Pair Tunneling in $ErRh_4B_4$ Films

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Pair-tunneling measurements have been carried out on $ErRh_4B_4$-Lu_xO_y-In thin-film junctions. The Fraunhofer-like pattern of the magnetic-field dependence of the dc Josephson current has been found to split at a temperature just above the reentrant transition of $ErRh_4B_4$. This suggests the onset of a magnetic structure of semimacroscopic scale which does not break up the superconductivity of the $ErRh_4B_4$ electrode into disconnected domains, at least within the surface region probed (~ 150 Å).

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The synthesis of superconducting compounds containing magnetic ions in an ordered array[1] reopened the issue of the possible coexistence of long-range superconducting and magnetic order.[2] Compounds such as $ErRh_4B_4$ [1] and $HoMo_6S_8$,[3] which become superconducting at a high temperature T_{c1}, and then reenter the normal state at a low temperature T_{c2} and become ferromagnetic, are of interest because of the possibility that superconductivity and ferromagnetism overlap in the vicinity of the lower transition. Indeed, neutron-diffraction experiments on bulk polycrystalline $ErRh_4B_4$ samples imply that the ferromagnetic regions of unknown size are present along with superconductivity at temperatures near T_{c2}.[4]

The superconducting state in the vicinity of T_{c2} has also been of interest because of the prediction of a new type of magnetic structure coexisting with superconductivity. Depending upon the choice of material-dependent parameters, this structure may be characterized as a spin-spiral,[5] a spontaneous vortex,[6] or, with magnetic anistropy included in the model,[7] as a linearly polarized sinusoidal configuration. Small-angle neutron-diffraction experiments on powdered samples of both $ErRh_4B_4$ [8] and $HoMo_6S_8$ [9] have confirmed the existence of a spatially modulated magnetic structure with a wavelength of the order of 100 Å. Recent neutron-diffraction measurements[10] on single crystals of $ErRh_4B_4$ have shown that the structure is linearly polarized in addition to providing strong evidence of the coexistence of superconducting and normal ferromagnetic domains near T_{c2}.

Recently it has been suggested that ferromagnetism and superconductivity coexist at temperatures above T_{c2} near the surface of bulk specimens of $ErRh_4B_4$ and $HoMo_6S_8$, and in films of these materials thinner than the penetration depth.[11] This is based on the idea that superelectrons near a surface do not diamagnetically screen rare-earth spins as well as do superelectrons in bulk, in a manner sufficient to strengthen the tendency towards ferromagnetic alignment near a surface. Electron tunneling, because it probes a volume near the surface determined by the coherence length (~ 150 Å in $ErRh_4B_4$ films), would be expected to be a useful probe of such an effect.

Previous oxide-barrier,[12,13] point-contact,[14] and vacuum-barrier[15] tunneling investigations of $ErRh_4B_4$ have been only preliminary in nature. In this Letter we report pair-tunneling studies of $ErRh_4B_4$-Lu_xO_y-In thin-film junctions which demonstrate for the first time the effect on superconducting long-range order of the onset of ferromagnetic order near T_{c2}. Single-particle tunneling data will be reported elsewhere. The $ErRh_4B_4$ film on which four essentially identical, but independent, junctions were formed was 4500 Å thick and was nearly single phase. Its superconducting properties were essentially identical to those of six other films. X-ray analysis indicated that the impurities in the films were RhB and $ErRhB_4$ and that the crystallites were not preferentially ordered on the substrate. The junctions were prepared using a dc sputtering technique[16] in which a 25-Å-thick layer of nonmagnetic Lu metal was sputtered directly onto a sputtered $ErRh_4B_4$ base layer without breaking vacuum. This thin rare-earth layer was then oxidized in air at room temperature for 15 min to form a Lu-oxide barrier. The junctions were completed with an evaporated In counterelectrode.

The temperature dependence of the zero-voltage current $I_c(T)$, which we have interpreted as a Josephson current, along with $R(T)$ of the $ErRh_4B_4$ electrode, are shown in Fig. 1. T_{c1} and T_{c2} as determined resistively were 8.2 and 0.94 K, respectively. Below T_{c2} the normal resistance was 95% of its value above T_{c1}. The coherence length $\xi(0)$, which determines the volume of the sample probed by tunneling, was estimated to be 150 Å from measurements of dH_{c2}/dT in the limit of $T \to T_{c1}$. In all four of the junctions no evidence of a dc Josephson current was observed below T_{c2}, in contrast with the results of point-contact studies on $(Er_{0.58}Ho_{0.42})Rh_4B_4$.[16] On the other hand, the dc Josephson current became small at temperatures above the resistively determined T_{c2}.

With the Josephson current quenched by a small magnetic field, the zero-bias resistances were only between 2 and 5 times the normal tunneling resistance. This "leakiness" is not too surprising given the magnetic Er^{3+} ions in the compound which would be expected to result in pair-breaking effects. In addition, the presence of even small amounts of impurity phases which are normal would also increase the conductance.

The observed zero-voltage currents can be interpreted as Josephson currents rather than shorts because their maximum values exhibit the well-known periodic variation with applied magnetic field.[17] However, the junction couplings were only uniform enough to permit the central maximum and one and sometimes two side peaks of the Fraunhofer pattern to be observed. The spacing between the nodes in the pattern, ΔH, was temperature dependent, increasing from zero at $T = 3.4$ K, which is T_c of the In counterelectrode, beginning to fall at 1.4 K, and approaching zero as $T \to 1.1$ K. The latter effect is consistent with both the susceptibility $\chi(T)$ and the penetration depth $\lambda(T)$ of $ErRh_4B_4$ becoming large as $T \to T_{c2}$, with $\lambda(T)$ growing as a consequence of strong pair breaking. The data on the temperature dependence of the maximum Josephson current (Fig. 1) and the period of the Fraunhofer patterns (not shown) could in principle be used to test a detailed theory of the microscopic interaction of magnetic and superconducting order.

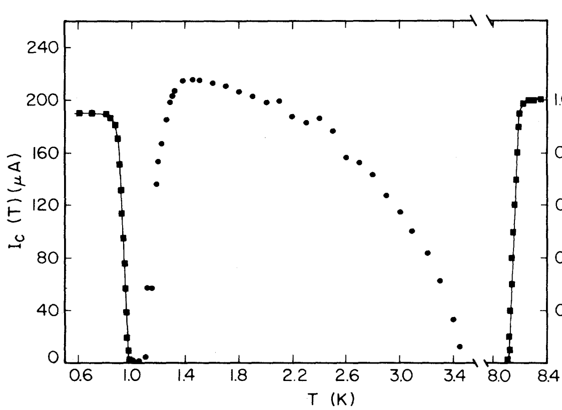

FIG. 1. The maximum dc Josephson current $I_c(T)$ of an $ErRh_4B_4$-Lu_xO_y-In junction (dots) and $R(T)/R(9\text{ K})$ (squares) of the $ErRh_4B_4$ electrode.

Before ΔH vanished, the central peak split by a few tenths of an oersted. This effect, shown in Fig. 2, occurred at $T \sim 1.16$ K on cooling from $T > T_{c1}$. On the other hand, if the junction were warmed from $T < T_{c2}$, irreproducible patterns with many irregular peaks were observed. Occasionally on cooling from $T > T_{c1}$ the central peak did not split, but shifted its position by a few tenths of an oersted, an effect which may be associated with flux trapping in the junction.

The Fraunhofer pattern of a Josephson junction exists because of long-range spatial coherence of the pair phase difference between the electrodes of the junction. The pattern is essentially the spatial Fourier transform of the supercurrent coupling. The onset of uniform magnetic order in the $ErRh_4B_4$ electrode should produce internal magnetic fields within the junction which in turn would result in only a shift of the Fraunhofer pattern and not a splitting. On the other hand, if the tunneling supercurrent were spatially inhomogeneous the Fraunhofer pattern would split as long as the long-range superconducting order were not disrupted as would be the case if the $ErRh_4B_4$ electrode were to consist of disconnected superconducting domains. A spatially inhomogeneous current density could result from normal ferromagnetic domains of a characteristic size such as those reported in Ref. 10, or from a periodic magnetic structure. These magnetic phenomena would affect the tunneling current either by introducing a modulation of the phase difference across the junction in addition to that due to the uniform applied magnetic field or by modulating the magnitude of the order parameter at the barrier. Both mechanisms could operate simultaneously.

There is currently no detailed theory of the field dependence of the Josephson current of an asymmetric junction where one electrode is a magnetic superconductor undergoing a transition

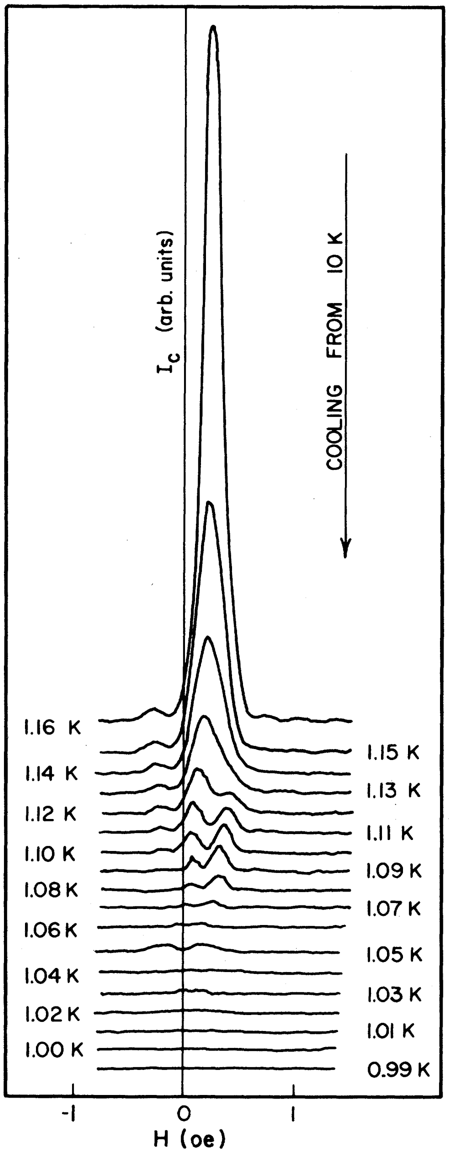

FIG. 2. Magnetic-field dependence of $I_c(T)$ at different temperatures. The offset is in the zero of the superconducting magnet.

to a magnetically ordered state. If one assumed in an *ad hoc* fashion the existence of modulation of the phase difference and coupling strength with a wavelength λ_M, then, as we will show in detail elsewhere, I_c vs H becomes a sum of Fraunhofer patterns described by integers n, centered at fields given by $C_1(H) \pm 2\pi n/\lambda_M = 0$ with amplitudes given by integer-order Bessel functions $J_n(C)$, where C is the amplitude of the sinusoidal variation of the phase difference and where

$$C_1(H) = H(2e/\hbar)[\lambda' \tanh(d'/2\lambda') + (1+4\pi\chi)\lambda_{\mathrm{eff}} \tanh(d/2\lambda_{\mathrm{eff}}) + t] \qquad (1)$$

is the wave vector of the variation of the phase difference due to the applied field. Equation (1) is a generalization of the standard form[17] to the case of a magnetic superconducting electrode of thickness d, susceptibility χ, penetration depth λ, and effective penetration depth[18] $\lambda_{\mathrm{eff}} = \lambda/(1+4\pi\chi)^{1/2}$, coupled across a barrier of thickness t to a nonmagnetic electrode of thickness d' and penetration λ'. The peaks in the split pattern are located at fields which correspond to λ_M being equal to a multiple of the wavelength of the modulation of Josephson current by the applied field. The number of peaks will be limited as the $J_n(C)$ rapidly become small with increasing C. Inhomogeneity of the junction coupling will wash out higher orders as in the usual Fraunhofer pattern.

At 1.5 K, where the pattern is not split, the period of the Fraunhofer pattern is $H_1 = 0.6$ Oe. Then, by taking d and $d' = 4500$ Å, $\lambda'(0) = 200$ Å, the junction width $W = 2.5\times 10^{-2}$ cm, and assuming $\chi = (0.184\text{ K})/(T - 0.94\text{ K})$,[18] λ is found to be 750 Å using the condition $C_1(H_1)H_1W/2 = 1$ to define the period.[17] Then at 1.2 K, where the main peaks, assumed to be given by $n = 1$, are offset from zero by about 0.2 Oe, the relation $C_1(H) \pm 2\pi/\lambda_M = 0$ gives $\lambda_M = 3.4\times 10^{-3}$ cm, a result much greater than both the 100-Å wavelength of the periodic magnetic phase and the 5000-Å characteristic grain size of the $ErRh_4B_4$ films. In arriving at the above we assumed that λ, as a result of pair breaking, was increased tenfold at 1.12 K relative to its value at 1.5 K. This factor is the reciprocal of that by which the dc Josephson current is reduced from its value at 1.5 K, presumably mostly by pair breaking. The reciprocal relation follows from the fact that pair breaking in reducing the order parameter ψ near T_{c2} would decrease the Josephson current $\sim |\psi|$ and increase $\lambda \sim |\psi|^{-1}$. Without this assumption, λ_M would be even larger. If there are very short-wavelength oscillations of the coupling and phase difference resulting from magnetic structure such as in the linearly polarized-sinusoidal configuration, then more homogeneously coupled junctions than those reported here would be needed to see them in the Josephson current.

There is an additional feature of the data which supports the model. The amplitudes of the various contributions to the Josephson current depend on the $J_n(C)$ which oscillate and decrease in magnitude as C increases. A nonmonotonic decrease in the amplitude of the pattern with decreasing temperature as is evident from Fig. 2 might be expected from such a Bessel-function dependence.

In summary, we have succeeded in preparing tunneling junctions with $ErRh_4B_4$ films. The dc Josephson current becomes small above T_{c2} and the Fraunhofer pattern splits. A possible explanation of the latter effect is the development of magnetic structure of semimacroscopic size which does not result in the $ErRh_4B_4$ film being broken into disconnected superconducting domains. However, because the tunneling amplitudes depend on the strength of the superconductivity within a volume at the surface defined by the 150-Å coherence length of $ErRh_4B_4$, the present experiments, in the absence of a detailed theory, do not reveal whether the presumably normal magnetic structure extends across the 4500-Å thickness of the film or is the surface effect suggested in Ref. 11.

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