Pair Breaking by Spin-Disorder Scattering at the Antiferromagnetic Transition of the Dy^{3+} Sublattice of $DyBa_2Cu_3O_{7-\delta}$ Films

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A peak in the temperature dependence of the resistance of ultrathin, granular $DyBa_2Cu_3O_{7-\delta}$ films has been found near the Néel temperature T_N of the Dy^{3+} sublattice which orders as a 2D Ising antiferromagnet. The shift of the position of the peak to lower temperatures with increasing magnetic field is consistent with it being a signature of T_N. These observations are attributed to the suppression of intergranular Josephson coupling by pair breaking, resulting from enhanced intragranular spin-disorder scattering of quasiparticles near the transition.

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The substitution of trivalent magnetic rare earth ions R^{3+} for Y ions in $YBa_2Cu_3O_{7-\delta}$ to form $RBa_2Cu_3O_{7-\delta}$ (R123) [1] results in materials which are reminiscent of those used earlier to investigate the interplay of superconductivity and magnetic ordering, i.e., the rare earth rhodium borides and Chevrel phase compounds [2]. The coupling between carriers and local moments in the R123 compounds, although weak on the scale of the superconducting condensation energy, may nevertheless be large enough to result in spin-disorder scattering which is pair breaking. The latter may be enhanced near the Néel temperature T_N of the antiferromagnetic transition because of fluctuations. However, values of T_N of the rare earth sublattices are much lower than superconducting transition temperatures, and the critical magnetic fields of the R123 compounds at low temperatures are very large. Thus one would not expect to observe any signature of antiferromagnetic ordering in measurements of either critical magnetic field or electrical resistance. As a consequence, information pertaining to the magnetic transitions of the R sublattices of superconductors has come from neutron scattering [3] and specific heat studies [4]. The Dy^{3+} sublattice has been found to be well described by the 2D Ising model.

In this Letter we report the observation of a peak in the temperature dependence of the resistance $R(T)$ of ultrathin, granular Dy123 films near T_N (<1 K) of the Dy^{3+} sublattice. The films were unusual in that they exhibited nonzero resistances down to temperatures well below T_N. This was a consequence of their properties being close to those of films at the zero-field superconductor-to-insulator transition which we reported on previously in Ref. [5]. We argue that the peak below 1 K results from the reduction of the *intergranular* Josephson coupling by pair breaking due to enhanced *intragranular* spin-disorder scattering near T_N. The peak moves to lower temperatures with increasing magnetic field (applied perpendicular to the film plane), consistent with expectations for an Ising antiferromagnet [6]. These observations provide evidence of the interaction between the magnetic degrees of freedom of the R sublattice and the superconductivity of the R123 compounds. They are also the first example of an observable effect of spin fluctuations associated with a magnetic transition in a Josephson-coupled system.

Ultrathin $DyBa_2Cu_3O_{7-\delta}$ films were prepared using the technique of ozone-assisted molecular-beam epitaxy. Their structural and chemical characterization has been described elsewhere [5,7]. The films, which were two-to-three unit cells thick, were grown heteroepitaxially on $SrTiO_3$ single-crystal substrates and consist of grains with characteristic linear dimensions estimated to be on the order of 100–200 Å. Films were typically 2.24 mm long and 0.75 mm wide and were measured using currents typically the order of 0.05 μA which for a 40 Å thick film corresponds to a current density of 1.65 A/cm^2. The I-V characteristics were linear up to currents in excess of the measuring current.

Figure 1 shows a plot of $R(T)$ from room temperature down to 100 mK for a film with a sheet resistance near the quantum resistance for pairs [5]. The onset of superconductivity begins at a temperature not far below 90 K. However, intergrain coupling is initially sufficiently weak that zero resistance is not established. Instead, the development of phase coherence within grains, accompanied by an opening of a gap in the quasiparticle density of states, leads to the first sign of superconductivity being an abrupt change in the sign of dR/dT and a *rise* rather than a *drop* in resistance. As the temperature is reduced further, Josephson coupling begins to overcome intergranular phase fluctuations, and near 40 K there is a downturn in resistance. This behavior has been found in the "weak-coupled" limit of models of granular films in which the films are treated as Josephson-coupled arrays [8]. These features imply that although the film is resistive, its constituent grains are superconducting at low temperatures and are coupled by tunneling.

Further support for this picture is the process of "aging" of films in vacuum at room temperature, an effect associated with oxygen depletion at grain boundaries, where oxygen is mobile [7]. With aging, the high

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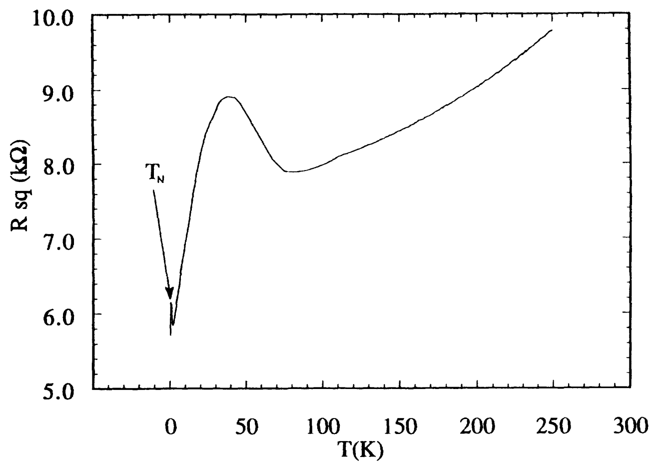

FIG. 1. Temperature dependence of the resistance of a $DyBa_2Cu_3O_{7-\delta}$ film with properties close to the insulating state on the superconducting side of the superconductor-to-insulator transition. The peak near 40 K is attributed to the gap opening up within the grains, whereas that below 1 K is attributed to the antiferromagnetic transition of the Dy^{3+} sublattice. The Néel temperature is given by the temperature at which the low temperature peak is found, after a linear background subtraction is made.

temperature resistance increases and the temperature of the onset of superconductivity is weakly suppressed, but the temperature at which the resistance turns down is strongly suppressed. This is consistent with a weakening of Josephson coupling with aging, and indicates that film resistances at low temperatures are associated with *intergranular* tunneling effects. When the films first become insulating ($R \to \infty$ as $T \to 0$), either through aging or application of a large (>6 T) magnetic field, the high temperature peak is still present, indicating that the grains are still superconducting, although the phase coupling is disrupted [5].

In analyzing magnetoresistance data in Ref. [5], we interpreted the low-field peak in $R(H)$ as a signature of a reentrant superconductor-insulator transition. Here, with the observation of a zero-field peak in $R(T,0)$, together with detailed curves of $R(T,H)$ exhibiting low-field magnetoresistance peaks, we are led to a reinterpretation of this phenomenon (observed in all six DBCO films). The fact that a zero-field peak is found near T_N of the Dy^{3+} ions strongly suggests that it is a consequence of spin-disorder scattering, associated with enhanced fluctuations found near T_N. Four possible mechanisms for this phenomenon involving tunneling are *intragranular* spin-disorder scattering, such as is found in antiferromagnetic metals [9]; *intergranular* Josephson tunneling with spin flip [10]; *intergranular* single-particle tunneling, involving localized magnetic moments in the regions between grains; and *intragranular* spin-flip scattering, resulting in pair breaking. The latter, which we argue is the relevant one, would suppress the magnitude of the order parameter on the grains, thus reducing the effective *intergranular* Josephson coupling [11,12]. Because the grains are superconducting, their *intragranular* resistance is zero, ruling out the first mechanism. The microscopic dynamics of the spins will not show up in the amplitude of the intergranular Josephson current, because the upper limit on the tunneling time is the lifetime of the virtual intermediate state in perturbation theory. This lifetime, proportional to the inverse of the superconducting energy gap, is much shorter than spin fluctuation times, which are proportional to T_N^{-1} and are the order of $(1\ \mathrm{K})^{-1}$ [13]. Fluctuating magnetic fields in the barrier associated with the spin dynamics which can affect the phase of the Josephson current will decrease the tunneling resistance rather than increase it [14]. These considerations rule out the interplay of the Josephson current and the spins in the barrier. Single-particle tunneling via localized magnetic states in the regions between grains can be ruled out, because it would be expected to lead to zero-bias conductance peaks which were not observed in any of the films [15].

The nature of pair breaking in superconductors near an antiferromagnetic transition of a magnetic sublattice has been treated by Ramakrishnan and Varma [11], and parts of their analysis were applied to the problem of Josephson tunneling in the antiferromagnetic superconductor $SmRh_4B_4$ by Vaglio, Terris, Zasadzinski, and Gray [12]. In that work, pair breaking *decreased* as the antiferromagnetic state was entered, and enhanced fluctuations above the Néel temperature were not observed. However, Ramakrishnan and Varma showed that in the quasielastic scattering limit, the pair-breaking parameter ρ of an antiferomagnetic superconductor is given by

$$\rho = \frac{3J^2}{\pi} \sum_{\mathbf{q}} \phi(\mathbf{q})\chi(\mathbf{q}), \tag{1}$$

where J is the exchange interaction between the carriers and the localized Dy^{3+} magnetic moments. The quantity $\chi(\mathbf{q})$ is the wave-vector-dependent spin susceptibility of those moments, and $\phi(\mathbf{q})$, the joint density of states, is given by

$$\phi(\mathbf{q}) = \frac{1}{N(\varepsilon_F)V^2} \sum_{\mathbf{k}} \delta(\varepsilon_{\mathbf{k}} - \mu)\delta(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}-\mathbf{q}}). \tag{2}$$

These equations determine the magnitude of the scattering by various wave-vector components of $\chi(\mathbf{q})$, which is peaked at the antiferromagnetic wave vector Q. The result for ρ resembles that obtained in the study of the temperature dependence of the resistivity near T_N in conducting antiferromagnets in which $\phi(\mathbf{q})$ is proportional to the momentum transferred to the scattered carriers by spin fluctuations [16]. In that problem there is no critical behavior in the largest contribution to the resistivity. The result for the pair-breaking parameter ρ is similar with the leading correction in the asymptotic limit of the transition $(\Delta\rho)_{\mathrm{crit}} \sim |(T - T_N)/T_N|^{1-\alpha}$, where α is the specific heat critical index which vanishes for a 2D Ising system [11]. In the present problem, that of a high-T_c superconductor

$Q > 2k_F$ [17], one expects a broad peak in the pair-breaking parameter slightly above T_N [16]. The nature of the complicated band structure, together with the spin susceptibility, will determine the actual behavior. If $\phi(\mathbf{q})$ is substantial in the region of the q space in which $\chi(\mathbf{q})$ peaks, then pair breaking should increase [13]. For a 2D Ising system, it might be possible to compute $\Delta\rho(T)$ in detail over an extended temperature range [18].

We consider how intragranular pair breaking affects the behavior of a single junction as a step towards explaining granular film behavior. If the I-V characteristic of a junction is broadened by phase fluctuations, then the voltage drop at fixed current, and thus the junction resistance, will be inversely proportional to the maximum Josephson current I_1. At $T = 0, I_1 = \pi\Delta/2eR_N$, where R_N is the normal tunneling resistance. The energy gap in the small pair-breaking limit is then $\Delta = \Delta_0 - [\pi^2/2]k_BT_c\rho$, where Δ_0 is the gap in the absence of pair breaking, and ρ is the full pair-breaking parameter [19]. The latter can be written as $\rho = \rho_0 + \Delta\rho(T)$, where ρ_0 is the pair-breaking parameter in the paramagnetic phase, and $\Delta\rho(T)$ is the temperature-dependent part of the pair-breaking parameter due to spin-disorder scattering near T_N. The resistance of the junction, for small pair breaking, is then proportional to the inverse of the Josephson coupling:

$$\frac{1}{I_1} = \frac{2eR_N}{\pi\bar{\Delta}}\left[1 + \frac{\pi^2 k_B T_c}{2\bar{\Delta}}\Delta\rho(T)\right], \qquad (3)$$

where $\bar{\Delta} = \Delta - (\pi^2/2)k_BT_c\rho_0$ is the average value of the gap in the paramagnetic phase [20].

The behavior of single junctions is relevant to that of granular films. If films were modeled as a square arrays of identical junctions, then the resistance of one junction would be that of the array. A more realistic model would be a 2D random network of junctions. The sheet resistance of such a network can be seen to be (approximately) the median resistance of the random array [21]. Thus these arguments predict a peak in the resistance of the granular film, the shape given by the temperature dependence of the pair-breaking parameter of a single junction.

Figure 2 shows a series of plots of the sheet resistance R_{sq} vs T in different fields. We have not carried out an analysis of background effects here, as the flux flow resistance and the superconductor-insulator transition which occur at high fields [Fig. 2(a)] are hard to quantify [5]. The peak at low fields and temperatures associated with the magnetic transition, as shown in Fig. 2(c), is clearly distinct from the high-field behavior of Figs. 2(a) and 2(b). The low temperature peak of Fig. 1, seen in detail as the zero-field trace of Fig. 2(c), is significant over roughly the same fractional temperature range as that over which there is a significant specific heat signature in the 2D Ising model [22]. In addition to the enhancement of pair breaking above T_N, the sharp drop in resistance below T_N suggests a reduction in pair breaking in the antiferromagnetic phase relative to the paramagnetic. The effective phase diagram of the antiferromagnetic transition, shown in Fig. 3, was mapped out taking the transition to be the locus of points in H and T of the maxima of the peaks of $R(T,H)$ at fixed H of Fig. 2(c). The effect of increasing magnetic field is

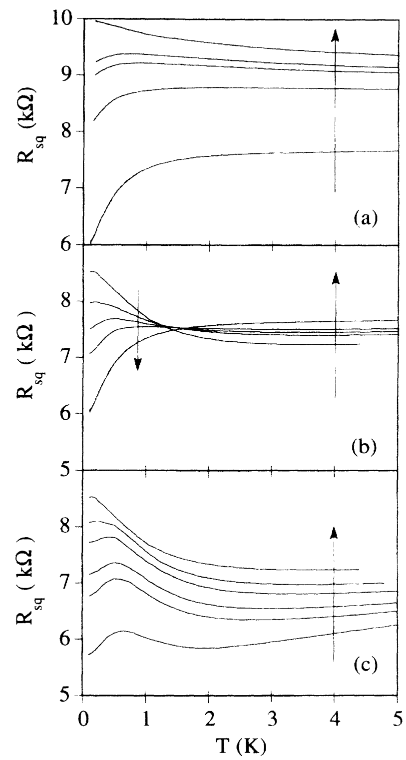

FIG. 2. Evolution of the temperature dependence of the resistance with magnetic field where the arrows point in the direction of increasing field. Some field values are repeated in the plots to maintain continuity in the data. (a) Fields of 1.3, 5.0, 6.0, 6.3, and 7 T showing a superconductor-insulator transition between 6.3 and 7 T, at fields much higher than those of interest here. (b) Fields of 0.55, 0.8, 0.9, 1.0, and 1.3 T showing decreasing resistance with field at temperatures below about 1.5 K. (c) Fields of 0, 0.05, 0.1, 0.2, 0.3, and 0.55 T displaying a peak in $R(T,H)$, which is suppressed with increasing magnetic field.

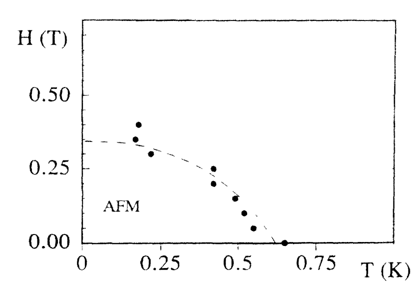

FIG. 3. The location in field and temperature of peaks in $R(T,H)$ of 2(c) are shown as points. The dashed line shows the extrapolation of the transition to $T = 0$.

to shift T_N downward as expected, since a uniform magnetic field is not thermodynamically conjugate to the order parameter of an antiferromagnet. The peak is driven towards $T = 0$ at $H \sim 0.33$ T. Above this field the low temperature resistance drops [Fig. 2(b)], indicating that the shift in T_N is not due to flux flow resistivity. In the $T \to 0$ limit, the critical field H_c of the 2D Ising model is given by $H_c = zJ/2\mu$ [23]. Taking the coordination number of the Dy^{3+} ions, $z = 4$, the exchange constant, $J = 0.44 k_B T_N$, where $T_N = 0.6$ K, and the magnetic moment of the Dy^{3+} spins, $\mu = 7.0\mu_B$ [15], one finds $H_c = 0.33$ T, which is close to the value extrapolated from the graph. We also expect the spin system to undergo a spin-flop transition which is not evident in our data. An independent phase diagram similar to ours has been mapped out for the Dy^{3+} ions in Dy123 by measuring specific heat [24].

In summary, we have observed a well-defined peak in $R(T)$ in zero magnetic field in granular Dy123 films at a temperature very close to the Néel temperature of the Dy^{3+} sublattice. We argue that this peak is a consequence of the suppression of intergranular Josephson coupling by intragranular pair breaking, resulting from the spin-disorder scattering of quasiparticle excitations near T_N. The width of the peak is qualitatively consistent with the 2D Ising character of the antiferromagnetic transition. The critical field at which the peak is suppressed to zero temperature is close to that expected for a 2D Ising antiferromagnet. These results are the first example of identifiable conventional pair-breaking effects in a high-T_c material caused by the R^{3+} lattice. They are also the first example of enhanced spin fluctuations near an antiferromagnetic transition in a Josephson-coupled system.

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[1] J.M. Tarascon, W.R. McKinnon, L.H. Green, G.W. Hull, and E.M. Vogel, Phys. Rev. B **36**, 226 (1987).
[2] For a review see, *Superconductivity in Ternary Compounds II: Superconductivity and Magnetism,* edited by M.B. Maple and Ø. Fischer (Springer-Verlag, Berlin, 1982).
[3] J.W. Lynn, in *High Temperature Superconductivity,* edited by J.W. Lynn (Springer-Verlag, New York, 1990); and also T.W. Clinton, doctoral dissertation, University of Maryland, 1992 (unpublished).
[4] M.W. Kirken and L.J. de Jongh, Solid State Commun. **64**, 1201 (1987); B.W. Lee, J.M. Ferreira, S. Ghamaty, K.N. Yang, and M.B. Maple, in *Oxygen Disorder Effects in High-T_c Superconductors,* edited by J.L. Moran-Lopez and Ivan K. Schuller (Plenum Press, New York, 1990).
[5] T. Wang, K.M. Beauchamp, A.M. Mack, N.E. Israeloff, G.C. Spalding, and A.M. Goldman, Phys. Rev. B **47**, 11619 (1993).
[6] P. Allenspach, A. Furrere, and F. Hulliger, Phys. Rev. B **39**, 2226 (1989), and references cited therein.
[7] V.S. Achutharaman, K.M. Beauchamp, N. Chandrasekhar, G.C. Spalding, B.R. Johnson, and A.M. Goldman, Thin Solid Films **216**, 14 (1992).
[8] B.G. Orr, doctoral dissertation, University of Minnesota, 1985 (unpublished).
[9] Y. Suezaki and H. Mori, Prog. Theor. Phys. **41**, 1177 (1969).
[10] Hiroyuki Shiba and Toshio Soda, Prog. Theor. Phys. **41**, 25 (1969).
[11] T.V. Ramakrishnan and C.M. Varma, Phys. Rev. B **24**, 137 (1981).
[12] Ruggero Vaglio, B.D. Terris, J.F. Zasadzinski, and K.E. Gray, Phys. Rev. Lett. **53**, 1489 (1984).
[13] This argument is due to L. Glazman. In the event of d-wave pairing, certain directions would have zero gap, and the Josephson amplitude might then be proportional to the spin susceptibility. The qualitative features of the discussion which follows would still hold.
[14] A.M. Goldman, C.G. Kuper, and O.T. Valls, Phys. Rev. Lett. **52**, 1340 (1984).
[15] J.A. Applebaum, Phys. Rev. Lett. **17**, 91 (1966); P.W. Anderson, Phys. Rev. Lett. **17**, 95 (1966).
[16] S. Alexander, J.S. Helman, and I. Balberg, Phys. Rev. B **13**, 304 (1976).
[17] T.W. Clinton, doctoral dissertation, University of Maryland, 1992 (unpublished).
[18] K.M. Beauchamp, doctoral dissertation, University of Minnesota, 1993 (unpublished).
[19] Kazumi Maki, in *Superconductivity,* edited by R.D. Parks (Marcel Dekker, Inc., New York, 1969).
[20] If the expression given for the Josephson current in the presence of weak pair breaking is used, $\pi^2/2$ in Eq. (4) must be replaced by 16/3. See, I.O. Kulik, JETP **23**, 529 (1966).
[21] V. Ambegaokar, B.I. Halperin, and J.S. Langer, Phys. Rev. B **4**, 2612 (1971).
[22] M.E. Fisher and R.J. Burford, Phys. Rev. **156**, 583 (1967).
[23] R. Navarro, in *Magnetic Properties of Layered Transition Metal Compounds,* edited by L.J. de Jongh (Kluwer Academic Publishers, Dordrecht, 1990), and references cited therein.
[24] A.P. Ramirez, L.S. Schneemeyer, and J.V. Waszczak, Phys. Rev. B **36**, 7145 (1987).