Thin-Film Superconductor in an Exchange Field

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Using the technique of spin-polarized tunneling, we studied tunnel junctions with EuS/Al bilayer electrodes. We found a large effective internal field in the Al film, which gives rise to extra Zeeman splitting in the superconducting quasiparticle density of states and which is attributed to the exchange interaction between the Eu ions and the Al conduction electrons. This exchange field, acting only on the electron spins, is inversely proportional to the thickness of the Al, causes no observable orbital depairing in the Al, and leads to a first-order transition to the normal state.

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Following the publication of the microscopic theory of superconductivity [1] with its assumption of electron pairs with opposite spins, the effect of an exchange field (which acts only on electron spins, not on electron motion) on superconductivity was soon investigated. In the limit that the spin-orbit interaction was neglected in the superconductor, theories predicted that the critical field of a superconductor is limited by the field-spin interaction (the Chandrasekhar-Clogston limit [2]) and that the field-spin interaction causes the superconductor to go normal through a first-order phase transition at low temperatures [3]. In general, when a superconductor is subjected to an applied magnetic field, the effect of the field on electron motion (orbital effect) is more important than the effect of the field on electron spins. However, for extremely thin films in a parallel magnetic field, orbital effects are greatly reduced, and the field-spin interaction becomes predominant. The above predictions of spin effects and their refinement which included spin-orbit scattering in the superconductor were later verified experimentally using very thin superconducting films in a parallel magnetic field [4]. Also, the effect of the exchange field on the superconducting state has been investigated extensively in the ternary compounds in which magnetic ions are uniformly distributed [5].

In this work, we show that there is a way to "apply" an exchange field on a superconductor, which leads to a first-order transition to the normal state. In studying Au/EuS/Al junctions, in which the EuS serves as a tunnel barrier, we found, in addition to the high degree of electron-spin polarization (up to 85%) in the tunnel current [6], a very large extra Zeeman splitting in the quasiparticle density of states (DOS) in the Al. For instance, in one Au/EuS/Al junction, an applied field of 0.3 T produced a Zeeman splitting in the tunnel conductance curve corresponding to a total field of 4.3 T: The effective internal field B^* was 4 T. In fact, in some cases the internal field existed before any external field was applied. This extra Zeeman splitting is similar to, but much more pronounced than, those observed in junctions with oxidized-Eu/Al bilayer electrodes [7].

de Gennes discussed the coupling between two ferromagnetic insulators (FMI) through a superconducting (SC) film in a FMI/SC/FMI sandwich [8]. His argument can be applied to the bilayer of a ferromagnetic insulator and a thin-film superconductor, such as the EuS/Al system. The large internal field in the Al film results from the exchange interaction between the ferromagnetically ordered magnetic ions in the EuS and the conduction electrons in the Al. de Gennes's simple argument indicates that the superconductor in the FMI/SC proximity bilayer is equivalent to a superconductor in a uniform exchange field; it also predicts that the internal field is inversely proportional to the thickness d of the superconducting film for $d < \xi$, where ξ is the superconducting coherence length. Tokuyasu, Sauls, and Rainer [9] recently proposed a model to explain the observations of Tedrow, Tkaczyk, and Kumar [7]. One of the predictions of their model is that the superconductor in a FMI/SC proximity bilayer is *not* equivalent to a superconductor in a uniform exchange field; as a result, the transition to the normal state due to the exchange interaction at the interface is always of second order in zero applied field (and in a small applied field, when the effect of the applied field is much smaller than the surface effect). Our study of the EuS/Al system indicates that the Al film in contact with the EuS behaves as a superconductor in a uniform exchange field.

Junctions of the type EuS/Al/Al_2O_3/Ag were used to study the proximity effect at the EuS/Al interface. First, a thin layer of EuS was deposited through a mask onto room-temperature glass substrates. Then, an Al long strip was deposited, half of which was over the EuS film and the other half directly on glass not covered by EuS. The surface of this Al long strip was subsequently oxidized by glow discharge in oxygen, at a bias voltage of 1.8 kV, for about 30 s to form the Al_2O_3 barrier. Junctions were completed by depositing six cross strips of 20 nm of Ag as counterelectrodes. These junctions show a resistance on the order of a few kΩ and have an area of about 3×10^{-3} cm^2. Three of the six junctions are formed over the part of the Al that is not in contact with EuS; they are of the type Al/Al_2O_3/Ag and serve as control junctions. The other three junctions are of the type EuS/Al/Al_2O_3/Ag and are used for the study of the proximity effect at the EuS/Al interface. Tunnel conduc-

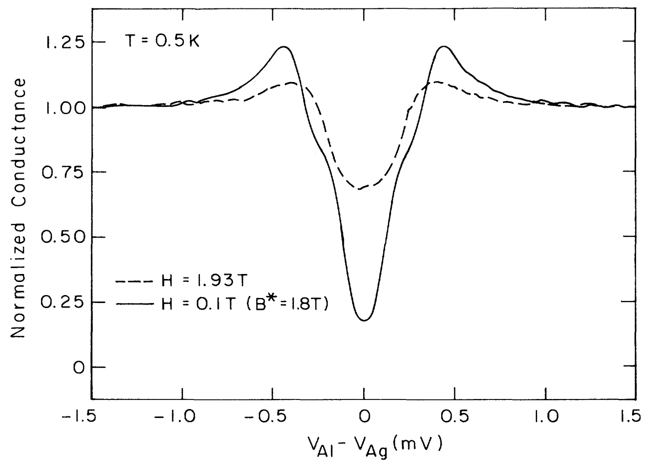

FIG. 1. The tunnel conductance of the control junction (dashed curve) was measured in an applied field of $H=1.93$ T; it is broadened by the orbital depairing of the applied field. The tunnel conductance of the junction with the EuS/Al electrode (solid curve) measured in a field of 0.1 T shows a Zeeman splitting corresponding to a total field of $B=1.9$ T, but does not show much depairing.

tances were measured in a ^{3}He cryostat at about 0.5 K and in small magnetic fields parallel to the junction surface.

All the conductance curves obtained on junctions with EuS/Al electrodes show enhanced Zeeman splitting. The internal field which gives rise to the extra Zeeman splitting does not noticeably increase the orbital depairing in the superconductor. In Fig. 1, the conductance curve of the EuS/Al/Al$_2$O$_3$/Ag junction (the solid curve) was measured in an applied field of 0.1 T. It shows two small shoulders in addition to the peaks, indicating a Zeeman splitting corresponding to a total field of 1.9 T. Also shown in Fig. 1 is the tunnel conductance curve (the dashed curve) of a control junction in an applied field of 1.93 T. The conductance curve of the control junction is very much broadened by the effect of orbital depairing, and the Zeeman splitting is not discernible because of the broadening. We see that the internal field in the EuS/Al bilayer is an exchange field: It produces a Zeeman splitting (which is a spin effect) in the quasiparticle DOS of Al, but does not cause orbital depairing in the superconductor.

For a superconductor in an exchange field, in the limit of small spin-orbit scattering (which Al satisfies) the transition to the normal state caused by the exchange field is of first order at low temperatures [3]. In a first-order phase transition the order parameter goes to zero abruptly at the transition point, whereas in a second-order phase transition, the order parameter decreases gradually and reaches zero at the transition point. Figure 2 shows the reduced order parameter $(\Delta/\Delta_{00})^2$ plotted against the reduced field $(H+B^*)^2/H_{c\parallel}^2$, for a few junctions. (Here $\Delta_{00}=1.76k_BT_{c0}$ is the unperturbed order parameter at zero temperature and $H_{c\parallel}$ is the extrapolated parallel critical field of the corresponding pure Al film.) In the case of pure Al films, which are used as a comparison, the total field is the applied field H; in the case of the EuS/Al bilayer, the total field is the applied field H plus the internal field B^*. The order parameter Δ and the internal field B^* are obtained by fitting the tunnel conductance curves using the theory of thin-film superconductors in parallel magnetic fields [10]. First the parameter c (which describes the relative strength of the magnetic-field effects on electron orbits and on electron spins) and the spin-orbit parameter $b_{\rm s.o.}$ were obtained by fitting the conductance curves of the control junctions. The c and $b_{\rm s.o.}$ values are listed in Table I; the parameter

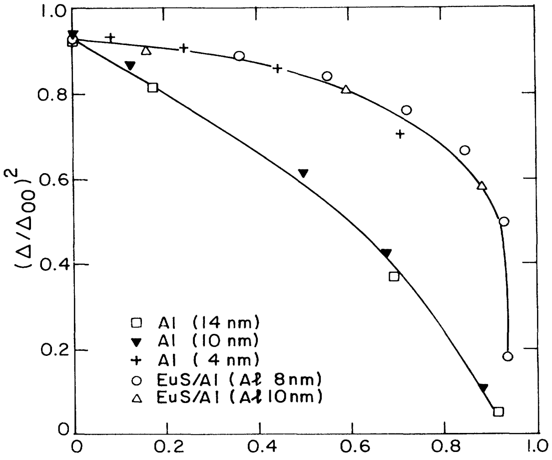

FIG. 2. The square of the reduced order parameter plotted against the square of the reduced field. The data points show that the 10- and 14-nm-thick Al films become normal through a second-order transition, which is expected from theoretical calculations. The superconductor-normal transition in the EuS/Al bilayers (each with 8 and 10 nm of Al, respectively) and in a 4-nm-thick Al film is of first order as indicated by the sharpness of the transition near the critical field.

TABLE I. Properties of thin Al films.

Control junction	Nominal Al film thickness (nm)	T_c (K)	c_F	$b_{\rm s.o.}$
6625-4	4	2.37	0.2	0.05
6723-4	4	2.50	0.15	0.03
6727-4	4	2.41	0.16	0.03
6625-6	6	2.00	0.5	0.05
6626-8	8	1.71	1.1	0.05
6668-8	8	1.64	1.05	0.04
6715-10	10	1.68	2.25	0.04
6655-12	12	1.64	3.5	0.04
6701-14	14	1.64	8.3	0.04
6668-15	15	1.44	9.0	0.02

c depends strongly on Al films thickness ($c \sim d^3$). The parameters c and $b_{\mathrm{s.o.}}$ so obtained are used as fixed parameters in fitting the conductance curves of EuS/Al/Al_2O_3/Ag junctions and the total field is separated into the applied field H and the internal field B^*. The internal field B^* is the only varying parameter in the fits and the order parameter Δ is calculated once a B^* is determined.

Numerical calculations show that, for a thin superconducting film in a parallel magnetic field, transition to the normal state is of second order for $c > 1.66$ [11]. Therefore, Al films with thicknesses of more than 10 nm ($c > 1.66$ for Al films thicker than 10 nm, see Table I) are expected to go to the normal state through a second-order phase transition. Indeed, Fig. 2 shows that the order parameters obtained from fitting control junctions with Al electrode thicknesses of 10 and 14 nm (not in contact with EuS) gradually decrease with increasing applied field, approaching zero as the applied field approaches the critical field. In contrast, a 4-nm Al film is known to become normal in a (parallel) magnetic field through a first-order phase transition at low temperatures [12]. The field dependence of the order parameter obtained from fitting the tunnel conductance curves of a control junction with a 4-nm-thick Al film is plotted in Fig. 2, along with the field dependence of the order parameter for two EuS/Al bilayers (one has an 8-nm Al film and the other has a 10-nm Al film); all three show a typical first-order behavior in which the order parameter decreases slowly with increasing field and drops to zero abruptly near the transition point. Because the orbital depairing is small, points for the EuS/Al bilayers are obtained much closer to the transition than with the 4-nm pure Al film. Note that in an applied magnetic field a 10-nm pure Al film goes to the normal state through a second-order transition, because of orbital depairing, but the 10-nm Al film in the EuS/Al bilayer goes to normal through a first-order transition owing to the internal exchange field.

Another indication that the superconducting-normal transition is first order is the zero-bias peak in the conductance curve, see Fig. 3, which results from the overlap of the Zeeman-split densities of states for spin-up and spin-down quasiparticles. The zero-bias peak usually appears right before the Al electrode becomes normal. The overlapping of conductance peaks owing to large Zeeman splitting has been observed in junctions with V electrodes and was interpreted as suggesting a first-order transition in the V from the superconducting to the normal state [13].

We also investigated the dependence of the internal field on the Al film thickness. Six sets of junctions with the EuS/Al bilayer electrode were measured. The Al film thickness varied between 4 and 15 nm. The EuS film was usually 5 nm thick. The exchange interaction with the electrons in the Al takes place in the first monolayer of EuS, so that the only reason for a greater thickness is that it is required for the EuS to be ferromagnetic, but greater EuS thickness has little effect on the exchange interaction at the interface. From fitting the conductance curves, the dependence of B^* on H is obtained. The internal field, which depends on the magnetic domain size in EuS, increases with the applied field, reaching saturation at $H \sim 0.5$ T. The saturation internal field is plotted against the inverse of the Al film thickness d in Fig. 4. The data points are fitted by a straight line using a least-squares fit. We interpret this result as showing that the average internal field varies inversely with the normalization volume of the exchange interaction, that is, as d^{-1} as predicted by de Gennes [8].

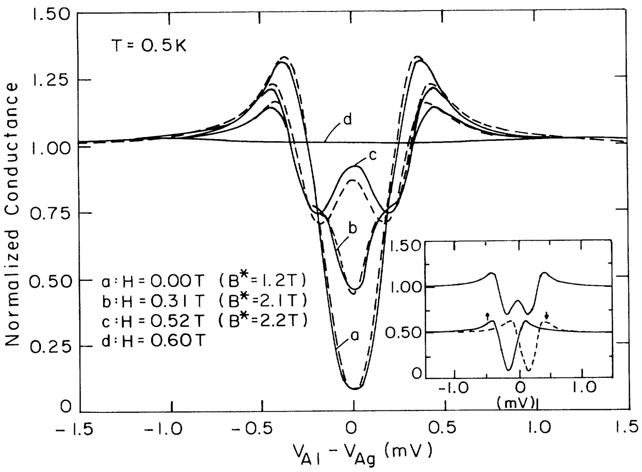

FIG. 3. Normalized tunnel conductance curves of a EuS/Al/Al_2O_3/Ag junction in various magnetic fields H. (The Al film is 10 nm thick.) The values of the internal field B^* are obtained from the fits (shown as dashed curves). The zero-bias peak is an indication that the superconducting-normal transition is of first order. Inset: The calculated total and individual-spin (labeled by the corresponding spin direction) conductance curves. The individual-spin conductance curves show a sizable gap. The merging of the peaks of individual-spin conductance curves gives rise to the zero-bias peak in the total conductance curve.

The order of the superconducting-normal phase transition in the EuS/Al bilayer indicates that the Al film behaves as a superconductor in a uniform exchange field. This disagrees with the prediction of the model proposed by Tokuyasu, Sauls, and Rainer [9] for the proximity system of a ferromagnetic-insulator/superconductor bilayer; the model shows that the interaction at the interface has the effect of rotating the spin orientation of the reflected wave function with respect to the incoming wave function of the quasiparticles. Because of the model assumption of a perfectly smooth and specularly reflecting interface, the magnetic surface gives rise to an internal exchange field in the superconductor, which depends on the quasiparticle

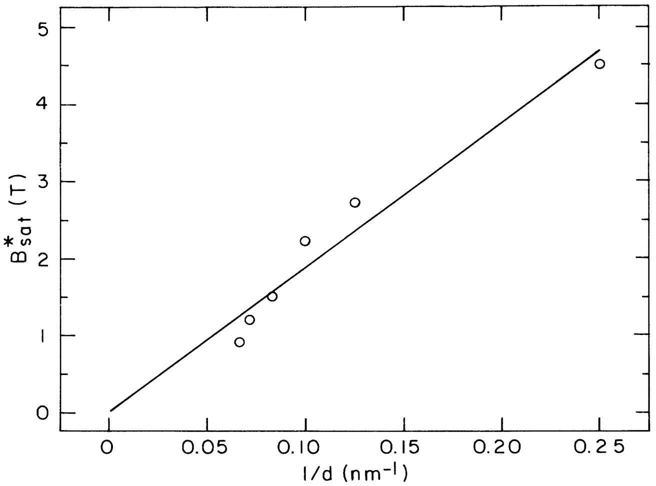

FIG. 4. The dependence of the saturation internal field on the Al film thickness. The line in the figure is a least-squares fit to the data points.

trajectory. This unusual trajectory dependence of the internal field dictates that the superconductor in the FMI/SC bilayer goes normal through a second-order transition in zero applied field. If the applied field is small, the superconducting-normal transition in the SC in contact with EuS remains second order in the model of Tokuyasu, Sauls, and Rainer. We estimate that when the internal field is greater than 1.4 times the applied field, the surface effect dominates and the transition is expected to be of second order. In all the junctions we studied experimentally, the internal field in the Al film in the EuS/Al bilayer is at least 3–4 times the external field when the Al film becomes normal, but the superconducting-normal transition in the EuS/Al is found to be of first order. We conclude that the assumption that the interface is specularly reflecting is not justified. That the surface scattering is mostly diffuse is also borne out by the film-thickness dependence of the resistivity in these Al films when analyzed by accepted theory [14].

An important property of this technique of "applying" a pure exchange field on a superconductor is that it allows measurements of spin properties of many superconductors which were previously impossible because of orbital depairing.

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