with increasing magnetic field as the contribution to the conductivity from superconducting fluctuations is gradually suppressed. As the field is increased further the magnetoresistance eventually becomes negative (in all the specimens and at all temperatures) because of the dominance of localization effects. In our analysis we also take into account electron-electron interaction effects which are, however, expected to be somewhat less important, especially at low fields and at temperatures near T_c. These effects become significant for the specimens with larger ρ_{RT} in high fields.[7]

TABLE I. Characteristics of the specimens.

Specimen	ρ_{RT} ($\mu\Omega$ cm)	T_c (K)	D (cm^2/s)	τ_{so} (10^{-11} s)	F
1	290	2.42	0.45	13	0 $\pm$0.5
2	520	2.37	0.37	8	-0.3 $\pm$0.4
3	940	2.33	0.25	10	-0.4 $\pm$1
4	1640	2.26	0.21	4.3	-0.4 $+0.1, -0.5$
5	2190	2.20	0.20	2.4	-0.4 $\pm$0.1
6	3530	2.15	0.18	2.0	0 $\pm$1
7	5800	1.92	0.17	1.8	0 $\pm$0.1

Although we analyze our results in terms of the most recent theories there are doubts about their range of applicability. In particular, it is not clear to what extent the calculations are appropriate for specimens for which $k_F l \lesssim 1$, where k_F is the Fermi wave vector and l the electron mean free path for elastic scattering.

In the analysis we fit the experimental magnetoconductance $\Delta\sigma = \sigma(T,H) - \sigma(T,0)$ at each temperature with the sum of four theoretical terms representing superconducting fluctuations, localization, and two interaction terms.[8-12] The only temperature-dependent fitting parameter is the inelastic scattering length L_{in} which appears by itself in the fluctuation term, and combined with the spin-orbit scattering time τ_{so} and the diffusion coefficient D in the localization term. In addition to L_{in}, τ_{so}, and D, our fitting procedure uses the coupling constant F, which was originally assumed to arise from the Hartree term in the screened Coulomb interaction between electrons.[11] The formulae which we use are as follows.

Superconducting fluctuations.[8,9]

$$\Delta\sigma = (e^2/2\pi^2\hbar)(eH/\hbar c)^{1/2}\beta f_3(4L_{in}^2 eH/\hbar c) \ ,$$

where $\beta = \beta[g(T,H)]$ is assumed to be the same function of $g(T,H)$ as it is of $g(T,0)$ in Ref. 8.[12]

$$1/g(T,H) = \ln(T_c/T) + \psi[\tfrac{1}{2}] - \psi[\tfrac{1}{2} + (DeH/2\pi ckT)] \ ,$$

where ψ is the digamma function and f_3 a function given in Ref. 9.

Localization.[9]

$$\Delta\sigma = (e^2/2\pi^2\hbar)(eH/\hbar c)^{1/2}[1.5 f_3(4D\tau_\phi^* eH/\hbar c) - 0.5 f_3(4D\tau_{in} eH/\hbar c)] \ ,$$

where $\tau_\phi^{*-1} = \tau_{in}^{-1} + \frac{4}{3}\tau_{so}^{-1}$ and $\tau_{in} = L_{in}^2/D$ is the time between inelastic collisions.

Electron-electron interactions.

Orbital effect (particle-particle channel):[9]

$$\Delta\sigma = -(e^2/2\pi^2\hbar)(eH/\hbar c)^{1/2} g(T,H)\phi_3(2DeH/\pi ckT) \ ,$$

where the function ϕ_3 is given in Ref. 9.

Zeeman splitting effect (particle-hole channel):[11]

$$\Delta\sigma = -(e^2/2\pi^2\hbar)(kT/8\hbar D)^{1/2} F g_3(2\mu_B H/kT) \ ,$$

where $2\mu_B H$ is the Zeeman splitting and the function g_3 is given in Ref. 11.

The data that are most sensitive to the choice of τ_{so} are in the temperature range near 8 K. At lower temperatures the fluctuation term (which does not contain τ_{so}) dominates, while at higher temperatures the localization term is dominated by τ_{in}. We therefore choose τ_{so} by fitting in the vicinity of 8 K with the constraint that the same value of τ_{so} must fit for the specimen at all other temperatures.

The fluctuation term is primarily determined by the function $\beta(g)$. Although the theory has been developed so far only for $H << H^* = (4ckT/\pi De)\ln(T/T_c)$,[12] in which case $g = 1/\ln(T_c/T)$, we have been able to obtain fits to higher fields by assuming $\beta = \beta(g)$ with $g = g(T,H)$ given by the formula from Ref. 12 quoted earlier. However, for fields greater than $H_{int} = \pi ckT/2De$ (Ref. 9) no set of parameters could be found to fit the data.

In Fig. 1 we show two examples of the experimental results together with the separate fitted contributions. The arrows indicate the values of H_{int} calculated from T and D. It can be seen that the theory begins to diverge from experiment near this field. Figure 1(a) is for a temperature just above T_c where the fluctuations dominate; Fig. 1(b) is for 10 K where the fluctuation contribution is smaller but still significant.

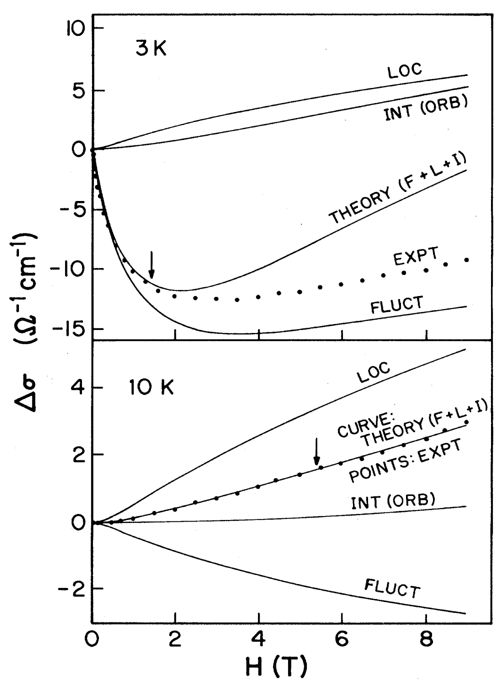

FIG. 1. Experimental values of $\Delta\sigma = \sigma(T,H) - \sigma(T,0)$ together with the theoretical contributions from superconducting fluctuations, localization, and orbital electron-interaction effects for specimen 1 at 3 K and at 10 K. (The Zeeman-splitting contribution is negligibly small here.) The arrows point to the value of H_{int} discussed in the text.

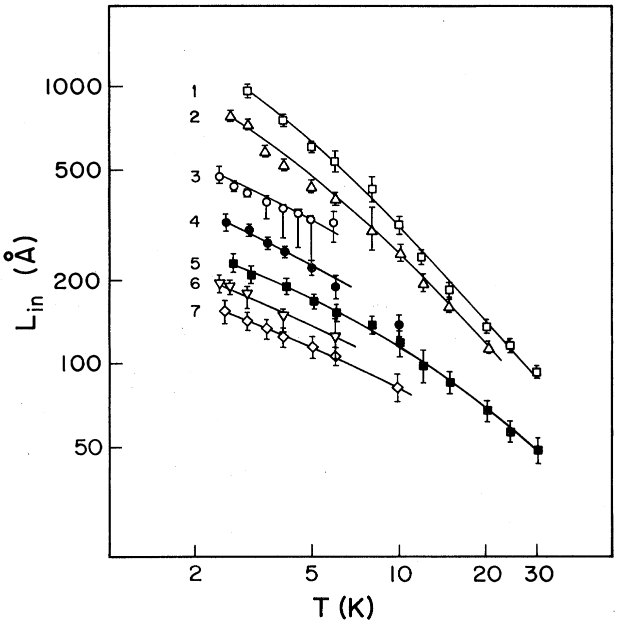

FIG. 2. Inelastic scattering length L_{in} as a function of temperature for all specimens. The lines are drawn as an aid to the eye.

Figure 2 shows the results for L_{in} as a function of T for all specimens. It may be seen that the estimated errors are smallest at the low-temperature end where the fluctuation contribution dominates, and largest in the middle region.

Figure 3 shows the inelastic scattering time $\tau_{in} = L_{in}^2/D$ which is of more interest from the theoretical point of view. It is, however, less precisely determined by our fitting procedure since it depends not only on L_{in} but also on D. One might expect the value of D to be quite well known either

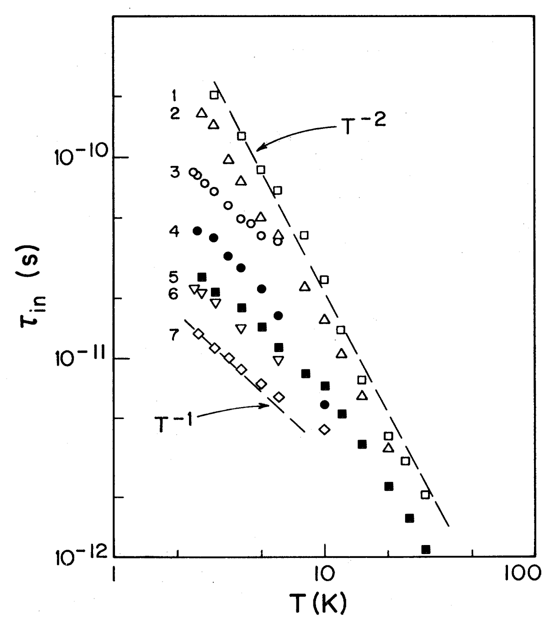

FIG. 3. Inelastic scattering time τ_{in} as a function of temperature for all specimens. The two dashed lines are proportional to T^{-2} and T^{-1}, respectively.

from the resistivity or from the slope of the critical field curve at T_c. It turns out that for our specimens the two methods lead to different values, probably because of the effect of Pauli paramagnetic limiting[13] on the critical field and perhaps also as a result of the effect of percolation.[14] Both of these effects lead to a slope of the critical field curve that is less and so a value of D that is larger than expected for a homogeneous dirty type-II superconductor. We therefore find D from the best fit to the magnetoresistance data. The fitting procedure is relatively insensitive to the values chosen for D. Acceptable values lie between the values obtained from the resistivity and the slope of the critical field curve. The ratio of the value of D found from the magnetoresistance to the one found from the critical field is 0.7 ± 0.1, i.e., in the same direction as would be produced by Pauli limiting or percolation. Because of uncertainties in the values we obtained it is not feasible to test for a possible temperature dependence of D by this method.

Figure 3 exhibits the following features: for the specimens with the lowest resistivities (1 and 2) τ_{in} is approximately proportional to T^{-2}. Specimens with higher resistivities have a slower dependence on temperature, except near the highest temperatures of the measurements where their magnitude of τ_{in} and its temperature dependence tend to the same values as for the low-resistivity specimens.

Qualitatively these observations are very suggestive of some aspects of the theory of Schmid[15] for inelastic scattering caused by the Coulomb interaction between electrons in a three-dimensional system.

The theory predicts a T^{-2} dependence in the pure limit with τ_{in} independent of resistivity ρ above a certain temperature T_x that increases with ρ. Below T_x a slower dependence on temperature is predicted and τ_{in} then depends on resistivity. At a given temperature, the values of $\ln\tau_{in}$ in Fig. 3 are then predicted to be proportional to $\ln\rho$. In Fig. 4 we show the experimentally determined values of $\ln\tau_{in}$ at 3 K plotted against $\ln\rho_{RT}$. (We use the room-temperature resistivity ρ_{RT} as the best measure of the Drude resistivity.)

Although some aspects of the theory are confirmed by the experiments, there are serious disagreements in the quantitative details. First of all, for $T < T_x$, τ_{in} is predicted to vary as $T^{-3/2}$ whereas the experimental dependence is

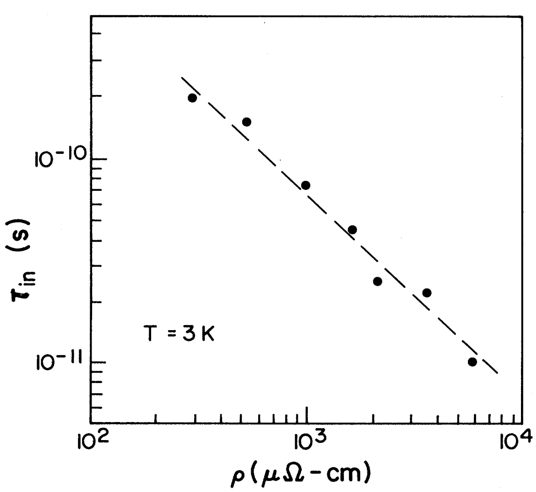

FIG. 4. Log-log graph of τ_{in} at 3 K as a function of ρ_{RT} for all specimens. The dashed line has a slope of -1.

slower. (The best-fit exponent for the five specimens 3–7 is -1.0 ± 0.2). Secondly, the slope of the line on Fig. 4 is predicted to be $-\frac{3}{2}$ but instead is close to -1. Thirdly, the constant value of $T^2\tau_{in}$ in the high-temperature, low-resistivity limit is predicted to be about $8\hbar E_F/\pi k^2$, where E_F is the Fermi energy. For aluminum $E_F = 8$ eV and so $T^2\tau_{in} = 2\times10^{-7}$ K^{2}s, compared to the experimental value of 2×10^{-9} K^{2}s. Finally, the crossover temperature is given by the theory as $T_x = nE_F\rho^3(e^2/\hbar)^3/9\pi^4k$, where n is the electron density. For aluminum $n = 2\times10^{23}$ cm^{-3} so that for a resistivity of 200 $\mu\Omega$ cm, $T_x = 2\times10^4$ K. This is evidently completely inconsistent with the experimental value which is of the order of 10 K.

It is possible that the T^{-1} dependence of τ_{in} at high resistivities and low temperatures is the result of scattering by two-level tunneling states, as reported also by Chaudhari and Habermeier.[16] In our specimens the material surrounding the aluminum grains is amorphous Al_2O_3 in which such states are expected to exist. In fact, thermal conductivity measurements[17] have shown the characteristic plateau ascribed to scattering of phonons by tunneling states.

On the other hand we cannot rule out the possibility that the quantitative details of our analysis for the higher resistivity specimens are artifacts of the application of theories which may only be valid for $k_Fl >> 1$.

We now turn to the other parameters used in our fitting to the magnetoresistance data. One is the electron interaction parameter F. The interaction contribution that depends on F is so small in the range of specimens studied here[7] that we gain little information about F although we include it in our fitting procedure. The results for F are between 0 and -0.5, with large uncertainties. (See Table I.)

The spin-orbit scattering times τ_{so} are also shown in Table I. They decrease as ρ_{RT} increases, roughly with ρ_{RT}^{-1}. Their magnitudes are between 10^{-11} and 10^{-10} s. These values span the same range as those found in 2D films.[3,4] However, for a given value of τ_{so} the resistivity of the 2D film is about two orders of magnitude smaller than our corresponding 3D film, thus supporting the view that specimen surfaces have a stronger effect than interior defects on spin-orbit scattering.[18] Because of their granular structure our specimens have internal metal-dielectric surfaces with a large total area, but these surfaces do not appear to be producing an unusually large amount of spin-orbit scattering.

We note, finally, that our results for τ_{in} are also surprisingly similar to those for 2D specimens. The values of τ_{in} from Refs. 2, 3, and 5 fall in the same range as ours on Fig. 3. Those from Ref. 4, which are for much purer specimens, are larger by up to one order of magnitude. In all cases the trend in the magnitude and temperature dependence as ρ_{RT} is changed is similar to that of our specimens. It should be noted that an essential feature of the analysis of our results is the use of formulae appropriate for the three-dimensional case. The formulas for two dimensions are quite different and could not be fitted to the experimentally measured values of $\Delta\sigma$.

We would like to thank G. Deutscher and J. M. B. Lopes dos Santos for many helpful discussions, Y. Z. Zhang for help with the experiment, and the authors of Refs. 4 and 5 for unpublished results. This research was supported by the National Science Foundation—Low Temperature Physics—Grant No. DMR-81-18062 and by the Rutgers Research Council.

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