Spin-Polarized Electron Tunneling Study of an Artificially Layered Superconductor with Internal Magnetic Field: EuO-Al

P. M. Tedrow and J. E. Tkaczyk
Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

and

A. Kumar
Physics Department, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 3 February 1986)

The tunneling conductance of junctions formed on thin films of Al in contact with films of the ferromagnetic semiconductor EuO implies that such composites behave like BCS superconductors with internal magnetization. In a magnetic field B applied in the plane of the films, the superconducting quasiparticle density of states shows a splitting $2\mu(B^* + B)$, where B^* can be greater than B by more than a tesla. The critcal field of composite films is reduced by approximately B^* compared to that of identical Al films without EuO. The observed B^* is of the same magnitude as $\mu_0 M$, the magnetization of EuO.

PACS numbers: 74.50.+r, 75.50.Dd, 75.70.−i

Because magnetic order and superconductivity are, in general, incompatible, the interaction between superconductivity and magnetism has been a topic of interest for many years. Recently, investigations of exotic new compounds containing rare-earth or actinide elements have uncovered remarkable superconducting and magnetic properties. Such behavior includes the reentrant superconductivity of the rare-earth rhodium borides[1] and tenary molybdenum chalcogenides,[2] the magnetic-field–induced superconductivity of $Eu_{0.8}Sn_{0.2}Mo_6S_8$,[3] and the superconductivity of $CePb_3$ which appears only in a high magnetic field.[4] In addition, improving thin-film deposition techniques are allowing new materials properties to be sought in artificially layered systems.[5] For example, a recent experiment involving tunneling between two Pb films with a $Ho(OH)_3$ barrier has been described as showing the formation of a bound state in the Pb due to the ferromagnetism of the barrier.[6] In the present experiment, we have examined the density of states, in a parallel applied magnetic field, of a thin superconducting Al film backed by a film of the ferromagnetic semiconductor EuO and have found that the composite behaves like a BCS superconductor with an internal magnetic field similar in magnitude to that arising from the magnetization of EuO. Classically, a quasiparticle in the Al would feel only the applied magnetic field. Therefore, we assume that the observed enhanced magnetic field arises because of tunneling of the quasiparticles into the EuO.

The samples were made by vacuum evaporation onto liquid-nitrogen–cooled glass substrates. First, 5 nm of Eu was evaporated. The substrate was warmed to room temperature and the Eu was exposed to an oxygen glow discharge. After the substrate was cooled again, an Al film 4–10 nm thick was evaporated. Oxidation of this film provided the tunnel barrier. A counter electrode of either Al or Fe was then added. Identical control junctions without EuO were made at the same time on the same substrate. The junctions were cooled to 0.4 K with an immersion ^{3}He cryostat. The magnetic field was supplied by either a superconducting solenoid or a water-cooled Bitter magnet. The field was applied parallel to the plane of the films.

EuO is a ferromagnetic semiconductor[7,8] with a Curie temperature of 70 K and with the NaCl structure. Its resistivity depends on stoichiometry and impurity content and can be as high as 10^8 Ω-cm at low temperature for oxygen-rich or stoichiometric examples. On the basis of published methods[7] of forming EuO, we would not expect any excess Eu in our films, but there may be some nonferromagnetic Eu_2O_3. We have not as yet attempted to analyze these films.

Previous experiments[9] have established the fact that the superconducting density of states of a thin Al film is split into spin-up and spin-down parts by an applied magnetic field B. The splitting energy at low temperature and field is $2\mu B$ where μ is the electron magnetic moment. The resulting density of states is shown schematically in Fig. 1(a). If the Al film is part of a tunnel junction with a normal-metal counter electrode, a conductance (dI/dV) curve such as shown in Fig. 1(b) would be observed[9] as a function of voltage. The density-of-states splitting of $2\mu B$ is reflected in the dI/dV curve. If the counterelectrode is Fe, a curve such as Fig. 1(c) would be observed.[10] The asymmetry arises from the polarization of the electrons at the Fermi surface of the Fe. This asymmetry makes possible the determination of the spin-dependent densities of states of the superconductor.[11] Note that if

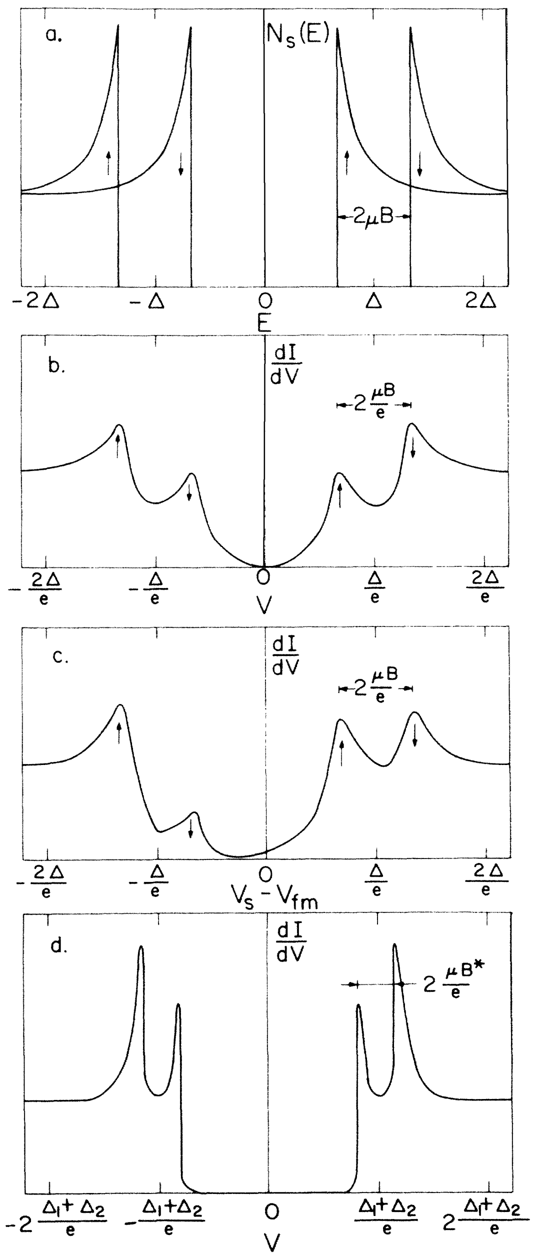

FIG. 1. (a) BCS density of states split into spin-up and spin-down parts by a magnetic field B. (b) Schematic tunneling conductance vs voltage of a junction between a thin Al film and a normal metal in a magnetic field B. (c) Schematic tunneling conductance vs voltage of a junction between a thin Al film and a ferromagnetic metal film in a magnetic field. (d) Schematic tunneling conductance vs voltage of a junction between a thin Al film in a magnetic field B and a thin Al film in a magnetic field $B+B^*$.

the counter electrode is another thin Al film, no splitting will be observed in the dI/dV curve with magnetic field applied to the junction,[12] because both films will have their densities of states split by the same amount and spin is conserved in the tunneling process. If one film should have different splitting from the other, however, structure will be observed in the conductance. This previously unobserved situation can arise if one superconductor has very large spin-orbit scattering and a high magnetic field is applied, or if the two superconductors are in different magnetic fields or if they have different electronic g factors. Thus, in S-I-N tunneling, the total splitting of the density of states is observed, while in S_1-I-S_2 tunneling, the difference in splitting of the densities of states of the two superconductors S_1 and S_2 is observed.

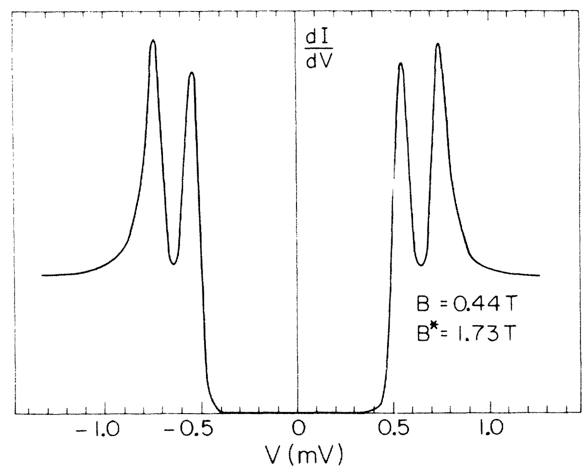

FIG. 2. Measured tunneling conductance vs voltage for an EuO-Al/Al_2O_3/Al junction with an applied field of 0.44 T and a voltage splitting equivalent to 1.73 T.

The tunneling conductance for an EuO-Al/Al_2O_3/Al junction in an applied field is shown in Fig. 2. Both Al films were 4 nm thick. In zero field, the conductance was the same as that for the control junction; that is, there was a single sharp peak at the voltage corresponding to the sum of the energy gaps of the two Al films. As the field was applied, this peak split into two. The splitting increased with applied field as shown by the filled circles in Fig. 3. The curve in Fig. 2 shows a

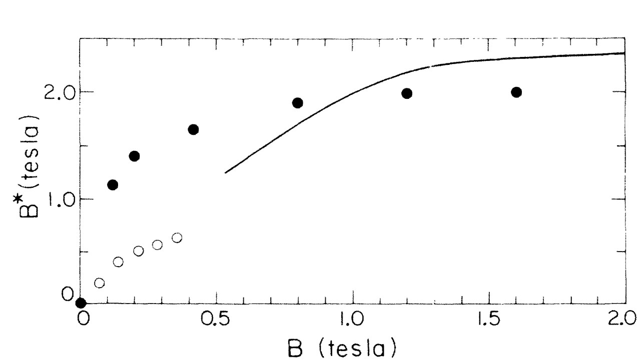

FIG. 3. Observed values of B^* vs applied field B for two EuO-Al films. The magnetization $\mu_0 M$ for EuO from Ref. 8 is shown by the solid line.

splitting equivalent to an enhanced field B^* of 1.73 T in an applied field of 0.44 T. The conductance of the control junction did not show splitting at any field. These results are in accordance with the discussion of Fig. 1(d). Above the critical field of the EuO-Al film, we obtained the symmetrical spin-split curves of the type shown in Fig. 1(b) (because the Al counter electrode was still superconducting), indicating that the electrons tunneling from the EuO-Al film were not polarized in the normal state. Otherwise, asymmetrical curves of the type shown in Fig. 1(c) would result. As expected, the normal Al film is not strongly affected by being in contact with a ferromagnet.

The normalized tunneling conductance for a EuO-$Al/Al_2O_3/Fe$ junction in a magnetic field is shown in Fig. 4(a). A curve in an applied field 0.8 T higher for an $Al/Al_2O_3/Fe$ junction on the same substrate is shown for comparison. The two curves are nearly identical, showing that the enhanced field causes the same depairing as an equivalent applied magnetic field. Analysis of these data by the method of Ref. 11 produced the spin-resolved conductances shown in Fig. 4(b). Comparison of these curves with earlier data indicates that the spin-orbit scattering is small in both films, with $b \sim 0.05$. Thus the proximity of the EuO does not affect the strength of the spin-orbit scattering. The dependence of B^* on applied field for this

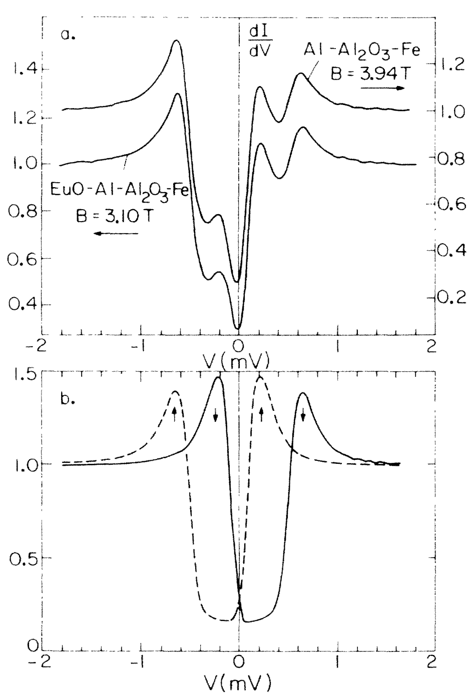

FIG. 4. (a) Normalized tunneling conductances of a EuO-$Al/Al_2O_3/Fe$ junction in an applied field of 3.1 T and an $Al/Al_2O_3/Fe$ junction in an applied field of 3.94 T. The curves have been offset vertically for clarity. (b) The spin-resolved conductance for the curves in *a*.

sample is shown by the open circles in Fig. 3, demonstrating that the size of B^* was sample dependent.

In Fig. 5, the parallel critical magnetic field $H_{c\parallel}$ of an EuO-Al film is compared to that of an Al film deposited at the same time but not in contact with the EuO, the same pair of films as produced the tunneling curves of Fig. 4. We see that $H_{c\parallel}$ for the EuO-Al film is about 0.9 T lower than $H_{c\parallel}$ for the plain Al film. This result is consistant with the tunneling results.

To summarize our findings, the EuO-Al layered system behaves like a superconductor with an internal magnetic field. The mechanism involved is presumably that the quasiparticle density of states is altered because the quasiparticles can tunnel some distance into the ferromagnet and are subjected to its exchange field or its magnetization. The fact that B^* is of the same magnitude as $\mu_0 M$, the magnetization of EuO, suggests the coupling of the quasiparticles to be with M. The exchange coupling between conduction electrons and the Eu ions has been estimated by Penney, Schafer, and Torrance[7] to be about 100 meV, 10^3 times our observed splitting. However, a proximity model described by de Gennes[13] in which the coupling is to the exchange field can account qualitatively for the results (see also Ref. 3). In this model, the coupling would be reduced by the ratio of the atomic spacing in EuO to the thickness of the Al film and by interface degradation effects such as Al oxide between the Al and EuO. The similarity between B^* and $\mu_0 M$ would then be coincidental. In either case, since the Al film is very thin compared to the coherence length and the spin mean free path, the effect of the ferromagnet is uniform throughout the film. The observed combination of superconductivity and apparent internal field results. The boundary conditions for a magnetic field applied parallel to a thin slab of ferromagnet guarantee that for a smooth interface, the quasiparticles in the Al would experience only the applied field B if they did not tunnel into the EuO. Any explanation of an enhanced field in the Al based on

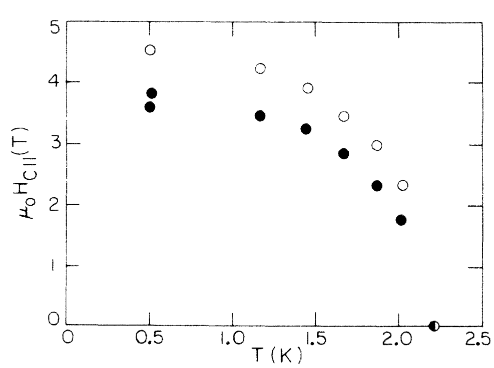

FIG. 5. Critical magnetic field vs temperature for two similar Al films, one of which (solid circles) was in contact with EuO.

roughness of the interface is difficult to construct because the tunneling curves are very sharp and are very sensitive to perpendicular magnetic fields. The Al film has a perpendicular critical field of only 0.08 T; a perpendicular field a fraction of this magnetic would cause orbital depairing and, consequently, broadening of the conductance peaks. Also, the Al counter electrode, only 2 nm away, did not experience any enhanced field. As a further check of the tunneling model, we made junctions with a layer of Al_2O_3 approximately 2 nm thick between the Al and the EuO. No splitting was seen at any field; the junctions behaved as though there were no EuO present.

We note that the phenomenon described here is not the same as that observed in zero field by Stageberg *et al.*[6] in the tunneling conductance of $Pb/Ho(OH)_3/Pb$ junctions. First, the Pb would be expected to have a high rate of spin-orbit scattering which would not allow the splitting to be seen. Second, both superconductors would have experienced the same enhanced field since they were both in contact with the ferromagnet. Again, no splitting would be seen [see discussion of Fig. 1(d)]. Conversely, we have not observed the interface states described by DeWeert and Arnold[14] in connection with the results of Ref. 6.

Finally, we mention that the ability to subject a superconductor to a local, intense magnetic field by applying a small one may have applications in electronic devices.

We thank S. Foner and R. Guertin for information about EuO. We appreciate discussions of the bound-state experiment with G. Arnold and R. Cantor prior to publication. We are grateful for encouragement and advice from D. Rainer. The films were made by R. K. MacNabb. The Francis Bitter National Laboratory is supported at the Massachusetts Institute of Technology by the National Science Foundation.

[1]W. A. Fertig, D. C. Johnston, L E. DeLong, R. W. McCallum, M. B. Maple, and B. T. Matthias, Phys. Rev. Lett. **38**, 987 (1977).

[2]M. Ishikawa and O. Fischer, Solid State Commun. **23**, 37 (1977).

[3]H. W. Meul, C. Rossel, M. DeCroux, Ø. Fischer, G. Remanyi, and A. Briggs, Phys. Rev. Lett. **53**, 497 (1984). Ø. Fischer, H. W. Meul, M. G. Karkut, G. Remenyi, U. Welp, J. C. Piccoche, and K. Maki, Phys. Rev. Lett. **55**, 2972 (1985).

[4]C. L. Lin, J. Teeter, J. E. Crow, T. Mihalisin, J. Brooks, A. I. Abou-Aly, and G. R. Stewart, Phys. Rev. Lett. **54**, 2541 (1985).

[5]For example, C. Uher, R. Clarke, G. Zheng, and I. K. Schuller, Phys. Rev. B **30**, 453 (1984). See also *Synthetic Modulated Structure Materials*, edited by L. Chang and B. C. Giessen (Academic, New York 1984).

[6]F. Stageberg, R. Cantor, A. M. Goldman, and G. B. Arnold, Phys. Rev. B **32**, 3292 (1985).

[7]A review of early work on EuO is contained in S. Methfessel and D. C. Mattis, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1968), Vol. 18, Pt. 1; G. Petrich, S. von Molnar, and T. Penney, Phys. Rev. Lett. **26**, 885 (1971); T. Penney, M. W. Shafer, and J. B. Torrance, Phys. Rev. B **5**, 3669 (1972).

[8]Y. Shapira, S. Foner, R. L. Aggarwal, and T. B. Reed, Rev. B **8**, 2316 (1973).

[9]R. Meservey, P. M. Tedrow, and P. Fulde, Phys. Rev. Lett. **25**, 1270 (1970); see also P. Fulde, Adv. Phys. **22**, 667 (1973).

[10]P. M. Tedrow and R. Meservey, Phys. Rev. Lett. **26**, 192 (1971).

[11]P. M. Tedrow, J. S. Moodera, and R. Meservey, Solid State Commun. **44**, 587 (1982).

[12]P. M. Tedrow and R. Meservey, Phys. Rev. Lett. **27**, 919 (1971); R. Meservey, P. M. Tedrow, and R. C. Bruno, Phys. Rev. **11**, 4224 (1975).

[13]P. G. de Gennes, Phys. Lett. **23**, 10 (1966).

[14]M. J. DeWeert and G. B. Arnold, Phys. Rev. Lett. **55**, 1522 (1985).