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/Al2O3/EuS structure, showing an infinite magnetoresistance can be produced by tuning the internal exchange field at the FI/S interface. This proximite effect at the interface can be suppressed by an Al2O3 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 proximite 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

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

[where $h(0)$ represents $h(\theta = 0)$ and $\Delta$ is the bulk BCS gap], the system is expected to show zero resistance when the two ferromagnetic layers align antiparallel ($\theta = 180^\circ$), whereas it would show a finite resistance when they are parallel ($\theta = 0$) [19]. This system was theoretically studied in further detail by Kulic et al., with similar results [20]. It has been shown experimentally that in the Fe3O4/In/Fe3O4 trilayer structure, the superconducting transition temperature $T_C$ of In depended on the relative magnetization alignment of the two magnetic Fe3O4 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 Al2O3 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 $\sim 2.0 \times 10^{-9}$ 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
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$_{0.8}$Fe$_{0.2}$ (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 $\xi_s$, 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 $\xi_s$ is defined by $\sqrt{\xi_0^2}$ as the dirty limit, where $l$ is the mean free path. For a 3.5 nm Al film $\xi_s$ is about 79 nm, using $l = 39$ 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 $\xi_s$, 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 $\Omega$. 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 $(< 2 \times 10^{-3} \Omega)$ 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 $MR = [(R_{\text{max}} - R_{\text{min}}) / R_{\text{min}}] \times 100\%$, where

![Figure 1](https://example.com/figure1.png)

**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 temperature measured with $H = 500$ Oe and typical dimensions of magnetic field. The samples were of macroscopic size 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
influenced by the exchange field of the insulating EuS observed is the intrinsic superconductivity of the Al film. 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.

\[ R_{\text{max}} \text{ and } R_{\text{min}} \text{ 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) \text{ curve. Again, these switching fields did not match the coercive fields from the } M(H) \text{ loop, but they match the switching fields seen in the } R(H) \text{ plots for the EuS/Al and Al/EuS bilayer structures.}

Another important feature in the } R(H) \text{ 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 \text{ of the Al layer in both the P and AP states (see Fig. 4). We obtained } T_{C}^{\text{AP}} \sim 1.55 \text{ K for the AP state using a midpoint definition. However, } T_{C}^{\text{P}} \text{ for the P state is below the temperature range in our pumped } ^4\text{He bath cryostat which is } 1.0 \text{ K. In the range } T_{C}^{\text{P}} < T < T_{C}^{\text{AP}}, \text{ the resistance change reaches its maximum value, namely, the complete normal-superconducting transition. When } T \text{ reaches the vicinity of } T_{C}^{\text{P}}, \text{ we see a partial transition. The resistance change was negligible for } T > T_{C}^{\text{AP}}. \text{ 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 \text{ 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.}

![Figure 2](image_url)

**FIG. 2** (color online). Resistance as a function of the external magnetic field } H \text{ 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 \text{ K, the MR values lead to infinity.}

films, acting on the quasiparticle spins. In our chosen 3.5 nm (\( \ll \xi_p \)) 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 \text{ field by steps, maintaining the P or AP configuration of } M, \text{ 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}^{\text{P}} \text{ and } T_{C}^{\text{AP}} \text{ of } 0.02 \text{ 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}

![Figure 3](image_url)

**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. \text{ The arrows denote the directions of the magnetic moment in the two EuS layers.}
faces may act as magnetic impurities lowering $T_C$ due to the blocking of the exchange field by the magnetic impurities, which in the present case of the magnetic moments at the EuS/Al2O3 interface, by examining the shift in $T_C$ for the P and AP alignments, as was shown for the Fe3O4/In/Fe3O4 system [21]. We obtain $\Gamma$ for our EuS/Al interface, by fitting the experimental data to the formula

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

where $T_{C0}$ is the transition temperature of the pure superconductor film (without the adjacent magnetic layer), $S$ is the spin angular momentum of Eu2+, $E_F$ 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 $T_{C0}$ 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 $\Gamma$ 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 $\Gamma$ should be larger than 16 meV because $T_{C0}$ for the 3.5 nm Al trilayer is lower than 1 K. Substituting this $\Gamma$ 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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