Superconductivity and the Metal-Insulator Transition: Tuning with Spin-Orbit Scattering

T. A. Miller, M. Kunchur, Y. Z. Zhang, P. Lindenfeld, and W. L. McLean
Serin Physics Laboratory, Rutgers University, Piscataway, New Jersey 08854-0849
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A small amount of bismuth was added to granular aluminum in order to enhance spin-orbit scattering. The metal-insulator transition was found to be shifted in accord with the antilocalization effect on an Anderson transition. The threshold for superconductivity is also shifted, to the same extent, remaining, as in the undoped system, separated from the metal-insulator transition by a narrow range of composition in which the samples are metallic but not fully superconducting. The correlation of the shifts demonstrates that superconductivity is suppressed by the vicinity of the metal-insulator transition.

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The relationship between the threshold for the occurrence of superconductivity and the metal-insulator (MI) transition has received considerable attention, but remains a controversial subject with conflicting speculations and few clearcut experimental results. Theoretical predictions include superconducting transitions from the insulating state,[1-3] as well as a nonsuperconducting metallic state in the presence of strong spin-orbit scattering.[4]

In this Letter we demonstrate that the enhancement of spin-orbit scattering shifts both the MI transition and to the same extent the superconducting threshold in granular aluminum, with the preservation of a narrow range of concentration on the metallic side where the material is not fully superconducting.[5,6]

The shift in the MI transition is in accord with the antilocalization effect of spin-orbit scattering[7] on an Anderson transition.[8] Percolation, in contrast, is not affected by spin-orbit scattering.

The fact that the superconducting threshold shifts along with the MI transition demonstrates that the position of that threshold is determined, at least in this material, by the vicinity of the MI transition, and that it is the impending transition to the insulating state which inhibits superconductivity. Later in this paper we discuss the general applicability of these conclusions to disordered superconducting systems.

The specimens are made by simultaneous electron-beam evaporation from two sources (Al and Bi), in the presence of a small amount of oxygen, onto glass substrates at room temperature. The evaporation rates are separately monitored and adjusted so that the Bi is 2 at.% of the metallic aluminum. Neighboring substrates have slightly different resistivities, and a series of specimens, from metallic to insulating, can be made in a single evaporation. There is no evidence or expectation that the small amount of bismuth has a more than insignificant effect on the structure. Based on the volume occupied by the Bi a change in the grain size of more than 1% is unlikely.

The specimens were characterized by transmission electron microscopy and Rutherford backscattering. The thicknesses were measured by crystal monitors during the evaporation as well as with a profilometer subsequently. All are larger than 4500 Å so that the specimens are three dimensional. They consist of grains of aluminum about 30 Å across, separated by amorphous Al_2O_3. The solubility of Bi in Al is 0.02 at.% so that the bismuth is presumably at the grain boundaries.[9] That the properties of the specimens are nevertheless profoundly affected is evident from even a cursory examination of the results for the superconducting transition temperature, the superconducting critical field, the magnetoresistance, and the variation of the resistance with temperature.

Figure 1 shows T_c vs ρ_{RT}, where T_c is the temperature at which the resistance has fallen to half the normal-state value. Each point on the figure represents a specimen with a complete superconducting transition. With higher values of ρ_{RT} the transitions first become incom-

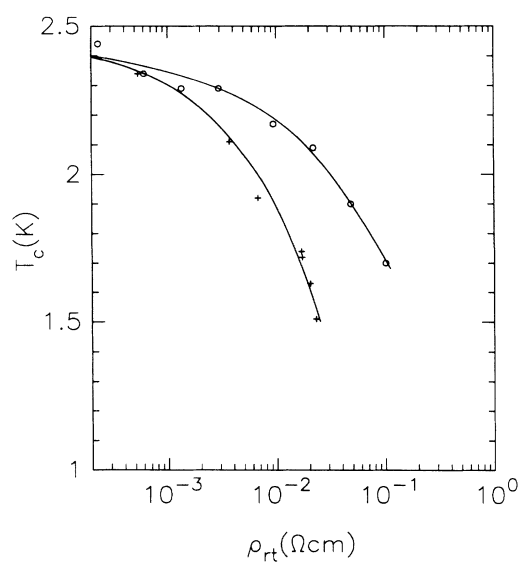

FIG. 1. T_c vs ρ_{RT}: circles, with Bi; plusses, without Bi.

plete or quasireentrant, and eventually disappear.[10]

The most compelling evidence that the added bismuth indeed results in a large amount of spin-orbit scattering comes from the enhancement of the superconducting critical field. For a specimen with Bi with a value of ρ_{RT} of 7.5×10^{-3} Ω cm, the critical field extrapolated to zero temperature, $H_{c2}(0)$, is 6.5 T, in contrast to 3.6 T for similar specimens without Bi. As we have discussed in earlier publications,[11] the critical magnetic field in high-resistivity granular aluminum is Pauli limited. Spin-orbit scattering reduces the Pauli-limiting effects[12] and therefore increases the value of $H_{c2}(0)$. From the theoretical treatment of Fulde[13] we estimate that the increase of $H_{c2}(0)$ to 6.5 T corresponds to a spin-orbit scattering time $\tau_{s.o.}$ of 7×10^{-13} s, smaller by a factor of about 15 than in similar specimens without Bi.

The normal-state magnetoresistance of metallic granular aluminum is negative because of the dominance of localization effects.[14] We have made an extensive study similar to that of Ref. 14 on the specimens with Bi, which shows that they exhibit large positive values in keeping with a shift of the MI transition[15] caused by the antilocalization effect of spin-orbit scattering. A detailed analysis of the specimen with $\rho_{RT}=7.5\times10^{-3}$ Ω cm leads to an upper limit for $\tau_{s.o.}$ of about 6×10^{-13} s, in reasonable agreement with the value from the critical field.

Finally we examine the property which gives the most direct information on the metallic character of a specimen, namely the variation of the resistivity with temperature. In granular aluminum specimens with values of ρ_{RT} greater than 10^{-3} Ω cm, the resistivity rises as the temperature is decreased, more and more steeply as the MI transition is approached. It is essentially to note that the negative slope by itself in no way indicates that a specimen is on one side or the other of the MI transition, and is only an indication of electron localization effects.

The result of adding Bi is dramatic. We observe that for the same ρ_{RT} the increase of $\rho(T)$ with lowering T is much smaller in the specimens containing Bi. We use the ratio $r=R_{4.2}/R_{RT}$ of the resistance at 4.2 K to the room-temperature resistance as a measure of the rate of change with T of the normal-state resistivity, and of the approach to the MI transition. Figure 2 shows r as a function of ρ_{RT} for both series of specimens. The figure shows that in the series with Bi, the divergence of ρ as zero temperature is approached, and therefore also the MI transition, occur at higher values of ρ_{RT} for the material with the Bi.

Figure 2 leads to the strong presumption that the MI transition for the series with Bi occurs at a value of ρ_{RT} at least 4 times as high as for the series without Bi. It is, however, well known (see, for example, Ref. 5) that the exact position of the MI transition, and the metallic or nonmetallic character of a particular specimen, can only be ascertained by measurements at sufficiently low temperatures, and in the case of superconducting specimens only by measurements in magnetic fields higher than H_{c2}.

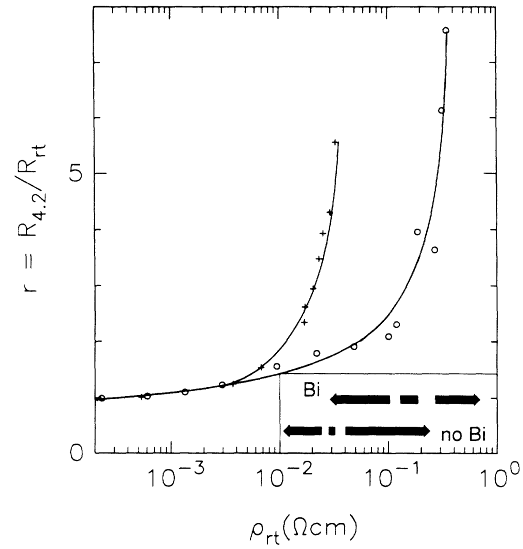

FIG. 2. r vs ρ_{RT}: circles, with Bi; plusses, without Bi. Inset: The three phases: superconducting and metallic on the left, insulating and normal on the right. The intermediate regions are metallic but without complete superconducting transitions. The regions between the phases where no specimens have been measured are left blank.

Figure 3 shows data down to 0.06 K with and without a magnetic field of 8 T on some specimens with Bi. The results confirm the conclusions of the previous paragraph and of Ref. 6, and in addition demonstrate the existence of a narrow range of values of ρ_{RT} for which the material is metallic but not completely superconducting, exactly as for specimens without Bi, but at values of ρ_{RT} about 4 times as high as in that case.

A comparison of our results with those on other materials is made difficult by the fact that few other systems have been subjected to the same detailed scrutiny as granular aluminum. In several granular and amorphous systems the MI transition and the superconducting-normal threshold appear to coincide at least approximately within the limits of the measurements. They are therefore like granular aluminum in this respect, except that the possible existence of a narrow range of composition where the specimens are metallic but not superconducting has not been investigated.[16] Systems of this type include tin-tin oxide,[8] indium-hydrogen,[17] aluminum-hydrogen,[18] mercury-xenon,[19] niobium nitride-boron nitride,[20] and indium oxide.[21]

The chief counterexamples are the amorphous systems NbSi[22] and AuSi[23] (probably also BiKr[24]), in which there are substantial ranges of composition where the specimens are metallic but not superconducting. These systems cannot, however, be considered to be merely

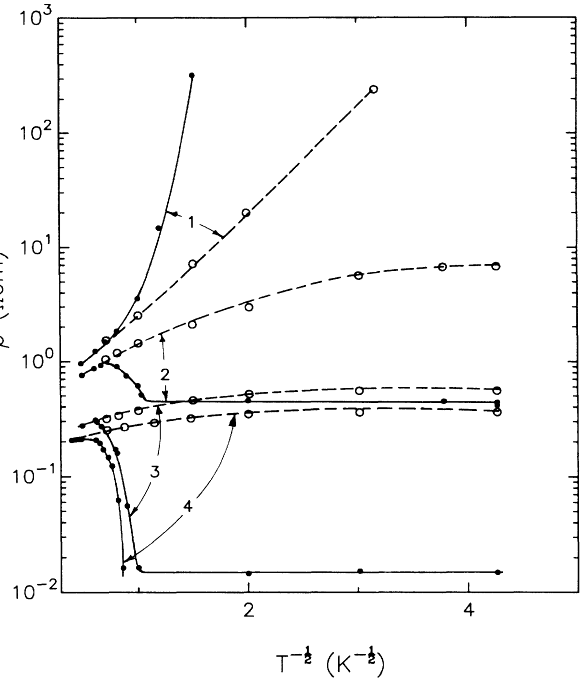

FIG. 3. ρ vs $T^{-1/2}$ for four specimens with Bi. Full lines: zero field; dashed lines: 8 T. Line 1 is insulating, lines 2 and 3 are metallic with incomplete superconducting transitions, and line 4 is metallic and superconducting.

noninteracting mixtures of metal and insulator. Certainly in the case of AuSi, where neither constituent alone is superconducting under ordinary conditions, there is evidently an interaction between the two kinds of atoms which leads to superconductivity in a range of composition, which is unrelated to the MI transition.

A further example of a system which may behave qualitatively differently is granular aluminum-germanium, which is similar to granular aluminum, but with a larger grain size (120–200 Å). As a possible consequence of the larger grain size and its effect on the competition between Josephson coupling and electrostatic charging, a superconducting transition was predicted in specimens which are insulating in the normal state.[25] This prediction was followed by the report of an experimental observation[26] of such a transition. The observation did not, however, include measurements in a magnetic field, nor measurements at low enough temperatures, and the conclusions must therefore be regarded with some reservations.

Several theoretical investigations consider aspects of the relation between superconductivity and the MI transition. Most relevant to our experiment is the examination of the role of spin-orbit scattering by Ma and Fradkin.[4] They consider only the limit of strong spin-orbit scattering and for this case predict the likelihood of a metallic but nonsuperconducting range of composition in three dimensions, however, without any quantitative predictions about its width. Our observations of a narrow range of this type, with and without Bi, can be considered to be compatible with their prediction only if the spin-orbit scattering is sufficiently strong even without the addition of Bi.[5]

Castellani, Kotliar, and Lee[27] come to the conclusion on the basis of their Fermi-liquid theory of interacting disordered systems that the nature of the MI transition may be quite different with and without spin-orbit scattering. They suggest that the diffusion constant goes to zero in the absence of spin-orbit scattering, but that in its presence it is the density of states which goes to zero. We observe only a shift in the transition without any indication that the character of the transition changes.

A number of investigations have considered the possibility of superconducting transitions in insulators, and have shown that such transitions should not be ruled out.[1-3] Our work shows that in granular aluminum such transitions apparently do not occur. Moreover, we know of no unambiguous demonstrations of their existence.

In this experiment we have changed the character of the electron localization in granular aluminum by reducing the spin-orbit scattering time. We observe a shift in the metal-insulator transition to higher values of ρ_{RT}, in keeping with the reversal of localization effects by the spin-orbit scattering.

The experiment also shows that superconductivity exists in this system in metallic specimens from a threshold close to the metal-insulator transition. The threshold shifts together with the metal-insulator transition, implying that it is the vicinity of the transition which interdicts the continued presence of superconductivity.

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