Pair tunneling in the low-temperature conductivity of the Cd–Sb alloy high-resistance state near the superconductor–insulator transition

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We report a study of the temperature dependence, down to 30 mK, of the magnetoresistance of Cd–Sb alloy in the insulating phase obtained by annealing the quenched metallic superconducting ($T_c \approx 4.5$ K) phase of the alloy. Even though the sample in this state is no longer superconducting, the observed negative magnetoresistance points to single-particle tunneling in the presence of a superconducting gap in the spectrum. At magnetic fields $B<2$ T the ratio $\alpha(T,B)=R(T,B)/R(T,B=4\ \text{T})$ is found to be maximum at a temperature of about 0.1 K. This behavior indicates a change of the conductivity mechanism from single-particle tunneling to incoherent two-particle tunneling as the temperature decreases. [S0021-3640(96)01122-X]

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The superconducting transition in the grains of a granular material can under some conditions promote insulating behavior of the material, namely, an exponential decrease of its conductivity σ_1 below the critical temperature T_c (Refs. 1 and 2):

$$\sigma_1(T) \approx \sigma_n \exp(-\Delta/T), \tag{1}$$

where σ_n is the expected conductivity at the temperature T in the absence of superconductivity in the grains, and Δ is the superconducting gap. Subject to the absence of dissipationless Josephson currents between the grains, the conductivity σ_1 is determined by the single-particle tunnel current, which is proportional to the exponentially small number of quasiparticles above the gap Δ. The Josephson currents can be suppressed for the following reasons: (i) the small average geometrical size d of the grains results in a small capacitance C between them and thus in the destruction, due to the large Coulomb energy $e^2/2C$, of long-range correlations of the phase of the superconducting wave function[3] (the Coulomb blockade); (ii) in tunnel junctions with large normal resistances there is phase slip;[4,5] (iii) a large fraction of the grains remain normal.

An exponential increase of the resistivity with decreasing temperature on account of the superconducting transition has been observed on ultrathin discontinuous films[1,2] and later on three-dimensional samples.[6,7] The phenomena can be recognized by negative magnetoresistance:[2,6] when the magnetic field exceeds the critical value B_{c2}, the superconducting gap closes and the resistance becomes $\exp(\Delta/T)$ times smaller. In principle, measuring the ratio $\beta=\sigma_n/\sigma_1$ affords the possibility of extracting the gap Δ from $\beta(T)$ and comparing it with T_c. This has been done, e.g., for metastable Ga–Sb alloy.[6] However, in most cases also the normal conductivity σ_n of the material turns out to be strongly temperature dependent because σ_n is determined by hopping. This hampers analysis of the function $\beta(T)$. Metastable Cd–Sb alloy is an example of such a material. According to measurements from Ref. 7, the ratio $\beta(T)$ in this alloy increases from 1.1 to 1.6 in the temperature interval from 1 K to 0.5 K, while the resistivity $1/\sigma_n$ becomes twice as large. It is interesting to investigate the behavior of the conductivities σ_1 and σ_n at lower temperatures. We have made low-temperature measurements of the high-resistance state of Cd–Sb alloy and here we present the results.

A sample of $Cd_{47}Sb_{53}$ alloy had a rod-like form, with all dimensions amounting to several millimeters. The sample was transformed into a metallic phase, which is a superconductor with $T_c\approx 4.5$ K, in a high-pressure chamber and quenched to liquid-nitrogen temperature. After cooling, the sample was clamped in a holder by two pairs of gold wires with pointed ends and placed into a cryostat. The low-temperature transport measurements were alternated with heating of the sample to room temperature. The heating slowly transformed the material into a disordered-insulator state. The transformation was monitored by resistance measurements and could be interrupted by returning to low temperatures. The experimental procedure is described in detail in Ref. 7.

To characterize the instantaneous intermediate states of the sample we use a parameter q determined as the ratio of the sample resistance R and R_{in} in the intermediate and initial states well above T_c, viz., at $T=6$ K:

$$q=\log(R/R_{\mathrm{in}})_{T=6\ \mathrm{K}}. \tag{2}$$

We note that R is the averaged value of resistance, because in the course of the transformation the sample becomes inhomogeneous. According to Ref. 8, below T_c the sample looks like a mixture of different ''weakly-superconducting'' elements, such as tunnel junctions, constrictions, thin wires, etc. The density and scales of these elements change during transformation, leading to evolution of the sample behavior below T_c (Refs. 6 and 7): with increasing q the transition first broadens, then becomes incomplete, and, at $q>4$, quasireentrant, i.e., at low temperatures the resistance starts to increase again. At $q\approx 5$, only a weak kink remains on the $R(T)$ curve at $T=T_c$, and the curve has a negative derivative everywhere. The value of T_c decreases slightly down to ≈ 3.8 K, while the average sample resistance R increases by five orders of magnitude.

The sample was transformed by steps from the metallic state $q=0$ to the high-resistance state $q\approx 5$ in a He^3 cryostat in a manner described in Ref. 7. In the course of this transformation the ''poorly-superconducting'' states were studied. The obtained results will be published elsewhere. After q approached a value of 5, the sample was placed in an Oxford TLM-400 dilution refrigerator with a base temperature of 25 mK. The low-temperature measurements were performed by a four-terminal lock-in technique at a frequency of 10 Hz. The ac current was equal to 1 nA and corresponded to the linear regime. The procedures for mounting the sample in the dilution refrigerator and of its dismounting require the sample to stay at room temperature at least for ten minutes. At this stage of transformation of Cd–Sb, such an exposure does not change the sample state significantly. This was checked after the sample had been returned into the He^3 cryostat.

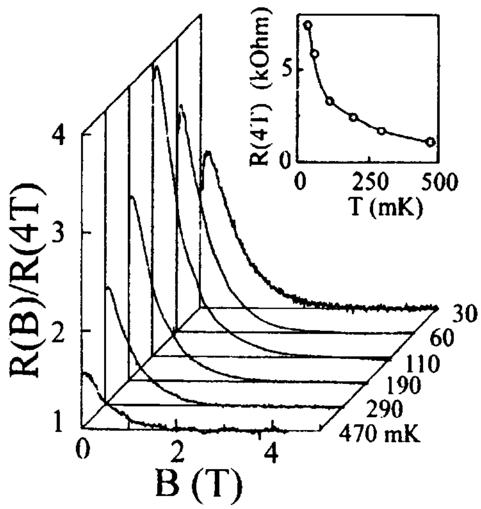

FIG. 1. Magnetoresistance of the Cd–Sb sample in a state $q \approx 5$ at different temperatures. Each curve is normalized by the value of the field-independent normal-state resistance taken at a field of 4 T. Inset: Temperature dependence of the normal-state resistance.

The experimental dependences of $\alpha(T,B)=R(T,B)/R(T,B=4\ \mathrm{T})$ on magnetic field at different temperatures are presented in Fig. 1. In the temperature range between 490 and 190 mK the value of $\alpha(T,0)$ increases with decreasing temperature, in qualitative agreement with Eq. (1). The magnetoresistance is negative and saturates ($\alpha \approx 1$) in magnetic fields exceeding 2 T. This corresponds to what has been observed previously at higher temperatures.[7] However, in the low-temperature limit the behavior of α changes drastically (Fig. 1). In weak magnetic fields there appears an initial increase on the dependence $\alpha(B)$, so that α is maximum at $B \approx 0.1$ T. At a fixed magnetic field $B < 2$ T the value of α achieves a maximum at a temperature of about 100 mK and then decreases on further lowering of the temperature. As is seen from the inset to Fig. 1, the temperature dependence of the normal-state resistance is monotonic in the range of temperatures used.

One can expect that at least in strong magnetic fields the sample resistance should follow one of the activation laws typical of the hopping process. Figure 2 shows a logarithmic plot of the conductance at $B=0$ and $B=6$ T as a function of $T^{-1/4}$, which corresponds to the slowest of the activation dependences—the Mott law. As is seen from the figure, at low temperatures both functions deviate from the Mott law and tend to saturate.

Let us discuss these results. The observed decrease of the ratio α with decreasing temperature unambiguously indicates that at low temperatures the conductivity σ_1, which originates from single-particle tunneling, is shunted by conductivity σ_2 of another kind:

$$\sigma = \sigma_1(T) + \sigma_2 . \tag{3}$$

We believe that σ_2 is due to incoherent pair tunneling. Indeed, the single-particle tun-

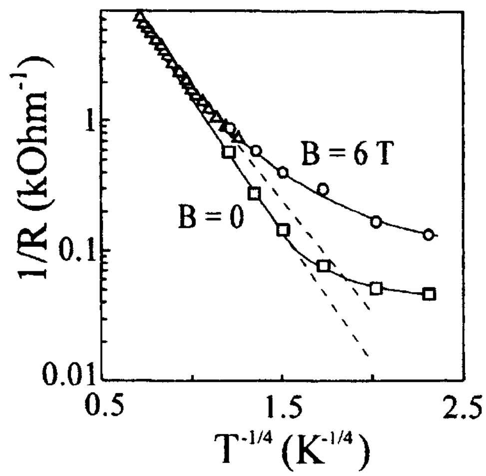

FIG. 2. Change of the sample conductance with temperature. The low-temperature data (circles and squares) were obtained using a dilution refrigerator and the remaining data (triangles) were measured in an experiment on a He^3 cryostat. The dashed lines fit the high-temperature points.

neling current i_1 is described in the first-order approximation by barrier transparency t: $i_1 \propto t\ \exp(-\Delta/T)$. It is proportional to the product of two small factors, one of which is temperature dependent. Since the Cooper pairs are at the Fermi level, the two electrons forming a pair do not need to be excited above the gap for tunneling simultaneously. Hence $i_2 \propto t^2$, where the exponential temperature-dependent factor is absent. When the temperature is sufficiently low, so that

$$t > \exp(-\Delta/T), \quad \text{i.e.,} \quad T < \Delta/|\ln t|, \tag{4}$$

the single-particle current is frozen out, and the two-particle tunneling comes into play.

In the second-order approximation, the two-particle contribution to the tunnel current of a junction between two different superconductors (SIS junction) has been calculated in Ref. 9. Given an SIS junction with gaps Δ and Δ_1, the two-particle current is significant above the threshold voltage $eV = \min(\Delta, \Delta_1)$. For tunneling into a normal metal (SIN junction, $\Delta_1 = 0$) the threshold is absent. Therefore the resistance of an SIN junction is finite at zero temperature.

Let us consider percolation on a simple cubic lattice (critical concentration $X^{(s)} = 0.31$ for site percolation and $X^{(b)} = 0.25$ for bond percolation[10]) with concentrations $x_n < X^{(s)}$ of normal (N) grains and $x_s = 1 - x_n$ of superconducting (S) grains in the sites of the lattice. The resistance of the lattice is determined by three types of contacts (bonds): SS, with concentration x_s^2; SN, with concentration $2x_s x_n$; and NN, with concentration x_n^2. According to Ref. 7, in magnetic fields above 0.1 T the Josephson currents, if any, through SS bonds are suppressed, and so at relatively high temperatures all the SS and SN junctions are in the σ_1 regime. When the temperature becomes lower than the threshold (4), the SS junctions remain in the poor σ_1 regime, so that the conductance is governed by the network of SN junctions. Their concentration may be rather large even at small x_n; for instance, when $x_n = 0.15$ (half of the critical value $X^{(s)}$), the concentration x_{sn} exceeds 0.25, which is sufficient for percolation. For interpreting the maximum on the experimental dependence $R(B)$ at low temperatures one has to suggest that in fields below 0.1 T the Josephson currents through some SS bonds are not negligible. Still, it is difficult to explain why the region of positive magnetoresistance shrinks at high temperatures (Fig. 1).

Obviously, the line of reasoning above implies that the size of grains is macroscopic, i.e., large compared to the relevant lengths. One of them is the coherence length ξ, which can be easily estimated. The characteristic field B_{ch} in superconductivity is related to the characteristic length λ_{ch} by the expression

$$B_{\mathrm{ch}} = \Phi_0/2\pi\lambda_{\mathrm{ch}}^2, \quad \Phi_0 = \pi c\hbar/e. \tag{5}$$

If the field B_{c2} is in question, then, depending on the relation between the coherence length ξ and the mean free path ℓ, the length λ_{ch} is written as[11]

$$\lambda_{\mathrm{ch}} = \xi, \quad \ell \gg \xi \ \text{(pure limit)},$$

$$\lambda_{\mathrm{ch}} = (\xi\ell)^{1/2}, \quad \ell \ll \xi \ \text{(dirty limit)}. \tag{6}$$

According to the experimental data, a field of 2 T destroys the remnants of the superconductivity and should be regarded as B_{c2}. Then, from the above relation in the dirty limit we get an estimate for the coherence length: $\xi \geqslant 120$ Å.

The other relevant length follows from a comparison of the size quantization $\delta\varepsilon \approx (g_F d^3)^{-1}$ (here g_F is the density of states at the Fermi level) and Δ: a separate grain cannot remain superconducting if $\Delta < \delta\varepsilon$. The critical grain size d_c proves to be even smaller than ξ.

Experimental estimates of the length d which determines the randomness of the structure are rather uncertain. Insensitivity of the field B_{c2} to the transformation parameter q implies that $d > \xi$. As an alternative, the authors of Ref. 8 argue that upon transformation of the sample to a high-resistance state the length d decreases until any macroscopic structure vanishes. Below we will show that the conclusion about the change of the conductivity mechanism from single-particle tunneling to incoherent two-particle tunneling is valid also at grain sizes smaller than d_c in terms of the concept of localized Cooper pairs in a quasihomogeneous disordered material.[12]

If there exists a field-dependent attractive interaction between two electrons localized on different centers at distances smaller than ξ, they can be regarded as a localized Cooper pair.[13] For an electron of a pair to hop, it has to ''pay off'' the binding energy Δ. However, two interacting electrons remain bound if their hops are correlated.[14] If the Cooper interaction fluctuates depending on the mutual disposition of electron sites, the binding energy Δ_1 in the final state may differ from that in the initial state. The final state can be even unbound (see Fig. 3). The energies of states 1_0 and 2_0 are well above the Fermi level if at least one of these states is empty. When both states are occupied they descend, due to the Cooper interaction, toward the Fermi level to energies 1_i and 2_i. The final state energies 1_f and 2_f do not depend on occupation numbers if either the barrier

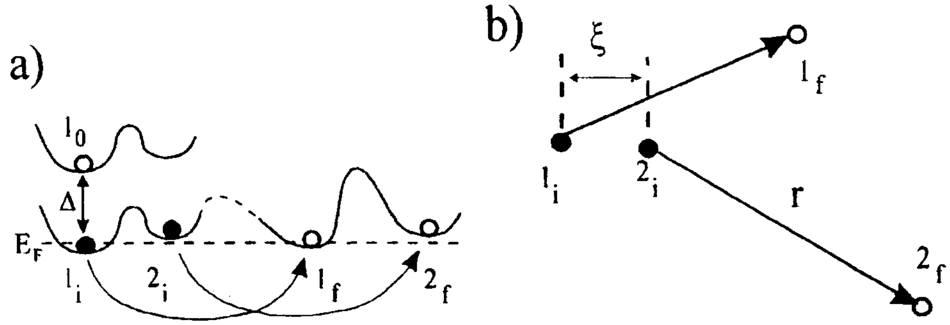

FIG. 3. Two-electron hop from interacting states 1_i and 2_i to noninteracting states 1_f and 2_f in the energy scheme (a) and geometrical scheme (b). If only one of these electrons hopped the other would have to increase its energy by Δ.

between the states is high (Fig. 3a) or they are far from one another (Fig. 3b).

Within the localized Cooper pair model, the appearance of a maximum on $R(H)$ at ≈ 0.1 T at low temperatures can be explained by reduction of the probability of pair hopping with magnetic field, which gives rise to an initial increase of the resistance R in weak magnetic fields. In this connection, we shall revisit the expression (5) that compares the magnetic flux through an area $\lambda_{\rm ch}^2$ with the flux quantum Φ_0. This relation is met not only in superconductivity but also in the theories of weak localization and of interference in hopping. If pair hops are coherent, the magnetic flux through the characteristic area $\approx \xi r$ (r is the hopping distance) should affect the wave function of the final state of an electron pair. Thus, assuming that coherent pair hops are present, one can account for the positive magnetoresistance observed in weak fields. However, there is no theory yet to make comparison with.

The tendency to saturation of the normal state resistance at low temperatures (Fig. 2) points to the existence of a shunting conductivity (cf. Ref. 15) which may, e.g., be due to resonant tunneling of localized electrons.

In summary, we have investigated the temperature and magnetic field dependence of the resistance of the metastable Cd–Sb alloy transformed into a high-resistance state. It has been established that below T_c the behavior of the resistance is determined by the competition between single-particle tunneling and incoherent two-particle tunneling. Considering the two limiting cases of macroscopic grains in the sample and of a quasi-homogeneous material in terms of localized Cooper pairs, we argue that regardless of the grain size, the incoherent pair tunneling is dominant in the low-temperature limit.

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[1] R. C. Dynes, J. P. Garno, and J. M. Rowell, Phys. Rev. Lett. **40**, 479 (1978).
[2] C. J. Adkins, J. M. D. Thomas, and M. W. Young, J. Phys. C **13**, 3427 (1980).
[3] K. B. Efetov, Zh. Éksp. Teor. Fiz. **78**, 2017 (1980) [Sov. Phys. JETP **51**, 1015 (1980)].
[4] A. Schmid, Phys. Rev. Lett. **51**, 1506 (1983).
[5] M. P. A. Fisher, Phys. Rev. Lett. **57**, 885 (1986).
[6] V. F. Gantmakher, V. N. Zverev, and V. M. Teplnskii *et al.*, Zh. Éksp. Teor. Fiz. **104**, 3217 (1993) [JETP **77**, 513 (1993)].
[7] V. F. Gantmakher, V. N. Zverev, V. M. Teplinskii, and O. I. Barkalov, Zh. Éksp. Teor. Fiz. **105**, 423 (1994) [JETP **78**, 226 (1994)].
[8] V. F. Gantmakher, S. E. Esipov, and V. M. Teplinskii, Zh. Éksp. Teor. Fiz. **97**, 373 (1990) [Sov. Phys. JETP **70**, 211 (1990)].
[9] Yu. N. Ovchinnikov, R. Christiano, and C. Nappi, Low Temp. Phys. **99**, 81 (1995) [*sic*].
[10] D. Stauffer, *Introduction to Percolation Theory*, Taylor and Francis, London, (1985).
[11] A. A. Abrikosov, *Fundamentals of the Theory of Metals*, North-Holland, Amsterdam, (1988).
[12] E. S. Soerensen, M. Wallin, S. M. Girvin, and A. P. Young, Phys. Rev. Lett. **69**, 828 (1992).
[13] V. F. Gantmakher, M. V. Golubkov, J. G. S. Lok, and A. K. Geim, Zh. Éksp. Teor. Fiz. **109**, 1765 (1996) [JETP **82**, 951 (1996)].
[14] Y. Imry, Europhys. Lett. **30**, 405 (1995).
[15] I. V. Lerner and Y. Imry, Europhys. Lett. **29**, 49 (1995).

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