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Magnetoresistance of amorphous indium oxide films on the insulating side near the superconductor–insulator transition

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Abstract

The magnetoresistance (MR) of amorphous indium oxide films on the insulating side near the superconductor–insulator transition was measured. Variable range hopping is evident in the presence of a high enough magnetic field even in the temperature range where simple activation prevails in the absence of a magnetic field, which strongly suggests the existence of localized superconducting granules. Consequently, junction breaking between superconducting granules and pair breaking effects dominate the MR at low enough temperatures. As those effects caused by the local superconductivity on MR decrease rapidly with increasing temperature, an intrastate interaction effect becomes significant. The observed MR is fitted to a theoretical expression which includes junction breaking, pair breaking and intrastate interaction terms. The temperature dependence of the fitting parameters shows qualitative agreement with theoretical expectations.

1. Introduction

Indium oxide is a well known material of which the electrical properties are easily controlled [1, 2]. A simple heat treatment changes the stoichiometry due to diffusion of the oxygen in the material and creates some short-range order on a microscope scale. This gives rise to a change in the carrier mobility and, thus, to a change in the resistivity, which ranges from metallic to insulating. In the case of three dimensions it is generally accepted that the material loses superconductivity as it crosses over from metallic to insulating with increasing disorder. A two-dimensional superconducting system, on the other hand, is expected to have a field-induced superconductor–insulator transition (SIT) [3] at temperatures below its bulk transition temperature. Recently, such a field-induced quantum phase transition has actually been found in amorphous indium oxide films [4]. A certain scaling behaviour near the transition is confirmed.

Although various experiments in amorphous indium oxide [1, 2, 4–9] have been carried out in connection with the properties of disordered systems, a remarkable observation is that

disorder-induced granularity may exist. According to Kowal and Ovadyahu [7] the system behaves as a granular system as far as charge conduction is concerned. On the insulating side near the SIT, they observed the electrical conduction simply activated below the liquid helium temperature, followed by variable range hopping (VRH) at higher temperatures. Such an observation cannot be reconciled with any typical conduction mechanisms which lead to an Arrhenius temperature dependence. An explanation for such behaviour is possible if one assumes localized superconducting granules exist in the films. In granular materials it is well known that superconducting granules with an insulating matrix surrounding them exist at a low enough temperature. Even in a deep insulating side localized Cooper pairs can exist [10], which limit the hopping conduction due to the reduced single-electron density of states (DOS). A large negative magnetoresistance (MR) and a peculiar form of the dynamic resistance enable them (Kowal and Ovadyahu) to conclude that the observed activation is due to localized superconducting granules induced by disorder, although the essential structure is amorphous. The existence of localized superconducting granules (or droplets) and a certain length of localized clusters composed of superconducting granules gives rise to anomalous MR at low temperatures. In this paper we try to explain the MR in thin films of amorphous indium oxide which lie on the insulating side near the SIT. The observed MR is fitted to an appropriate theoretical formula which includes the effects of the destruction of local superconductivity and the effect of intrastate interaction [11, 12].

2. Experiment

The indium oxide samples were prepared by rf-sputtering targets of indium oxide onto microscope-glass substrates in a vacuum chamber with a base pressure of $(1-2) \times 10^{-6}$ Torr. During the deposition at a deposition rate of 0.5 \AA s^{-1} , an oxygen partial pressure of 1×10^{-4} Torr was maintained while the substrate temperature was kept at 40°C . A stainless-steel mask was used to obtain sample strips with thickness $d \sim 150 \text{ \AA}$. The room-temperature resistivities of the as-deposited samples were $\sim 10 \text{ \Omega cm}$, which clearly indicated that they were insulators. It was necessary to carry out heat treatment at $50-60^\circ\text{C}$ in air in order to have sample resistivity of $\sim 0.01 \text{ \Omega cm}$. According to Ovadyahu [1], indium atoms with different valences are spatially disposed in the prepared material and may exhibit varying degrees of short-range order. It is known that the heat treatment changes the stoichiometry due to diffusion of the oxygen in the material and creates some short-range order on a microscopic scale. This gives rise to a change in the carrier mobility and, thus, to a change in the resistivity. The detailed procedure for obtaining samples can be found elsewhere [1, 13].

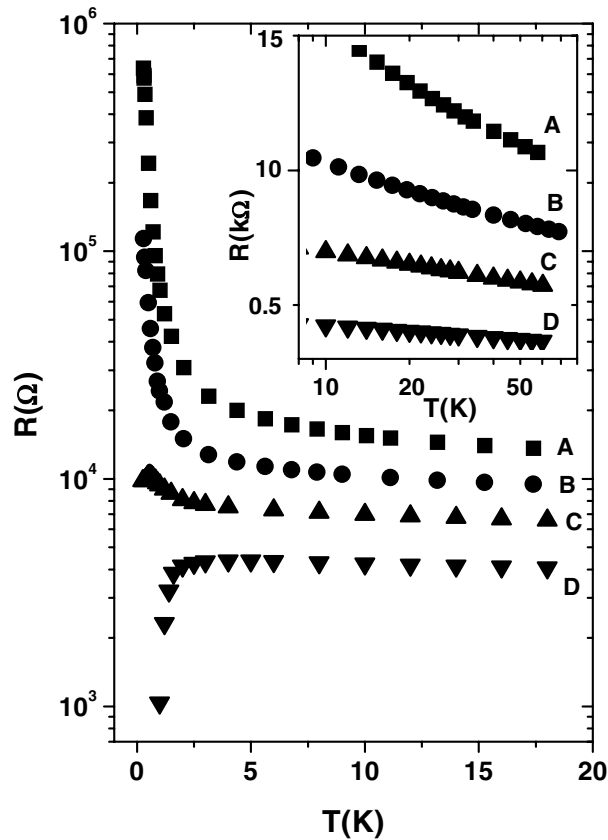
Scanning electron micrographs, as well as x-ray diffraction patterns showed that the samples obtained as described above were amorphous [13]. The resistance was measured by using a He(3) cryostat which was equipped with a superconducting magnet of 9 T maximum. A four-probe dc technique was employed. A capacitance sensor and a carbon thermometer were used to control and to read the temperature, respectively.

3. Results and discussion

Several samples were originally taken from a batch. Each sample has its own resistivity (or, sheet resistance) at room temperature (table 1) as a result of different heat treatment. The temperature dependence of four samples is depicted in figure 1. As can be seen the resistance of two samples (A and B) increases rapidly with decreasing temperature, which is a typical behaviour of an insulator. Sample D shows a superconducting transition, while

Table 1. Characteristics of the samples.

Sample	R_{\square} (300 K) (k Ω)	R_{\square} (4.2 K) (k Ω)	T_0 (K)	ξ (\AA)
A	7.89	20.1	4.91	433
B	6.40	12.1	1.2	876
C	4.84	7.47	0.92	1000
D	3.0	4.36	.	.

**Figure 1.** R against T in a semilogarithmic scale. The inset shows R against $\log T$.

sample C exposes ambiguous behaviour between insulating and superconducting states. At higher temperatures ($T > 10$ K) all the samples show a $\log T$ dependence of the resistance, which indicates that samples are effectively two dimensional (inset of figure 1). For $T \leq 4$ K, however, samples A and B follow an Arrhenius law except at low temperature ($T \leq 0.7$ K) where some degree of deviation is evident (inset of figure 2). There seems a certain temperature range where the VRH conduction of $R = R_0 \exp(T_0/T)^{1/3}$ is plausible, between 4–10 K, below and above which the Arrhenius and the $\log T$ dependences are valid, respectively (figure 2). Here T_0 is a constant temperature which is related with the localization length ξ as $T_0 \sim 1/(k_B N(0)\xi^2 d)$, where $N(0)$ is the DOS at the Fermi energy.

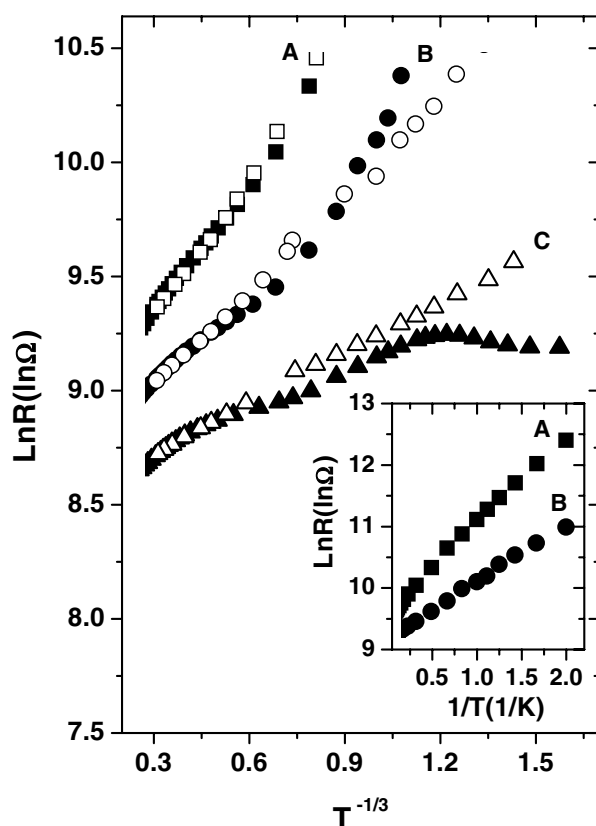


Figure 2. $\ln R$ against $T^{-1/3}$ for samples A, B and C. The open symbols represent the resistance of the sample measured in the presence of $H = 8$ T. The inset shows $\ln R$ against $1/T$ for samples A and B.

As mentioned in the introduction, the observation of a VRH regime in the temperature range below which simple activation is valid cannot be explained by typical conduction mechanisms which lead to an Arrhenius dependence. Another significant observation is that Arrhenius dependent charge conduction changes to VRH conduction when a strong enough magnetic field is applied (figure 2). Even sample C, which does not show activation at low temperatures, reveals the VRH behaviour in a magnetic field. For samples A and B, they need a thermal energy at a low enough temperature to break a localized Cooper pair for the normal conduction and, thus, the conduction should be activated. However, a strong magnetic field destroys those Cooper pairs, restores the reduced DOS and enhances the hopping conduction. Thus the main mechanism of the charge conduction changes from activation to VRH. Observations of such VRH conduction and a large negative MR, which will be shown below, strongly suggest the existence of localized superconducting granules.

In arguments on MR we presume that our amorphous indium oxide samples which are strongly localized are composed of localized superconducting granules embedded in amorphous insulating matrix. Thus one can imagine that weak links between localized superconducting granules break down in the presence of a certain strength of magnetic field while stronger links still maintain the regional superconducting clusters. The results of MR measurement for sample B are depicted in figure 3. Very similar results are also obtained for

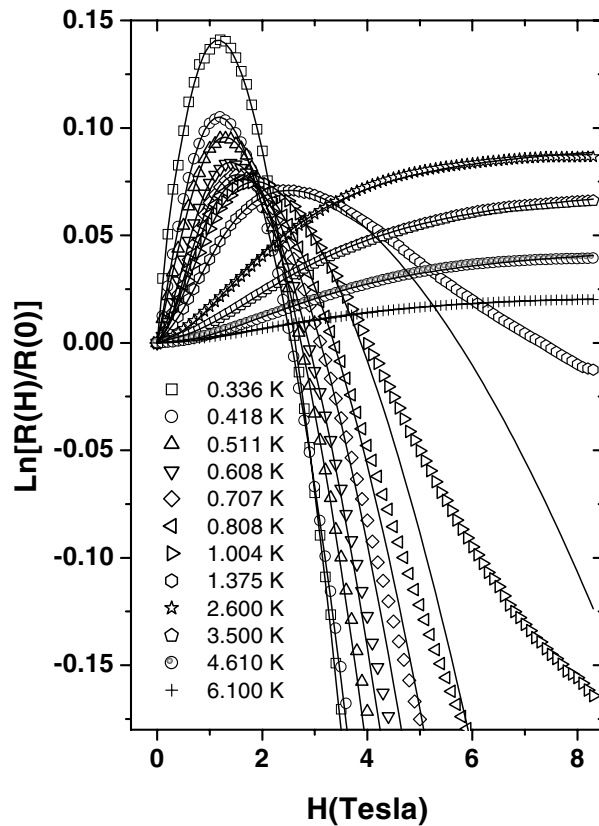


Figure 3. The MR of sample B at various temperatures. The full curves are theoretical fits using equation (1).

sample A. At low enough temperatures positive MR at low fields, which is followed by large negative MR at higher fields is conspicuous. This is more evidence for the existence of local superconducting clusters in our materials. In the case of an inhomogeneous superconductor, such as a granular one, the global superconductivity is destroyed in the presence of a magnetic field in two steps. First, weak links between the superconducting granules break down in low magnetic fields, while localized electron pairs separate into two individual electrons in relatively high fields. Consequently, positive MR at low fields can be explained as the result of junction breaking while negative MR at high fields is obviously due to the pair breaking, which results in a sharp increase in localized single-electron DOS. One may think of the initial positive MR as a result of the fluctuations in the order parameter of electron pairs. However, the superconducting fluctuation (either the Maki–Thompson term [14] or the Aslamasov–Larkin term [15]) is concerned with a conductor when a superconductor is in a normal state. Since the Cooper pairs are confined to reside within a localized region in our samples and the effect of the fluctuation must be restricted, unless very large clusters exist. Thus we believe that the superconducting fluctuation effect is not likely to be a main source of positive MR in low fields.

As the temperature increases above 1 K, effects due to the superconductivity decreases—i.e. the positive MR at low fields and the negative MR at higher fields become small at the

same time. With a further increase in temperature, another positive MR becomes significant. This second positive MR extends over a wider field range and tends to saturate at 4–5 T of magnetic field, which is typical behaviour of the spin-dependent intrastate interaction effect [11, 12]. In order to investigate how such effects on MR vary with the magnetic field and the temperature we try to fit our data to a theoretical expression considering junction breaking (JB), pair breaking (PB) and intrastate interaction (IS) effects. We use the fitting formula

$$\ln \left[\frac{R(T, H)}{R(T, 0)} \right] = \ln \left[1 + A_{JB} \left(1 - \frac{I_S(H)}{I_S(0)} \right) \right] - A_{PB} \left(\frac{H}{H_1} \right)^2 + A_{IS} \frac{H^2}{H^2 + H_{IS}^2} \quad (1)$$

The first term on the right-hand side of equation (1) represents the junction breaking effect. In the case of flow a constant dc current I_0 (using a constant current source) through a sample we simply assume $I_0 = I_n + I_S$, neglecting the capacitive effect in the resistively shunted junction model [16]. Here I_n and I_S denote normal and super currents, respectively. Since a magnetic field destroys weak links, the normal current increases with the magnetic field by the same amount as the supercurrent decreases. Assuming homogeneity¹ and thus the ohmic resistivity of samples in the normal state, i.e. $\Delta R/R(0) = \{R(H) - R(0)\}/R(0) = \Delta I_n/I_n(0) = \Delta I_S/I_S(0)$, we get the above expression (equation (1)) where $A_{JB} = I_S(0)/\{I_0 - I_S(0)\}$. We also use a field dependent Josephson tunnelling current for $I_S(H)/I_S(0)$ [17], i.e.

$$\frac{I_S(H)}{I_S(0)} \sim \frac{J_C(H)}{J_C(0)} = \left| \frac{\sin \pi H/H_0}{\pi H/H_0} \right| \quad (2)$$

which represents a Fraunhofer diffraction pattern. Here H_0 is a characteristic field determined by the size and the shape of the granule. Since a sample is composed of many junctions of random size and shape one needs to average them. We adopt the averaging of junction lengths and orientations performed by Peterson and Ekin [18], which results in

$$\frac{J_C(H)}{J_C(0)} = \frac{L_m}{\pi/2 - \Theta} \int_0^{\pi/2} dx p(x) \int_{\Theta}^{\pi/2} d\theta \left| \frac{\sin(yx \sin \theta)}{yx \sin \theta} \right| \quad (3)$$

where Θ defines the range of integration over θ , which is the angle between the external magnetic field and the normal to the plane of the junction; $x = L/L_m$; $y = \pi H/H_0$; and $p(x)$ is the probability distribution over lengths, normalized to unity. L_m is one of the parameters of the length distribution. The evaluation turns out to be insensitive to the value of Θ , which must not be far from $\pi/2$, since the least favourable orientations are those which control the current in a percolative flow. The second term on the right-hand side of equation (1) represents the pair breaking effect which is obtained from the field dependence of the energy gap in a weak field [19]. H_1 is a constant field of unit magnitude, which makes A_{PB} a dimensionless parameter. The last term consider the intrastate interaction effect on MR. Since the original expression of Kurobe and Kamimura [11] for the intrastate interaction effect is too complicated for actual use without detailed information such as localization lengths (inner and outer) and the DOS, we use a phenomenological formula proposed by Frydman and Ovadyahu [20]. H_{IS} is a characteristic field for spin alignment and is given by $H_{IS} = a_1(k_B T_0^{1/3}/\mu_B)T^{2/3}$, where a_1 is a constant of the order unity and μ_B is the Bohr magneton. A_{IS} is the saturation value of the MR and has a temperature dependence $A_{IS} = a_2(T_0/T^{1/3})$ where a_2 is another constant of order unity.

The results of the best fitting are expressed by full curves in figure 3. We put $\Theta = 80^\circ$, $H_0 = 0.44$ T and use the normal distribution for $p(x)$. The fitting is excellent at low fields

¹ As far as normal conduction is concerned, sample homogeneity should be examined in the scale of the hopping length. Scanning electron micrographs of a typical sample shows that our amorphous samples are homogeneous at the scale of 100 Å, which should not be longer than the hopping length of our sample B of which the localized length is 876 Å (table 1).

$(H \leq 4 \text{ T})$ where the expression for the pair breaking effect in equation (1) is presumably valid. To justify the fitting we examine the temperature dependence of fitting parameters (figures 4 and 5). All three parameters (A_{JB} , A_{PB} and A_{IS}) that control the magnitudes of each effect decreases with increasing temperature, which is expected theoretically. First of all, we notice the expression of Ambegaokar and Baratoff [21] for the amplitude of the tunnelling supercurrent through a Josephson weak link between two superconductors $J_S \sim \Delta(T) \tanh \Delta(T)/(2k_B T)$, where $\Delta(T)$ is a temperature dependent gap energy. According to the BCS theory in the weak coupling limit, $\Delta(T) \approx \Delta(0)(1 - T/T_C)^{1/2}$ near the critical temperature T_C and $\Delta(T) \approx \Delta(0)$ far below T_C [17]. For $T \leq T_C$ we get $J_C \sim \tanh(T_G/T)$ and

$$A_{JB} \sim \frac{J_S(0)}{J_0 - J_S(0)} \sim \frac{\alpha \tanh(T_G/T)}{1 - \alpha \tanh(T_G/T)}.$$

Here α is a constant and $T_G = \Delta(0)/2k_B$. As shown in figure 4, the best fit was obtained with $\alpha = 0.11$, $T_G = 0.20 \text{ K}$ and, thus, $\Delta(0) \sim 10^{-4} \text{ eV}$, which corresponds to a typical gap energy.

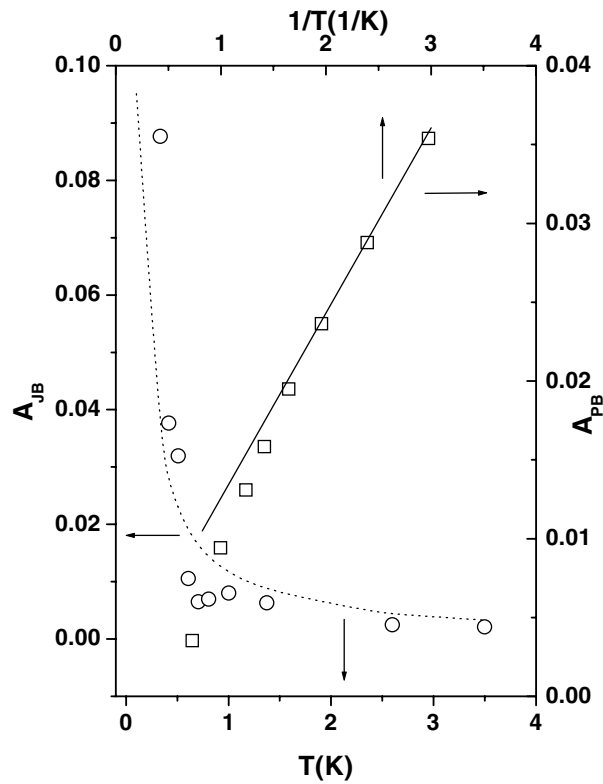


Figure 4. A_{JB} against T (circles) and A_{PB} against $1/T$ (squares). The broken curve represents $A_{JB} = [\alpha \tanh(T_G/T)]/[1 - \alpha \tanh(T_G/T)]$ with $\alpha = 0.11$ and $T_G = 0.20$. The full line is a guide for the eyes.

Apart from the effect to the MR, pair breaking gives rise to a temperature-dependent conduction of $\sim \exp[-\Delta(T)/k_B T]$. Thus we expect $A_{PB} \propto T^{-1}$ at far below the critical temperature where $\Delta T \approx \Delta(0)$. The result of our fitting is consistent with this expectation at low temperatures as shown in figure 4. At higher temperatures a square-root deviation is

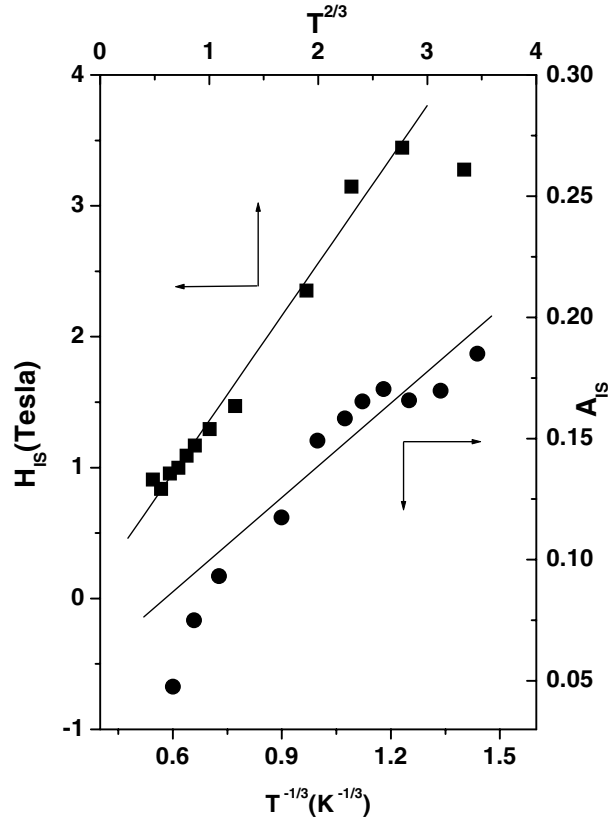


Figure 5. H_{IS} against $T^{2/3}$ and A_{IS} against $T^{-1/3}$. The full lines are guides for the eyes.

evident in the plot due to $\Delta(T) \approx \Delta(0)(1 - T/T_C)^{1/2}$. Finally, the best fit results of H_{IS} and A_{IS} are depicted in figure 5. Although data points are scattered to some extent H_{IS} shows qualitative agreement to the theoretical expectation. The slope in the plot H_{IS} against $T^{2/3}$ gives a value of $a_1(k_B T_0^{1/3}/\mu_B)$. A_{IS} also reasonably well matches the theoretical prediction, except at high temperatures where the crossover between weakly and strongly localized regimes is approached. Since A_{IS} should vanish at the crossover, the theoretical expression $A_{IS} \sim T^{-1/3}$ is no longer appropriate near the crossover. With $T_0 = 1.2$ K obtained from the slope estimation in the $\ln R$ against $T^{-1/3}$ plot of the sample (table 1), the best accordance between empirical observation and theory is obtained with $a_1 = 1.13$ and $a_2 = 0.12$.

The above analyses show that our fitting results are qualitatively in accord with theoretical predictions. One may wonder that a quantum interference (QI) effect [22] is missing in our MR analysis. QI is a well known effect which gives rise to a negative MR in the hopping regime. Detailed analysis, however, implies that the QI effect is negligible (not significant at least) in our samples near the SIT. At first we included QI effect in fitting the data but found that the effect is not compatible with other effects, especially with junction breaking and pair breaking effects. One may imagine that QI effect is suppressed by relatively larger effects caused by the superconductivity. Actually, QI effect is found to exist in more strongly localized samples that are away from the SIT. In those highly resistive samples coupling between superconducting granules is so weak that superconducting junctions are hardly formed. Thus PMR due to

junction breaking effect is strongly suppressed and NMR due to QI effect appears to be significant. NMR observation in more strongly localized samples suggests that NMR due to QI effect may exist in our samples near the SIT. Even in that case, however, the QI effect should be small compared to intrastate interaction effect as we can easily imagine from positive MR observation at higher temperatures above 1 K where local superconductivity presumably disappears. Further study may be needed to explore the basis of this QI negligibility in samples near the SIT.

4. Conclusions

the MR of amorphous indium oxide films on the insulating side near the SIT was measured. Anomalous MR and VRH conduction in the presence of a high enough magnetic field strongly suggest that the Arrhenius dependence of the resistance at low temperatures is caused by the existence of the localized superconducting granules. The observed MR is analyzed on the assumption that the films are composed of localized superconducting granules embedded in an amorphous insulating matrix. The initial positive MR and the subsequent large negative MR at low temperatures are attributed to junction breaking and pair breaking effects, respectively. As the temperature increases, effects due to the local superconductivity are suppressed and the intrastate interaction effect becomes significant.

The observed MR is fitted to a theoretical expression which includes junction breaking, pair breaking and intrastate interaction terms. The temperature dependences of the fitting parameters show qualitative agreement with theoretical expectations. A simple resistively shunted junction model explains the junction breaking term reasonably well, resulting in the estimation of the superconducting gap energy of order 10^{-4} eV. The temperature dependence of pair breaking effect can be explained by considering an activated conduction caused by localized superconducting granules. The saturation value of the MR due to the intrastate interaction effect matches with theoretical expectation at low temperatures, but shows significant deviation at relatively high temperatures where the crossover between weakly and strongly localized regimes is approached.

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