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Anomalous magnetoresistance of ultrathin films of $DyBa_2Cu_3O_{7-x}$ near the superconductor-insulator transition

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An anomalous peak in the magnetoresistance was observed in granular ultrathin films of $DyBa_2Cu_3O_{7-x}$ whose low-temperature behaviors span the zero-field superconductor-insulator transition. The magnitude of this peak increased with the weakening of the superconductivity of the films. Frustration effects in a Josephson coupled superconducting array are not sufficient to account for the peak. Instead, this unusual behavior may result from an interaction of the carriers with antiferromagnetically ordered Cu^{2+} ions on the copper-oxygen sheets, providing a crucial link between superconductivity and magnetism.

The interplay between superconductivity and localization has been studied extensively in uniform[1] and granular[2] ultrathin films of conventional superconductors in which disorder-driven superconductor-insulator transitions were found. Ultrathin films were studied because superconductivity is weaker and the effects of localization are more pronounced in two than in three dimensions. Superconductor-insulator transitions have also been studied in a variety of bulk, single-crystal, and thin-film high-temperature superconducting samples by varying doping,[3] and grain-boundary resistances.[4] A number of mechanisms for the transition in these materials have been identified, including localization, competition between Josephson coupling and intragranular capacitance, and spin disorder scattering. Here we report a study of magnetoresistance in a series of disordered ultrathin $DyBa_2Cu_3O_{7-x}$ (DBCO) films. We found that films whose sheet resistances fell closely on both sides of the zero-field superconductor-insulator transition exhibited an anomalous magnetoresistance which may be due to an increase in spin disorder scattering at a magnetic phase boundary.

Recently, we reported a zero-field superconductor-insulator transition in a sequence of ultrathin *c*-axis-oriented DBCO films[5] grown on $SrTiO_3$ by molecular beam epitaxy.[6] The transition was observed both by studying films of decreasing thicknesses, and by aging a single film by maintaining it in vacuum at room temperature for an extended period. Increased normal-state sheet resistances and reduced transition temperatures were correlated with decreased inverse Hall coefficients. The Hall coefficient was temperature independent, and therefore may give a meaningful indication of carrier concentration. The decreased carrier concentration suggests that this superconductor-insulator transition develops as oxygen is depleted and the electronic and magnetic configurations of the insulating donor compound are approached.

The temperature dependences of the sheet resistance in zero magnetic field measured in this study on a single 35-Å DBCO film are shown in Fig. 1. Each curve corresponds to a distinct aging step. Before aging, the DBCO film first deviated from its normal-state resistance at temperatures of 40 K and achieved global superconductivity at 2 K. In the aged films the superconducting transitions were always broad and only the onset of superconductivity was observed. Zero resistance may only be achieved below the limiting measuring temperature. Films exhibiting positive and negative values of dR/dT at the lowest temperatures will be referred to as superconducting and insulating, respectively, since these are expected to be the only two states for two-dimensional films in the $T \rightarrow 0$ limit,[7] except right at the superconductor-insulator transition.

A magnetic field of up to 14 T monotonically increased the resistance of the least resistive film, but at the lowest temperature was not sufficient to drive it normal. For each higher resistance stage the application of a magnetic field caused a *nonmonotonic* change in the resistance. The magnetoresistance was measured with the current flow in the CuO_2 planes, and the field perpendicular to the planes. The variation of sheet resistance with magnetic field at distinct, fixed temperatures is shown in Fig.

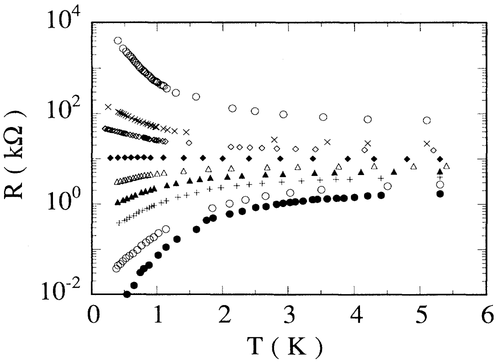

FIG. 1. Sheet resistance vs temperature, $R(T)$, for a series of films prepared by successive aging of a 35-Å-thick film at room temperature in vacuum. Different symbols correspond to different aging stages.

2 with each panel corresponding to a separate aging stage. For the superconducting stage [Fig. 2(a)], the resistance always decreases with decreasing temperature in a given field, and for the insulating stage [Fig. 2(c)], the resistance always increases with decreasing temperature. In the case of the resistive stage closest to the superconductor-insulator transition [Fig. 2(b)] the curves of $R(H)$ at temperatures below 1 K cross at three well-defined values of sheet resistance and magnetic field. We will interpret these crossings as successive magnetic-field-driven superconductor-insulator transitions.

The field at which the peak occurs varies systematically with temperature and age. The magnitude of the peak in $R(H)$, R_p, which was determined by subtracting the high-field magnetoresistance background, increases with decreasing temperature for all aging stages. At the lowest temperature measured, the peak magnitude at various aging stages is nearly linearly dependent on the zero-field resistance, as shown in Fig. 3. The temperature corresponding to the onset of nonmonotonicity increases with aging as well, from below 1.5 K in the least resistive film to above 5 K in the most insulating film. A more thorough study is currently being conducted to determine the field- and temperature-dependent phase diagram of the peak.

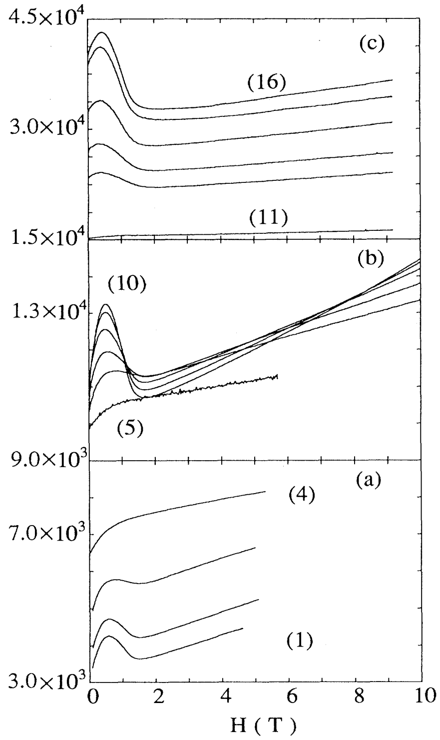

FIG. 2. Nonmonotonic dependence of sheet resistance on applied magnetic field at various temperatures. (a) The sample shows a tendency toward superconductivity. Curves (1)–(4) represent *increasing* temperatures of 0.38, 0.58, 1.1, and 4.2 K. (b) The sample in the transition region. Curves (5)–(10) represent *decreasing* temperatures of 4.2, 1.5, 1, 0.72, 0.47, and 0.38 K, respectively. (c) The sample is insulating. Curves (11)–(16) represent *decreasing* temperatures of 4.4, 1.2, 0.90, 0.64, 0.50, and 0.37 K, respectively.

We may consider whether the anomalous magnetoresistance can follow from modeling a film as a disordered two-dimensional Josephson coupled array, without considering the unique phenomenology of high-T_c superconducting grains. Periodic modulation of the magnetoresistance is observed in regular arrays due to frustration effects.[8] Resistance minima occur for applied fluxes that are integer multiples of ϕ_0/A, where $\phi_0=2.07\times10^{-7}$ G cm^2 is the flux quantum, and A is the area enclosed by a loop. The minimum in resistance in our films occurs near 1 T, which in this model corresponds to grain areas of about 450 Å, roughly consistent with the film microstructure.[9] Only the first-order oscillation is observed in our samples, as would be expected in positionally disordered arrays.[10]

Although a Josephson array model gives a qualitative explanation of a magnetoresistance peak, there are several features not explained by this picture. First, as the material evolves from superconductor to insulator, the magnetoresistance peak should become less pronounced. Instead the peak is much larger in the insulating than in the superconducting state. Second, this model does not explain the drop in resistance at high field to below that of zero field in the most insulating film. Finally, the field at which the peak occurs varies with temperature and aging stage, which would not be possible if the peak were due to film geometry.

To explain all of the features of the magnetoresistance peak, we must consider the properties of DBCO itself. In particular, $DyBa_2Cu_3O_{6+x}$ may be doped with oxygen from an insulator to a superconductor. Oxygen stoichiometry also controls the magnetic behavior of the Cu^{2+} ions in the conducting planes. In the insulating, oxygen-deficient phase of DBCO, the Cu^{2+} ions order antiferromagnetically within a CuO_2 layer, and couple antiferromagnetically between layers.[11] As the insulating compound is doped by addition of oxygen, the Néel temperature drops. However, antiferromagnetic spin fluctuations persist into the superconducting regime, and antiferromagnetism and superconductivity may coexist, at least in films close to the superconductor-insulator transition.

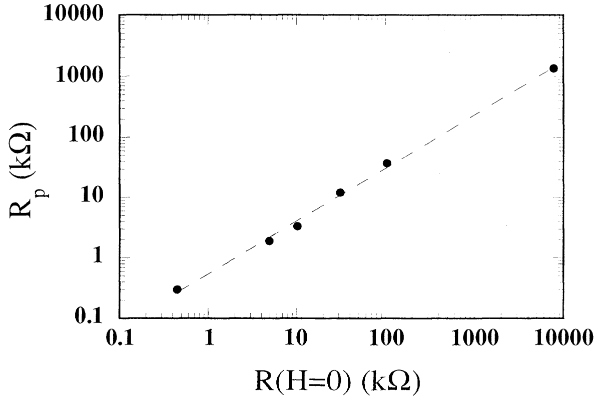

FIG. 3. Magnitude of peak in $R(H)$, R_p, plotted vs the zero-field resistance for each aging stage at 370 mK. The dotted line is a guide to the eye.

Peaks in magnetoresistance have been observed in antiferromagnetic materials such as ErAr,[12] a semimetal, and in Er_3Si_5,[13] a metal, at the magnetic-field-driven transition between antiferromagnetic and saturated paramagnetic phases. Yamada and Takada[14] have shown that there will be a peak in the magnetoresistance due to critical scattering[15,16] by spin fluctuations at this boundary. Since the low-temperature phase of the insulating parent compound of DBCO is antiferromagnetic, one might expect to observe a similar peak in its magnetoresistance at the critical field, although such a peak has never been reported. A peak may not be observed in fully insulating compounds because their antiferromagnetic exchange energy is very high. However, right at the superconductor-insulator transition, the Néel temperature is strongly reduced, and the field necessary to induce the antiferromagnetic-paramagnetic phase transition may be accessible. A peak in the *derivative* with respect to field of the magnetoresistance has been reported in a single crystal of La_2CuO_4, and was associated with a weak ferromagnetic transition.[17] However, in that study the current was directed *perpendicular* to the CuO_2 planes, where conduction occurs by interlayer hopping.

The evolution of the magnetotransport properties in our ultrathin DBCO films can be explained by a combination of intergranular and intragranular effects. As a film is aged, oxygen can be depleted preferentially from grain boundaries, reducing the intergranular coupling as well as the intragranular transition temperatures. The peak magnitude is a measure of the strength of the spin disorder scattering and the amount of material producing the scattering. The linear dependence of the peak resistance on the zero-field resistance over more than three decades (Fig. 3) is strong evidence that both scattering mechanisms have the same origin. The evolution of the temperature at which the nonmonotonic magnetoresistance begins is consistent with an increase in the magnetic correlation of the Cu^{2+} ions as the oxygen concentration is reduced. Whether the major source of the scattering is from magnetic material in the grain boundaries or randomly located magnetic sites within the grains will require further study which includes detailed investigations of I-V characteristics.

Having explained the peak in the magnetoresistance as primarily a scattering process, we may understand the behavior of the film nearest the zero-field superconductor-insulator transition [Fig. 2(b)]. At temperatures below 1 K, three distinct crossing points occur at fields of 0.10 ± 0.04 T, 1.08 ± 0.02 T, and 7.4 ± 0.2 T, and at sheet resistances of 11.7, 11.7, and 13.5 kΩ, respectively. At each crossing dR/dT changes sign, revealing transitions between superconducting and insulating behavior with increasing field. The first two field-driven transitions result from the modulation of the resistance, or the effective disorder, caused by the magnetic field and can be identified as zero-field transitions driven by disorder. The third, or high-field transition can be identified as a field-induced transition, which occurs at a critical vortex density[18] as described by the theory developed for nonmagnetic materials.[7] This transition occurs at approximately the same field and resistance as was observed in an oxygen-depleted Y-Ba-Cu-O single crystal.[19] A nonmonotonic magnetoresistance was not observed in the reduced T_c Y-Ba-Cu-O single crystal, and thus only the high-field transition was found.

We can rule out a direct role for the magnetic Dy^{3+} sublattice of DBCO, which orders antiferromagnetically near 1 K independent of oxygen concentration,[20] both because there is little interaction between charge carriers and the rare-earth ions, and such an interaction could not account for the evolution in peak magnitude, position, and onset temperature with aging of the film. Further work is being carried out to determine whether the Dy^{3+} ions play any role in these results.

In summary, we have observed a nonmonotonic magnetoresistance in a series of vacuum aging steps carried out on a DBCO film near its superconductor-to-insulator transition. We tentatively interpret the peak in $R(H)$ as evidence for critical scattering at a field-dependent transition from an antiferromagnetic to an aligned paramagnetic phase. This effect has been seen in studies of reduced-T_c single crystals possibly because the more granular and greater two-dimensional character of ultrathin films may play a role. There is also the possibility that although crystals may be more geometrically homogenous than films their oxygen doping is not, resulting in an inhomogeneity in the magnetic properties which hides the effects reported here. Finally, the observation of strong interactions between charge carriers and the underlying magnetic structure in the region where superconductivity and antiferromagnetic correlations appear to coexist is crucial evidence for a number of proposals[21] which suggest a magnetic basis for superconductivity in the cuprates.

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