4-24-2006

Negative magnetoresistance, negative electroresistance, and metallic behavior on the insulating side of the two-dimensional superconductor-insulator transition in granular Pb films

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(Received 24 June 2005; revised manuscript received 21 February 2006; published 24 April 2006)

Granular Pb thin films on the insulating side of the two-dimensional superconductor-insulator transition are observed to exhibit a large negative magnetoresistance and electroresistance (change in resistance with electric field) at low temperatures. At high measurement voltages and low temperatures, the film resistances become temperature independent creating a "metallic" state. These phenomena are explained as manifestations of transport due to intergranular quasiparticle tunneling. This explanation might also provide insights into the similar behavior observed in other superconductors.

DOI: 10.1103/PhysRevB.73.134516 PACS number(s): 74.78.−w, 74.25.Fy, 74.25.Ha

The puzzling behavior of underdoped high temperature superconductors (HTSC) and ultrathin films of conventional superconductors near a superconductor to insulator transition (SIT) has sparked new interest in the properties of inhomogeneous superconductors. The appearance of a pseudogap,[1] spatial variations in the tunneling density of states,[2] and evidence of vortices at temperatures far above T_c[3,4] suggest a model where Cooper pairs are formed well above T_c. In this model, these "preformed" pairs develop long range phase coherence at T_c.[5] Structurally "homogeneous" systems near the SIT exhibit properties that suggest the development of similar inhomogeneities. For example, near the magnetic field tuned SIT, hysteretic behavior,[6,7] metallic conduction at low fields,[8] and negative magnetoresistance at high magnetic fields,[9,10] suggest the existence of "puddles" of Cooper pairs that are not phase coherent between puddles.

In granular films of superconducting materials near an SIT such inhomogeneities are known to exist. Islands with well developed order parameter amplitudes (i.e., high densities of Cooper pairs) defined by the connections between grains can become phase coherent depending on the interisland coupling.[11] As such, their properties can serve as a model[12] for descriptions of inhomogeneous behavior in more complex systems including high T_c superconductors,[13,14] indium oxide,[9,15] and "homogenous" films[16] near the SIT. In this paper, we present measurements and compare with previous results on granular Pb films near their SIT that provide electrical transport "signatures" of a system with islands of Cooper pairs.

The superconductor-insulator transition SIT has been observed in granular films for a range of elements (Pb, Sn, Ga, Bi, Al) quench-condensed onto insulating, inert substrates.[11,17–22] Qualitatively, these films are not electrically continuous below a critical mass per unit area that corresponds to an average film thickness of many atomic layers. This observation has been used to support the idea that these films have a granular or clustered morphology. In addition, direct scanning tunneling microscope imaging of Pb films prepared in this way unambiguously confirms this conclusion.[23] Transport measurements have revealed extremely large electrical resistance in films deposited just above this critical areal mass density, consistent with transport due to intergranular tunneling. In most cases below a temperature near the bulk T_c of the material, resistance versus temperature $R(T)$ curves exhibit a kink. This kink suggests that superconductivity is present even in the thinnest samples, and the change in slope with temperature reflects a change in the intergrain tunneling properties due to that superconductivity. Direct tunneling measurements into granular Pb films have shown that even in highly resistive films there is superconductivity in the grains with nearly bulk values for the T_c and the energy gap Δ and the signature of a BCS density of states.[24]

Here, Pb films were evaporated onto fire-polished glass or quartz substrates held at temperatures between 8 and 10 K in an ultrahigh vacuum environment. Film average thicknesses were monitored with a quartz crystal microbalance to control the evaporation. Resistance and current-voltage I-V measurements were performed *in situ* using standard four-wire techniques. Resistance reported here was typically determined from the linear low-current regimes of the I-V curves, however, in some cases (described below in the text) constant DC currents were applied and the resultant voltage drop was measured. Perpendicular magnetic fields up to 8 T were applied using a superconducting magnet solenoid.

Figure 1 shows typical $R(T)$ curves for an incrementally deposited Pb film (as adapted from Ref. 25). Care was taken to derive resistance values from the linear low-current region of the I-V curves. When this procedure is followed in four different laboratories of the authors, there is no evidence of temperature independent resistance in the low temperature limit ("metallic" behavior). The sheet resistance of films on the insulating side of the SIT grows as $\exp[-(T_0/T)^{1/2}]$ at low temperatures similar to earlier work.[17] The onset of this rapid rise coincides with the T_c of the individual grains and thus the opening of the superconducting energy gap in the density of states at the Fermi energy.[24]

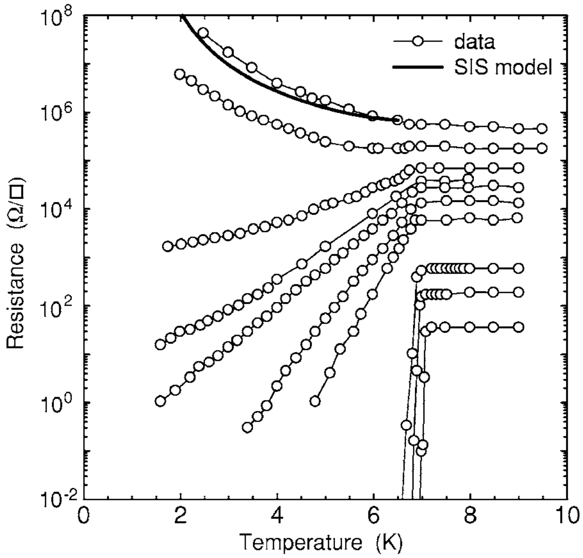

FIG. 1. Sheet resistance vs temperature for a typical series of quench-evaporated Pb films. Note the gradual broadening of $R(T)$ with increasing $R_N=R(8\text{ K})$ that eventually leads to insulating behavior. $R(T)$ for the single junction model calculation with $R_N = 1.36\text{ M}\Omega$ (see text) is shown as the solid curve.

In addition to the experimental data, we have also plotted in Fig. 1 results for a simple model. *If* the transport in these films is dominated by intergranular tunneling *and* the grains are known to be superconducting, then the high-resistance "insulator" regime should be dominated by quasiparticle superconductor-insulator-superconductor (SIS) tunneling. The resistance of a random array of such SIS junctions should have the same temperature dependence as one SIS junction. Furthermore, if the superconductive properties of the sample are bulklike,[24] the temperature dependence can be calculated with *no* free parameters except the overall resistance scale. We have calculated this temperature dependence using the zero-bias resistance of a Pb-Pb SIS tunnel junction ignoring the Josephson effect. We believe that Josephson tunneling will be suppressed almost entirely due to phase fluctuations in this high resistance regime.[26] The integral calculations were performed with MATHEMATICA for $I(V)$ using the standard form for tunneling between two identical superconductors[27]

$$I(V) = \frac{G_n}{e}\int_{-\infty}^{\infty} N_S(E'-eV)N_S(E')[f(E'-eV)-f(E')]dE'$$

assuming a BCS density of states[28]

$$\frac{N_s(E)}{N_n(0)} = \mathrm{Re}\left\{\frac{E-i\Gamma}{[(E-i\Gamma)^2-\Delta^2]^{1/2}}\right\}$$

with the gap $\Delta(T)$ from a strong coupling calculation,[29] $N_n(0)$ the normal density of states, and the parameter Γ being a phenomenological broadening term. Γ was set to be 0.001 meV, however, large variations in Γ had negligible effects on our results. The normal state tunnel resistance $R_n = 1/G_n$ was chosen to be 1.36 MΩ for the curve as plotted in Fig. 1 in order for the 6.5 K resistance of the model to match the experimental data at the same point. It is important to note that there are no other free parameters, and except for matching the calculated curve to the data at one point, this is *not* a fit. The good agreement between the temperature dependence of this extremely simple model and the temperature dependence of our data strongly supports the validity of SIS tunneling as the dominant mechanism on the insulating side of the SIT. A similar result was obtained for an array model of quench condensed Sn films.[19]

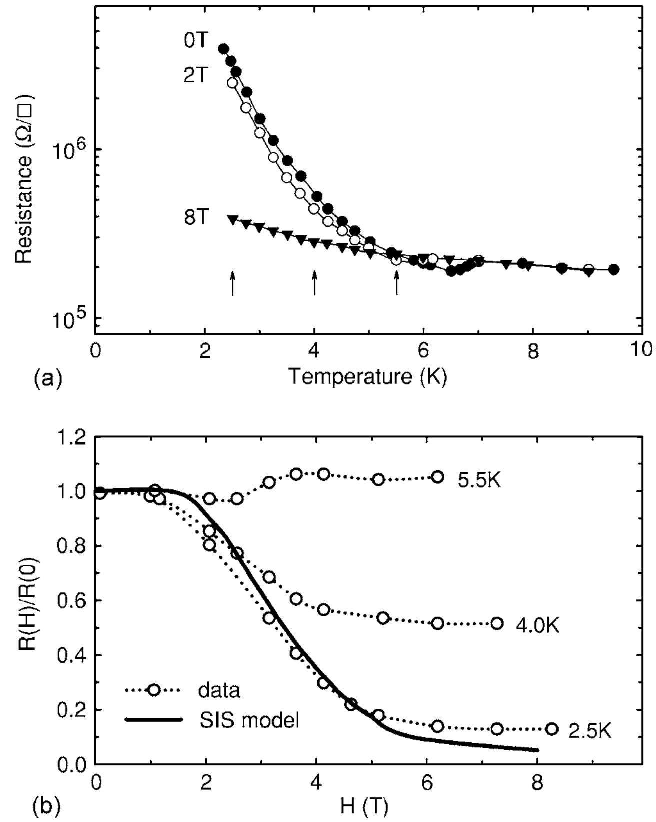

FIG. 2. Magnetoresistance on the insulating side of the SIT. (a) $R(T)$ in fields of 0, 2 T and 8 T. (b) Resistance normalized to its value in zero field as a function of magnetic field, at three different temperatures as indicated in Fig. 2(a). $R(H)/R(0)$ for the single junction model calculation at 2.5 K is shown as the solid curve.

Films on the insulating side of the SIT exhibit a giant negative magnetoresistance at low temperatures as illustrated in Fig. 2. In Fig. 2(b), an 8 T field drives the resistance lower by an order of magnitude at 2.5 K. The flattening of the magnetoresistance in all three curves at higher fields suggests that 8 T is sufficient to suppress the on-grain superconductivity and hence the spectral gap to zero. This critical field is consistent with the upper critical field, H_{c2} derived from transport[25] and tunneling,[24,30] experiments on granular Pb films on the superconducting side of the SIT.

A prediction from our single junction model is also possible for the case of magnetic field-induced pair-breaking. We simply used calculations of the modified density of states for samples in the presence of magnetic field[31] to replace the density of states in our tunneling model as described above. We have shown this calculation at a temperature of 2.5 K as a solid line in Fig. 2(b). The only additional information required is a value of the critical field, H_{c2}, at 2.5 K which is set to be 7 T. To estimate this value we used the experimentally accepted relationship

$$H_c(T) \approx H_c(0)(1-(T/T_c)^2)$$

with an $H_c(0)$ of 8 T[24,25,30] and T_c=7.2 K. Again, the resistance is derived from the zero-bias limit of the calculated I-V curves. Since we plot the experimental data and the model curve as normalized to the zero-field resistance, this calculation has *no* free parameters. Given that the magnetic field dependence of this simple model agrees well with experimental data in addition to the good agreement as temperature is varied, we are confident that it describes the dominant mechanism of transport in the insulating regime. To summarize, these films are unambiguously granular and dominated by intergranular tunneling, and the negative magnetoresistance is consistent with this understanding.

The I-V characteristics of quench condensed Pb films are very nonlinear.[19,21,32,33] Figure 3 shows typical results for a different film on the insulating side of the SIT (adapted from Ref. 21 and supporting results). In Fig. 3(a) are plotted the surface current density ($K \equiv I/W$) vs electric field ($E=V/L$) at five different temperatures where W and L are the film width (3.3 mm) and length (10.7 mm), respectively. We have chosen to plot K vs E, in order to eliminate the specific geometrical details of the films. $R_\square(T) \equiv E/K$ for a fixed K is shown in the inset with the value of K=0.6 mA/m denoted as the dashed line in the figure. It is important to clarify that our definition of resistance in this case *does not* derive from the linear, zero current limit definition. In fact $R_\square(T)$ (inset) is derived from data in the clearly nonlinear regime. Note that at the lowest two temperatures the K-E curves appear to approach a limiting form at larger E, consistent with the saturation of $R_\square(T)$ at low temperatures in the inset. This evolution with temperature is also qualitatively similar to I-V curves of single SIS junctions (see for example Ref. 28). In the lower frame, we have plotted the normalized electroresistance $R(E)/R(0)$ at fixed temperatures where $R_\square(E) = [\partial K(E)/\partial E]^{-1}$. Note the strong (more than two orders of magnitude) negative electroresistance observed at the lowest measured temperature and the similarity of these data with those in Fig. 2.

Figure 4 presents an alternative illustration of negative electroresistance. Here are plotted $R_\square(T)$ curves for three films (a, b, and c) in one experimental series with the same width (3.3 mm) and length (10.7 mm). In this case, $R_\square \equiv E/K$ and is plotted for three fixed surface current densities for each film (denoted as different symbols). The experimental configuration limited the maximum measurable resistance to 1 MΩ. Again we observe a significant negative electroresistance at the lowest temperatures as current (or equivalently voltage) is increased. Note that for the thickest of these three films, film "c," the $R_\square(T)$ curves are somewhat different from those for "a" and "b." This difference is likely an indication that this sample is crossing into a regime where the simple single-particle intergranular tunneling picture does not fully describe the behavior. Since sufficiently thick films show well-developed superconducting properties, there must be a region where tunneling dominated transport is overtaken by growing regions of phase coherence as the intergranular Josephson coupling begins to dominate. It is nonetheless interesting to note that for film 'c' an increase in current changes an apparently quasi-reentrant $R_\square(T)$ (resistance shows a resistance minimum) into an apparently "metallic" state (finite R at low T). It is important to reiterate that this metallic behavior is *not* seen in the zero-current linear regime ("intrinsic" resistance), but rather in a higher current nonlinear part of the $K(E)$ curve. Furthermore, in all but the lowest current $R_\square(T)$ curves for the highest resistance film (a), we observe a similar "apparently metallic" state. In those two lowest current curves for film a we note that they are effectively identical indicating that both the measurements are within the linear regime of the $K(E)$ curves. It is also valuable to note that there is no appreciable difference in the $R_\square(T)$ curves above the T_c of the grains in all the data indi-

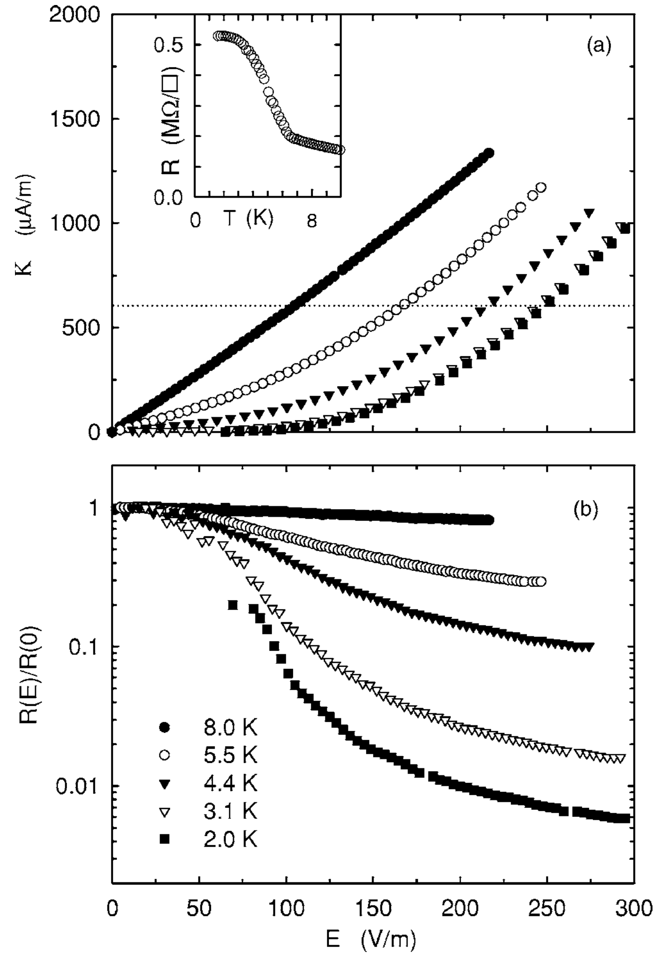

FIG. 3. Electroresistance on the insulating side of the SIT. (a) Surface current density K vs electric field E for a film at 8.0, 5.5, 4.4, 3.1, and 2.0 K. $R_\square \equiv E/K$ for a 600 μA/m applied current density is shown in the inset. Resistances for the two lowest temperature curves are nearly identical except in the low current limit, illustrating the metallic behavior induced by negative electroresistance in the regime where applied voltages exceed the energy scale of the superconducting energy gap. (b) Resistance normalized to its value at zero field as a function of electric field, calculated from the data in Fig. 3(a).

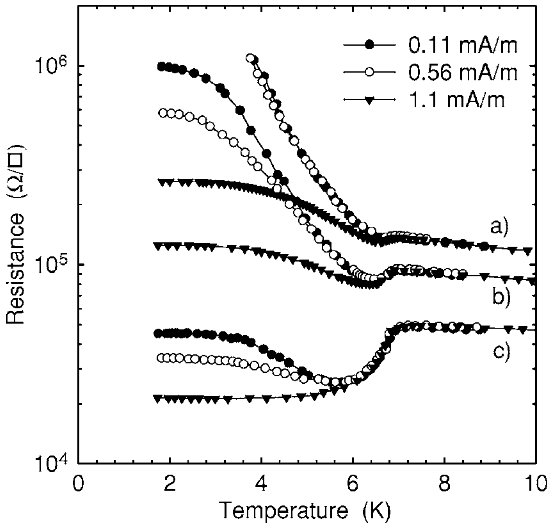

FIG. 4. Resistance per square $R_{\Box} \equiv E/K$ vs temperature for three films (a,b,c) in the insulating regime measured at three different applied surface current densities K as denoted in the legend. Metallic behavior $(dR/dT \approx 0)$ is observed at the lowest temperatures, with a diminishing temperature range as the applied current is decreased.

cating that the differences are due to nonlinearities in the superconducting tunneling characteristics. All of these results are consistent with tunneling-dominated transport in a granular superconducting film. The nonlinear $K(E)$ curves have been interpreted within the framework of a simple intergranular model.[21] The $R_{\Box}(T)$ effects of negative electroresistance and metallic behavior are simply manifestations of the nonlinearity in the regime where applied voltages exceed the energy scale of the superconducting energy gap.

Finally, it is interesting to note that the lowest current density $R_{\Box}(T)$ curves for films a and b in the temperature range between 4 and 6 K (no significant negative electroresistance) look like they are simply shifted vertically on the logarithm-linear plot in Fig. 4. That relationship is equivalent to an overall resistance scale factor, precisely the assumption used in our SIS single-junction model. In other words, if both samples are in the intergranular tunneling dominated regime, we expect them to have very similar temperature dependence with only a different resistance magnitude. For films in the insulating (and therefore intergrain tunneling dominated) regime, we expect that thinner films have some combination of a higher density of quasiparticle tunnel junctions and higher characteristic resistance per junction that will make the overall resistance scale larger.

It is useful to examine the extent to which the quasiparticle tunneling model can account for the behavior of other disordered, superconducting film systems that qualitatively resembles that of granular Pb films. Nonlinearities in I-V curves on the insulating side of the SIT appear in many systems. In some, quench condensed Sn, for example,[19] the behavior is very similar to that presented here. Quench condensed Ga films[34] and granular Al films,[33] however, exhibit strongly nonlinear I-V curves at voltages as low as the mV range. In the case of Ga a temperature independent threshold for conduction was observed that suggested that a charge unpinning process, similar to the type occurring in charge density wave compounds, was dominating.[34] In the case of granular Al films, the nonlinearities were strong enough that some films with negative dR/dT or "insulating" behavior at low currents exhibited positive dR/dT or "superconducting" behavior at high currents.[33] Again, these nonlinearities were present at the mV scale. They were attributed to a current induced suppression of the Coulomb barrier to intergrain tunneling and restoration of Josephson coupling. The agreement of the SIS model with the data presented above strongly suggests that neither of these processes control the nonlinearities in quench condensed Pb films. The differences could be rooted in morphological variations. In particular, the Ga films exhibited a thickness dependent mean field transition temperature[32] unlike the Pb films. Unlike Pb, Ga has several metastable phases each with a different T_c which can be formed under pressure or in thin films. This behavior generally indicates smaller grains or a more uniform morphology as is observed in "homogeneous" films where the T_c is also thickness dependent.[18,35,36] This effect is also observed in Bi samples[37] and is perhaps an indication of a morphology that is somewhat different from Pb where the T_c is nearly the bulk value even in the thinnest measured samples.[24]

Metallic conduction at low T has also been observed by others in zero magnetic field[32] and in magnetic field[8] on the superconducting side of the SIT. In one series of investigations, the existence of metallic behavior at low temperatures was correlated with the normal state resistance of the films.[32] Films with normal state resistances exceeding the quantum resistance for pairs showed metallic behavior while the others became superconducting. Figure 4 (lowest curve in group c) suggests that this flattening could arise from negative electroresistance in a granular system. Some homogeneously disordered films very close to the SIT also exhibit metallic conduction in magnetic fields.[8] Interestingly, it has been suggested that the magnetic field induces an effective granular structure in the form of puddles of superconductor in a metal background. Quantum phase slips between puddles are thought to produce the temperature independent resistance.[38] Despite the purported granularity, this flattening cannot be simply attributed to a negative electroresistance effect. Unlike granular systems, the homogeneous films contain a large density of quasiparticles at low energies,[39] and thus, not a well defined gap edge.

Our data give us confidence that a clear signature of the development of granular structure in an insulating film is negative magnetoresistance. Beloborodov has shown that even in magnetic fields sufficient to suppress the energy gap to zero, pair fluctuations can still give rise to a negative magnetoresistance.[40] This theory seems to account for the very high field negative magnetoresistance of three-dimensional AlGe films[41] and granular high T_c superconductors,[13] and perhaps effects in more homogeneous TiN films.[16] It might also account for similar, but more spectacular behavior recently observed in indium oxide films at high magnetic fields.[9,15,42,43]

To summarize our results, we have focused on the insulating side of the SIT in quench-condensed granular Pb films. We have identified three signature behaviors: negative magnetoresistance, negative electroresistance, and metallic behavior at low T in the explicitly nonlinear I-V regime. These three signatures can be explained within the context of the granular morphology of the samples and the dominance of intergranular tunneling.

We would like to acknowledge helpful discussions with W. Wu, P. Adams, I. Beloborodov, and B. Granger and support from NSF DMR Grant No. 0203608.

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