Superconducting Spin Switch with Infinite Magnetoresistance Induced by an Internal Exchange Field

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A theoretical prediction by de Gennes suggests that the resistance in a FI/S/FI (where FI is a ferromagnetic insulator, and S is a superconductor) structure will depend on the magnetization direction of the two FI layers. We report a magnetotransport measurement in a EuS/Al/EuS structure, showing that an infinite magnetoresistance can be produced by tuning the internal exchange field at the FI/S interface. This proximity effect at the interface can be suppressed by an Al_2O_3 barrier as thin as 0.3 nm, showing the extreme confinement of the interaction to the interface giving rise to the demonstrated phenomena.

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The proximity effect between a ferromagnetic metal (F) and a superconductor (S) has attracted considerable attention because of the rich physics governing the competition between the ferromagnetism and the superconductivity [1,2]. A number of theoretical and experimental works have reported that the superconducting transition temperature T_C of the superconductor in metallic F/S/F as well as in F1/F2/S sandwich structures depends on the relative orientation of the magnetization of the two ferromagnetic layers [3–18]. However, the interplay between the superconductivity and ferromagnetism in a metallic system is complicated, since a spin-polarized current flows from the F layer through the S layer. The resistances for the parallel (R_P) and antiparallel (R_{AP}) configurations, both normal ($R_P > R_{AP}$) [6,7,9,15] and inverse ($R_P < R_{AP}$) [8,10,14], spin switch effects have been reported in this type of metallic system. A more ideal and well-defined system would consist of a ferromagnetic insulator (FI) instead of a ferromagnetic metal. In the FI/S/FI system the proximity effect is limited to the interface because the wave function of the electron decays in the insulator within an atomic distance [2]. Such a case was discussed by de Gennes nearly five decades ago [19]. When two ferromagnetic insulators couple through a superconducting layer, the average exchange field seen by a conduction electron is

$$\bar{h} = 2|\Gamma|S(a/d_s)\cos(\theta/2), \tag{1}$$

where $2\Gamma S \cdot S_e$ is the exchange coupling between a ferromagnetic ion spin S and a conduction electron with spin S_e, θ is the angle between the magnetization of the two ferromagnetic layers, and a and d_s are the lattice constant and thickness of the superconductor, respectively [19]. In the strong exchange field scenario, namely, $h(0) > \frac{\sqrt{2}}{2}\Delta$ [where $h(0)$ represents $h(\theta = 0)$ and Δ is the bulk BCS gap], the system is expected to show zero resistance when the two ferromagnetic layers align antiparallel ($\theta = 180°$), whereas it would show a finite resistance when they are parallel ($\theta = 0$) [19]. This system was theoretically studied in further detail by Kulić *et al.*, with similar results [20]. It has been shown experimentally that in the Fe_3O_4/In/Fe_3O_4 trilayer structure, the superconducting transition temperature T_C of In depended on the relative magnetization alignment of the two magnetic Fe_3O_4 layers [21]. In this Letter, we report magnetotransport measurements in a FI/S/FI system with europium sulfide (EuS) as the ferromagnetic insulator, which for thick films has a bulk magnetic ordering temperature $T_C \sim 16.6$ K [22–25]. We demonstrate infinite magnetoresistance and well-defined sharp resistance switching between the superconducting and normal states by controlling the exchange field at the interface. At zero field in the remanence state of magnetization, two clear resistance states are maintained, creating nonvolatile memory states. Our results clearly confirm de Gennes' prediction, showing that the intrinsic superconductivity is affected through the coupling of two ferromagnetic insulators.

All of our films were deposited on precleaned glass substrates that were processed by an oxygen plasma to further remove the organic residue. An Al_2O_3 seed layer with a thickness of 1 nm was first deposited on the glass substrate to facilitate smooth film growth. The substrates were cooled to liquid nitrogen temperature in a thermal deposition system with a base pressure of $\sim 2.0 \times 10^{-8}$ Torr. The thin film layer structure was EuS(1.5)/Al(3.5)/EuS(4) (thickness in nanometers) (see Fig. 1). Different thicknesses were chosen for the two EuS layers to give rise to different coercivities, making it

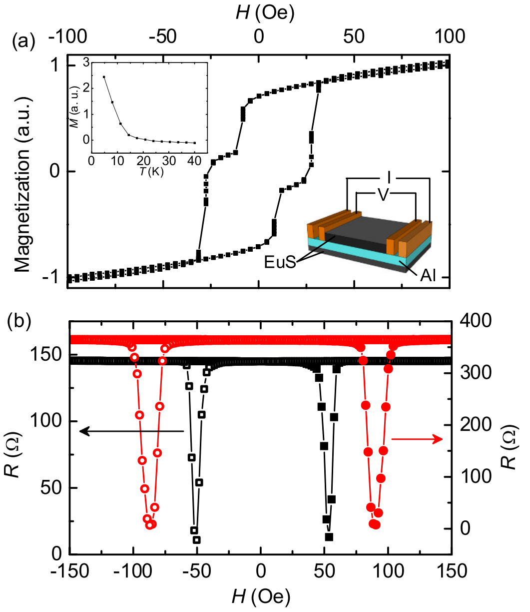

FIG. 1 (color online). (a) Magnetic hysteresis loop of EuS(1.5)/Al(3.5)/EuS(4) structure at 2 K. Top inset: magnetization as a function of temperature measured with $H = 500$ Oe. Bottom inset: schematic view of the device structure. (b) Resistance as a function of the external magnetic field H for bilayer structures at 1.2 K (thickness in nanometers): EuS(1.5)/Al(3.5) (black squares) and Al(3.5)/EuS(4) (red circles). The closed (open) data points are for increasing (decreasing) field.

possible to achieve an antiparallel alignment. The Al film thickness was optimized to be 3.5 nm. We could not obtain continuous films with reduced thicknesses, while the exchange field decreases in thicker Al films, which affects sharp and clean resistive transitions [24,26]. After the film growth, the samples were capped with 4 nm Al_2O_3 protection layers. All the low temperature measurements were performed in a pumped liquid ^{4}He bath with samples immersed in the liquid, and the temperature was determined from the ^{4}He vapor pressure (temperature stability ±2 mK). The transport measurements were conducted using a LR-700 ac resistance bridge with an in-plane magnetic field. The samples were of macroscopic size with typical dimensions of $5\ \mathrm{mm} \times 25\ \mathrm{mm}$ to avoid Joule heating at the probe current of 1 μA. The magnetization measurements were conducted with a superconducting quantum interference device (SQUID) magnetometer from Quantum Design.

Figure 1(a) shows the magnetization $M(H)$ of a EuS/Al/EuS structure as a function of the external magnetic field at 2 K. From the $M(H)$ loop, we see two distinct coercive fields for the EuS layers: $H_c^1 \sim 10$ Oe (1.5 nm layer) and $H_c^2 \sim 30$ Oe (4 nm layer). These coercivities are similar to our results reported previously [25] that showed that a EuS film grown at 77 K is magnetically soft compared with a room-temperature grown film. The magnetic ordering Curie temperature is approximately 16 K obtained from the $M(T)$ curve [see Fig. 1(a)]. We first investigated the magnetotransport characteristics of EuS(1.5)/Al(3.5) and Al(3.5)/EuS(4) bilayer structures. These two bilayer structure geometries correspond to the bottom and top sections of the EuS/Al/EuS structure with the same thicknesses. The resistance of the films was recorded at 1.2 K as a function of the applied magnetic field. The external field H was swept from -200 Oe to 200 Oe and then back to -200 Oe. As shown in Fig. 1(b), we observed steep dips in the resistance at specific values of the magnetic field and clear hysteresis behavior in both structures. A similar dramatic drop in resistance due to the onset of superconductivity has been reported in a $Ni_{0.80}Fe_{0.20}$ (Py)/Nb bilayer structure [27]. We attribute this large resistance drop to the weakening or disappearance of the exchange field from the EuS film in the vicinity of the switching field. If the magnetic domain size is comparable to or larger than the superconductor coherence length ξ_s, the Cooper pair will feel a uniform exchange field as large as several tesla [1,23,24,28,29]. The experimentally determined intrinsic coherence length at $T = 0$ for Al is $\xi_0 = 1.6\ \mu$m [30,31], and follows a temperature dependence of $\xi(T) \propto \frac{\xi_0}{\sqrt{1-T/T_C}}$ [32]. In thin films, the coherence length ξ_s is defined by $\sqrt{\xi_0 l}$ as the dirty limit, where l is the mean free path. For a 3.5 nm Al film ξ_s is about 79 nm, using $l = 3.9$ nm from the resistivity measurement. Near the magnetization switching (coercive) field, in these ultrathin polycrystalline EuS films the domain size is expected to become much smaller than ξ_s and thus the net field seen by Al would be dramatically reduced. Thus the average exchange field experienced by quasiparticles in the superconductor is much smaller and the pair breaking effect is weaker, leading to the recovery of superconductivity. Interestingly, the switching fields in the $R(H)$ curves did not match the coercive fields obtained from the $M(H)$ loop, but were increased to 50 Oe (1.5 nm) and 85 Oe (4 nm).

Next we discuss the behavior of the EuS/Al/EuS trilayer structure. The $R(H)$ data for the EuS(1.5)/Al(3.5)/EuS(4) sample are shown in Fig. 2. By sweeping H, the relative alignment of the magnetic moments in the two EuS layers could be switched between the parallel (P) and antiparallel (AP) configurations. At high fields where the magnetizations of the two EuS layers are aligned parallel, the sandwiched Al layer shows a normal state resistance which is about 110 Ω. As H is reduced and reversed, the magnetization of the 1.5 nm EuS layer switches first, giving rise to the AP configuration, where the resistance of the Al film drops to zero dramatically. [The residual resistance ($< 2 \times 10^{-3}$ Ω) was within the fluctuation of the LR-700 ac resistance bridge.] This demonstrates that the Al transitions from a normal state to a superconducting state. Further increase of the field in the reverse direction brings back the normal state resistance. We define the magnetoresistance as $\mathrm{MR} = [(R_{\max} - R_{\min})/R_{\min}] \times 100\%$, where

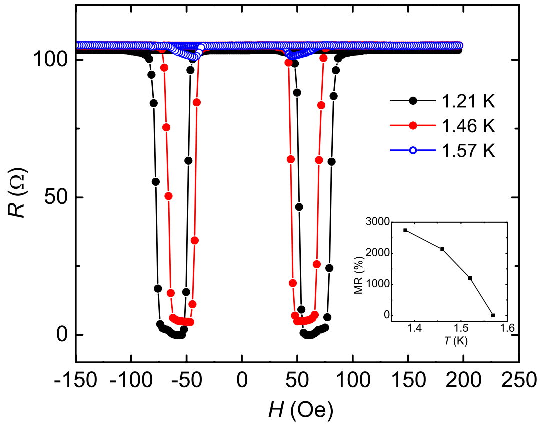

FIG. 2 (color online). Resistance as a function of the external magnetic field H at different temperatures for the EuS(1.5)/Al(3.5)/EuS(4) structure. The change of resistance diminishes with increasing temperature. Inset: magnetoresistance as a function of temperature. For $T < 1.25$ K, the MR values lead to infinity.

R_{max} and R_{min} are the maximum and minimum resistance values, corresponding to the P and AP configurations, respectively. Since the Al film undergoes a complete transition from the normal state to the superconducting state, an infinite magnetoresistance (MR) results. It is worth noting that the zero resistance state forms a plateau in the $R(H)$ curve. Again, these switching fields did not match the coercive fields from the $M(H)$ loop, but they match the switching fields seen in the $R(H)$ plots for the EuS/Al and Al/EuS bilayer structures.

Another important feature in the $R(H)$ curves is the strong temperature dependence of the MR, shown for slightly higher temperatures (see the inset of Fig. 2). This temperature dependence is further identified by comparing the superconducting transition temperature T_C of the Al layer in both the P and AP states (see Fig. 4). We obtained $T_C^{\mathrm{AP}} \sim 1.55$ K for the AP state using a midpoint definition. However, T_C^{P} for the P state is below the temperature range in our pumped ^{4}He bath cryostat which is 1.0 K. In the range $T_C^{\mathrm{P}} < T < T_C^{\mathrm{AP}}$, the resistance change reaches its maximum value, namely, the complete normal-superconducting transition. When T reaches the vicinity of T_C^{AP}, we see a partial transition. The resistance change was negligible for $T > T_C^{\mathrm{AP}}$. We would like to point out that this infinite MR in EuS/Al/EuS is different from the infinite MR we reported in the Fe/V/Fe metallic system [15]. In a metallic F/S/F system, in addition to the presence of the exchange field, another mechanism also plays an important part which is the spin-polarized current flowing from one F through the S to the other F. The relative alignment of M in the two F layers acts as a valve to control the supercurrent flowing through the S [4]. Here what we observed is the intrinsic superconductivity of the Al film influenced by the exchange field of the insulating EuS films, acting on the quasiparticle spins. In our chosen 3.5 nm ($\ll \xi_s$) Al films in the trilayer structures, all the quasiparticles experience the exchange field at both interfaces. Thus in the P configuration the net exchange field is much higher than the typical spin critical field of the 3.5 nm Al film [26], whereas in the AP configuration the net exchange field seen becomes negligible or zero, thus leading to the full recovery of superconductivity.

Sharp and reliable resistance switching is a prerequisite for candidates of memory and logic circuit applications. To demonstrate this characteristic, we performed resistance measurement while varying the H field by steps, maintaining the P or AP configuration of M, as shown in Fig. 3. Well-defined switching between the normal and superconducting states was realized. As soon as the magnetization configuration is switched, the system undergoes a resistive transition with an infinite MR. Note that we have two distinct resistance states with zero applied field: one for the AP state and one for the P state, depending on the magnetic history. This characteristic can lead to nonvolatile memory applications.

The superconducting spin switch effect is also observed in trilayer structures with thicker Al films. The superconducting transition temperature was found to increase in a trilayer with a 5 nm Al film, but with a smaller difference between T_C^{P} and T_C^{AP} of 0.02 K. This is expected in de Gennes' model, as the exchange field is inversely proportional to the film thickness [19,24,33]. In the metallic F/S/F system, theoretical calculations carried out by Buzdin *et al.* showed that the interface transparency is an important control parameter for the superconducting spin valve effect [1], which is usually tuned by thin barriers at the F/S interfaces [11,34]. We emphasize this point in the present experiment with a clean FI/S/FI system because

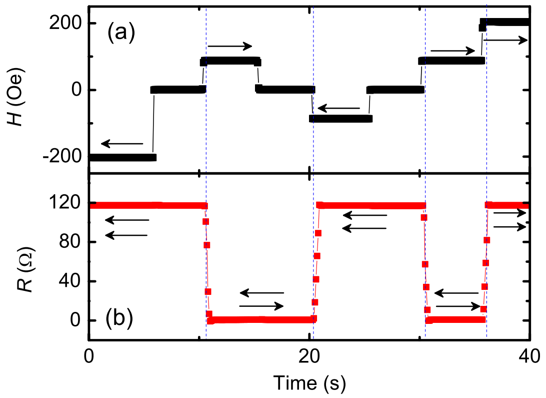

FIG. 3 (color online). Stepwise increase and decrease of the applied magnetic field (a) as a function of time show a corresponding change in the measured resistance (b) of the EuS/Al/EuS trilayer at 1.2 K: resistance switches between the normal and superconducting states at fixed field values. Note that the trilayer device can be in either the high or zero resistance state at $H = 0$. The arrows denote the directions of the magnetic moment in the two EuS layers.

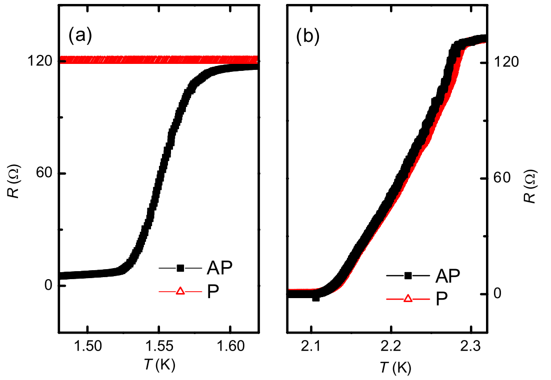

FIG. 4 (color online). Temperature-dependent resistance for the parallel (P) and antiparallel (AP) configurations for (a) EuS(1.5)/Al(3.5)/EuS(4), and (b) EuS(1.5)/Al_2O_3(0.3)/Al(3.5)/Al_2O_3(0.3)/EuS(4). No MR transition is observed with the Al_2O_3 layers inserted. Black data squares are for the antiparallel state and red data triangles are for the parallel state.

the wave function of the conduction electrons decays on an atomic scale in the insulator [2,24].

To demonstrate that the suppression of superconductivity is due to the exchange field, and is interface sensitive, we changed the interface transparency. We fabricated a set of control samples by inserting an ultrathin Al_2O_3 barrier between the EuS magnetic insulator and the Al film for the sample structure EuS/Al_2O_3/Al/Al_2O_3/EuS. The superconducting transition temperatures were obtained for both the P and AP states for these devices with the Al_2O_3 thickness ranging from 0.3 to 0.9 nm. As shown in Fig. 4 for both cases, contrary to the samples with transparent interfaces, no MR transition for the P and AP states was found in these control samples. Al_2O_3 barriers as thin as 0.3 nm at the interface were enough to destroy the proximity effect. Also, it is observed that T_C is much higher in the EuS/Al_2O_3/Al/Al_2O_3/EuS sample compared with T_C^{AP} of the EuS/Al/EuS structure, indicating less interplay between the superconductivity and ferromagnetism in the former due to the blocking of the exchange field by the barrier, whereas in the EuS/Al/EuS trilayers possible unaligned Eu^{2+} magnetic moments at the EuS/Al interfaces may act as magnetic impurities lowering T_C [35,36]. Given the extreme sensitivity of the superconductivity to magnetic impurities, which in the present case of the Eu^{2+} ion carry 7 μ_B per Eu^{2+} magnetic moment, a T_C change is not surprising.

Here we discuss the noticeable difference between the coercive fields H_c obtained from the $M(H)$ loop and the switching fields obtained from the $R(H)$ curves. There are two plausible explanations for this differences. First, we observe a near doubling of the coercive fields in the EuS films from 4 to 2 K from SQUID measurements. It is likely this increasing trend in H_c may continue below 2 K (whereas the SQUID system is limited to 2 K). Alternatively, the above difference could come from the surface or interface anisotropy of EuS. Considering that the present films were deposited onto liquid N_2 cooled substrates, the interfaces are expected to be smooth and sharp [37]. The $M(H)$ loop is a manifestation of the collective average of the ensemble of all the domains. Given the observation that a 0.3 nm Al_2O_3 barrier completely prevented the proximity effect, it is clear that the interface magnetization of EuS controls the switching.

We estimated the value of the exchange integral Γ for the interface, by examining the shift in T_C for the P and AP alignments, as was shown for the Fe_3O_4/In/Fe_3O_4 system [21]. We obtain Γ for our EuS/Al interface, by fitting the experimental data to the formula

$$T_C/T_{C0} = 1\text{–}10(\Gamma S/E_F)(\sqrt{\xi_0 l}/d), \qquad (2)$$

where T_{C0} is the transition temperature of the pure superconductor film (without the adjacent magnetic layer), S is the spin angular momentum of Eu^{2+}, E_F is the Fermi energy of Al, $\sqrt{\xi_0 l}$ is the coherence length in the dirty limit, and d is the Al film thickness [21]. Although T_C^P for the trilayer with 3.5 nm Al is below our available temperature range, we can use our lowest temperature of 1 K for a rough estimation. Given that T_C vs $1/d$ follows a linear relation [21], we can estimate Γ from the slope. We obtain $\Gamma_{min} = 16$ meV using as parameters $T_{C0} = 2.22$ K [from Fig. 4(b)], $S = 7/2$, $E_F = 11.6$ eV, $\xi_0 = 1600$ nm. and $l = 3.9$ nm [26,31]. The actual value of Γ should be larger than 16 meV because T_C^P for the 3.5 nm Al trilayer is lower than 1 K. Substituting this Γ value into Eq. (1), we obtain the exchange field $h(0) = 13$ meV. Using $T_{C0} = 2.22$ K and the BCS relation, we obtain a BCS gap of 0.68 meV for our 3.5 nm Al film. Comparing this gap to $h(0)$, we are in the strong exchange field condition.

In conclusion, we studied the transport properties of a superconductor subjected to an exchange field using a FI/S/FI sandwich structure with the ferromagnetic insulator EuS. We demonstrated switching between the superconducting and normal states by tuning the proximity effect induced by the exchange field at the EuS/Al interfaces. Clean and sharp transitions, as well as an infinite MR has been realized, confirming the theoretical prediction of de Gennes [19]. This system has potentials for logic circuits and memory applications. It also provides a platform to engineer structures with an s-wave superconductor and a ferromagnetic insulator in the search for Majorana fermions [38].

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