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Citation: Li, Bin, Guo-Xing Miao, and Jagadeesh S. Moodera. "Observation of Tunnel Magnetoresistance in a Superconducting Junction with Zeeman-Split Energy Bands." Phys. Rev. B 88, no. 16 (October 2013). © 2013 American Physical Society

As Published: <http://dx.doi.org/10.1103/PhysRevB.88.161105>

Publisher: American Physical Society

Persistent URL: <http://hdl.handle.net/1721.1/88763>

Version: Final published version: final published article, as it appeared in a journal, conference proceedings, or other formally published context

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Observation of tunnel magnetoresistance in a superconducting junction with Zeeman-split energy bands

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(Received 15 August 2013; published 21 October 2013; corrected 22 October 2013)

The change of the quasiparticle density of states in a superconductor due to the Zeeman splitting can lead to highly responsive spintronic devices. This is demonstrated here in the magnetotunneling studies of a superconductor/insulator/ferromagnet tunnel junction. A tunnel magnetoresistance (TMR) as large as 36% is obtained, resulting from the conductance variation due to spin-split quasiparticle tunneling, and only occurs in the superconducting state. Our results show that in addition to the naturally existent spin imbalance in ferromagnets (upon which conventional TMR is obtained), we can manipulate tunnel conductance by tailoring spin dependent density of states with interfacial exchange fields, and even with a tunnel junction with both superconducting electrodes.

DOI: [10.1103/PhysRevB.88.161105](https://doi.org/10.1103/PhysRevB.88.161105)

PACS number(s): 71.70.-d, 73.43.Qt

In the conventional BCS superconductors spin-up and spin-down electrons form singlet Cooper pairs via the electron-phonon interaction,¹ whereas in ferromagnetic materials the parallel alignment of the electron spins is favored. Given the intrinsic incompatibility, the interplay between the superconductivity and the ferromagnetism leads to rich physics.^{2,3} It is particularly appealing to employ zero or finite resistance states of superconductors in spintronic applications. Observation of spin-triplet supercurrent has been reported in several S/FM/S structures (S: superconductor; FM: ferromagnetic metal).⁴⁻⁸ Spin-polarized quasiparticle transport over distances of several micrometers has also been demonstrated in Al.⁹⁻¹¹

The interaction between the superconductivity and the ferromagnetism can also be explored by taking advantage of the Zeeman splitting of the quasiparticle density of states (DOS).¹² This has been amply shown and studied by Meservey and Tedrow using the BCS *s*-wave superconductor Al. In the presence of an in-plane applied field, an ultrathin film of Al, with negligible orbital depairing, the quasiparticle DOS splits into separate spin-up and spin-down levels, as demonstrated by Meservey, Tedrow, and Fulde.¹²⁻¹⁴ The well separated spin-up and spin-down DOS can be expected to give rise to a spin dependent conductance, a large magnetoresistance in a tunnel junction. This is achievable by tuning the energy and/or the spin state of the tunneling electron, to create a cryogenic spintronic device.^{15,16} The splitting $2\mu H$ of the DOS is proportional to the applied magnetic field H , where μ is the Bohr magneton. In order to create enough splitting to well separate spin-up and spin-down states, an external field of the order of several tesla is required,¹⁴ which is unreasonably high, to obtain a TMR by this approach.

Alternatively, instead of an external magnetic field, an internal exchange field from an adjacent ferromagnetic insulator (FI) layer could serve to split the quasiparticle DOS.¹⁷⁻¹⁹ Compared to FM, there are considerably less studies in superconducting junctions with FI, in spite of the fact that using FI could avoid the complications of spin injection at

the FM/S interfaces. Several europium chalcogenides have been identified as FI. For example, EuO has a ferromagnetic ordering temperature of 69 K.²⁰ However, it is not stable, whereas the stable oxide Eu₂O₃ is nonferromagnetic.^{12,20} A better candidate is the stable compound EuS, with a ferromagnetic ordering temperature of 16.6 K and a band gap of 1.64 eV.^{21,22} It has been reported that the equivalent internal field of the Zeeman splitting in Al film due to EuS could be as large as 5 T.^{18,23-25} Here, we report magnetotunneling studies of a structure S/I/FM, showing a TMR response from the spin-split quasiparticle energy landscape tunneling into a ferromagnet.

As presented in Fig. 1(a), the magnetic tunnel junction (MTJ) structure investigated is EuS(4)/Al(4)/Al₂O₃/Co(8)/CoO (thickness in nanometers; for Al, 4 nm before oxidation), where materials are listed in the order in which they were deposited. The junctions were fabricated in a vacuum chamber with a base pressure $\sim 2.0 \times 10^{-8}$ Torr using *in situ* shadow masks. To facilitate the growth of smooth films, a thin Al₂O₃ (1 nm) seed layer was deposited onto chemically, and further *in situ* oxygen-plasma cleaned glass substrates. The substrate was cooled to liquid-nitrogen temperature for the growth of EuS and Al layers. After warming to room temperature (RT), a thin Al₂O₃ barrier was formed by plasma oxidization of the 4-nm Al film surface. Co film was deposited at RT over this and then slightly oxidized to form a thin layer of CoO. This antiferromagnetic CoO exchange biased the Co layer fixing its magnetization in a given direction.²⁶ Subsequently, the junctions were capped with 6 nm Al₂O₃ for protection. Low temperature magnetotunneling measurements were performed with sample immersed in a pumped liquid ⁴He bath. An in-plane magnetic field could be applied on the sample during tunneling measurements. The temperature was determined from the ⁴He vapor pressure with a temperature stability of ± 2 mK. The magnetization and the coercivity of the EuS and Co films were measured with a superconducting quantum interference device (SQUID) magnetometer from Quantum Design.

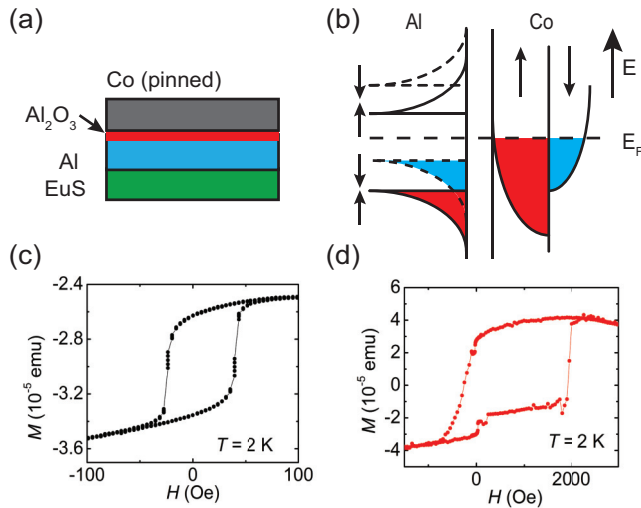


FIG. 1. (Color online) (a) Schematic view of the structure of the EuS(4)/Al(4)/Al₂O₃/Co(8)/CoO junction. The magnetization of the Co layer is pinned by the exchange bias between Co and CoO (not shown). (b) Illustration of the quasiparticle DOS of the superconducting Al using the semiconductor model. Below T_C , in the presence of the exchange field, the quasiparticle DOS is split into spin up (red) and down (blue) states. The simplified spin up (red) and down (blue) DOS at the Fermi level for the Co is also shown. (c) Minor magnetic hysteresis loop of the junction EuS(4)/Al(4)/Al₂O₃/Co(8)/CoO measured at 2 K. The coercive field of EuS film is about ± 30 Oe. The constant negative shift comes from the exchange biased Co layer. (d) Complete hysteresis loop up to $+3000$ Oe for the same junction. The sample was cooled down from room temperature in a magnetic field of -300 Oe. Linear background has been subtracted. The magnetization of the Co is pinned in the negative field direction, thus an external field of $+2000$ Oe is needed to reverse its magnetic moment.

Compared to the RT growth, EuS film grown at the liquid-nitrogen temperature is magnetically soft.²⁷ This helps to keep the applied H low, needed in our magnetic tunnel junction, so as not to significantly perturb the superconducting Al. To fix the magnetization of the Co layer, the samples were cooled from RT to 4.2 K in an applied field $H_{\text{appl}} = -300$ Oe, for both transport and magnetization measurements.²⁶ The Néel temperature T_N for bulk CoO is 291 K,²⁸ whereas it is lower in thin films.²⁹ Figure 1(c) shows the minor magnetic hysteresis loop of the tunnel junction in the range of -100 to $+100$ Oe measured at 2 K. Because the magnetic moment of the Co film is pinned in the negative direction, only the switching of EuS is observed, which happens at the coercive fields around ± 30 Oe. It is worth noting that the magnetic moment has a constant negative shift coming from the exchange biased Co layer. The complete hysteresis loop (major loop) is presented in Fig. 1(d), showing that an external field of $+2000$ Oe was required to switch the magnetization of the Co layer. Since the external field H was swept at best only between $+200$ Oe and -200 Oe in the magnetotunneling measurements, the magnetic moment of the Co layer stayed aligned in the negative field direction during the measurements.

We measured the resistance of the junction EuS/Al/Al₂O₃/Co/CoO as a function of the applied H field. The TMR value is defined as $\text{TMR} = [(R_{AP} - R_P)/R_P] \times 100\%$, with R_{AP} and

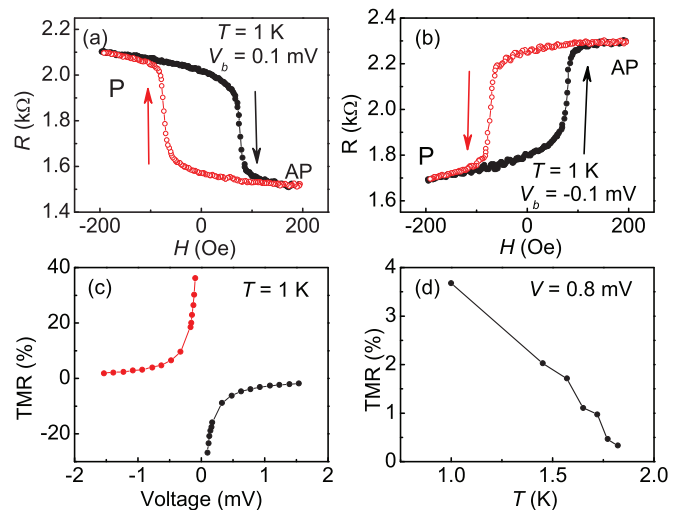


FIG. 2. (Color online) Resistance of the junction EuS/Al/Al₂O₃/Co/CoO as a function of the applied magnetic field H under the bias voltage (a) $V_b = +0.1$ mV and (b) $V_b = -0.1$ mV at 1 K. The bias voltage is defined as $(V_{Co} - V_{Al})$. (c) Bias dependence of the TMR at $T = 1$ K. The TMR values show opposite signs for positive bias and negative bias. The absolute TMR value diminishes with increasing voltage. (d) Temperature dependence of the TMR under a bias voltage of 0.8 mV. The TMR decreases as the temperature increases.

R_P corresponding to the junction resistances in the antiparallel (AP) and the parallel (P) magnetization configurations, respectively. At $T = 4.2$ K, we did not observe any TMR where the Al film was in normal state. This may be attributed to the fact that in the normal state the exchange coupling is restrained within a few Fermi wavelengths at the F/I/S interface, as shown in the theoretical model by Tokuyasu *et al.* and tunneling experiments by Tkaczyk *et al.*^{30,31} The absence of TMR rules out the possibility of EuS directly polarizing the whole Al layer. As the Al film enters a superconducting state below the transition temperature $T_C = 2.0$ K, the quasiparticle DOS becomes sharp¹ and TMR appears. The junction resistance as a function of the external H field measured at 1 K is shown in Fig. 2(a). The bias voltage is defined as $(V_{Co} - V_{Al})$. Under positive bias, when the H is swept from -200 to $+200$ Oe, the junction resistance drops at $+80$ Oe, which is the coercive field of EuS and where the magnetization configuration changes from a P to an AP state. When the H sweep is reversed to -200 Oe, the junction resistance increases at -80 Oe. Referring to the definition of TMR, we thus obtain a negative TMR. As we reverse the bias polarity, the sign of the TMR changes from negative to positive, as shown in Fig. 2(b). Since the magnetization of Co is pinned, only one resistance switching appeared for each sweep, at ± 80 Oe when EuS magnetization switches. It is worth noting that the switching field at 1 K is larger than the coercive field of the EuS film obtained from the $M(H)$ loop at 2 K, reflecting the increase of H_c as the temperature decreased, as we observed in the EuS/Al/EuS structure recently.^{27,32} The bias dependence [see Fig. 2(c)] shows that the absolute value of the TMR decreases with increasing voltage, and the TMR diminishes as the temperature increases [see Fig. 2(d)]. The exchange field at

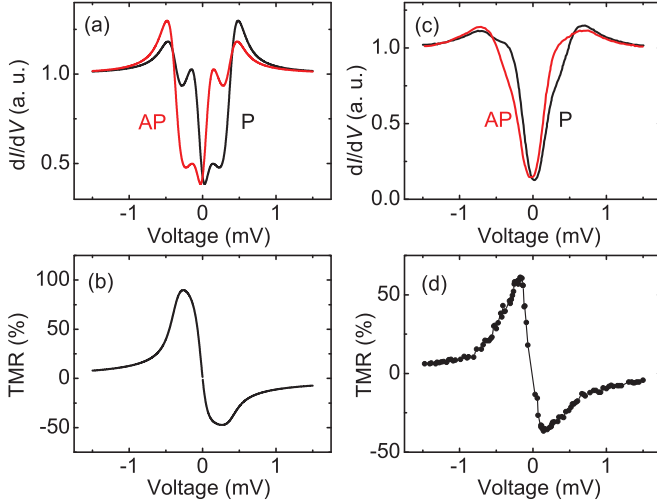


FIG. 3. (Color online) (a) Simulated tunneling conductance (dI/dV) for the MTJ EuS/Al/Al₂O₃/Co/CoO in the parallel (P, black) and antiparallel (AP, red) states. Junction resistance can be obtained by integrating the dI/dV curves. (b) Calculated bias dependence of the TMR based on the simulation, showing the TMR changing sign with bias polarity. (c) Measured tunneling conductance for the EuS/Al/Al₂O₃/Co/CoO junction in P (black) and AP (red) states at 0.4 K. (d) Calculated bias dependence of the TMR from the measured dI/dV curves.

the FI/S interface is purely interfacial, as proved by the disappearance of the TMR when a thin Al₂O₃ barrier was inserted between the EuS and the Al. Compared to the conventional magnetic tunnel junction structure of FM/I/FM, in the present experiment the combined FI/S bilayer acts as an artificial FM layer.

The internal exchange field acting on the quasiparticle DOS of superconducting Al can be determined by measuring the tunneling conductance.¹² An exchange field of 3.3 T was determined from the measurements [see Fig. 3(c)]. In the S/FI model proposed by Tokuyasu *et al.*, the relation between the Zeeman splitting field H and the exchange energy h_{ex} is expressed as³⁰

$$H = \left(\frac{h_{ex}}{\mu} \right) \left(\frac{1}{k_F d} \right) \sqrt{\frac{2E_F}{E_g}}, \quad (1)$$

where k_F is the Fermi wave vector, d is the thickness of the Al film, E_F is the Al Fermi energy, and E_g is the band gap of EuS. By substituting H with the experimental value of 3.3 T and using parameters²⁸ $k_F = 1.75 \times 10^{-8} \text{ cm}^{-1}$, $E_F = 11.63 \text{ eV}$, $E_g = 1.64 \text{ eV}$, and $d = 3.2 \text{ nm}$ (estimated Al thickness after oxidation), we infer an exchange energy $h_{ex} = 2.6 \text{ meV}$, which is of the same order of magnitude as we reported recently in a FI/S/FI sandwich structure.³² We also obtained a spin polarization $P_{Co} = 40\%$ using the Meservey-Tedrow method for the Co electrode, quite consistent with the directly measured value.^{14,16} The sign of the spin polarization of Co can depend on the choice of the barrier material. As De Teresa *et al.* reported, Co shows a negative spin polarization when SrTiO₃ or Ce_{0.69}La_{0.31}O_{1.845} barriers were present.^{33,34} For our sample configuration having the Al₂O₃ barrier, the spin polarization of Co is known to be positive.^{14,16} We fit our

conductance curve to the standard S/I/F tunneling model and obtained the Fulde's spin-orbit scattering parameter of 0.05 and orbital depairing parameter of 0.1, showing that they are negligible in this system.³⁵

In order to understand the unusual bias dependence of the observed TMR, we carried out a simulation. The conductance is calculated as the sum of contributions from spin-up and spin-down electrons as¹⁴

$$\begin{aligned} \frac{dI}{dV} \sim & \int_{-\infty}^{\infty} a N_s(E + \mu H) \frac{\beta \exp[\beta(E + eV)]}{\{1 + \exp[\beta(E + eV)]\}^2} dE \\ & + \int_{-\infty}^{\infty} (1-a) N_s(E - \mu H) \frac{\beta \exp[\beta(E + eV)]}{\{1 + \exp[\beta(E + eV)]\}^2} dE, \end{aligned} \quad (2)$$

where $\beta = 1/kT$, a is the fraction of the electrons with their magnetic moment aligned with the applied field, and $N_s(E)$ is the superconducting density of states with energy E . Figure 3(a) shows the simulated tunneling conductance curves of a EuS/Al/Al₂O₃/Co/CoO junction for the P (black) and AP (red) states. We used the following experimentally obtained parameters: superconducting Al energy gap $\Delta = 0.3 \text{ meV}$, $P_{Co} = 40\%$, and internal exchange field $H = 3.3 \text{ T}$. Clear Zeeman splitting of the quasiparticle DOS can be seen in the simulated conductance, as well as their asymmetric peaks. The height of the peaks reflect the different tunneling probability between up and down spin states of the Al quasiparticle DOS and the asymmetric spin subbands at the Fermi level of the Co electrode. Reversing the magnetization direction of the EuS results in a switch from the P to AP state, leading to an inversion of the conductance curve.

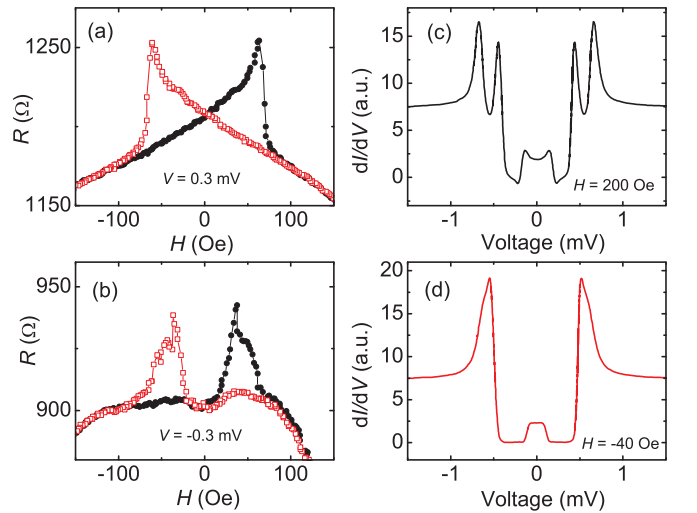


FIG. 4. (Color online) Resistance of the junction EuS(1.5)/Al(4)/Al₂O₃/Al(3.5) as a function of the applied magnetic field under the bias voltages (a) $V_b = +0.3 \text{ mV}$ and (b) $V_b = -0.3 \text{ mV}$ at 1 K. The bias voltage is defined as ($V_{\text{top}} - V_{\text{bottom}}$). (c) Tunneling conductance (dI/dV) measurements show the Zeeman splitting of the DOS of Al in the presence of the EuS exchange field with an external field $H = 200 \text{ Oe}$. (d) The Zeeman splitting of the Al DOS disappears at the coercive field of EuS ($H = -40 \text{ Oe}$), where the EuS exchange field is weakened.

The junction resistances can be calculated by integrating the dI/dV curves, which can then give us the theoretical TMR value as shown in Fig. 3(b). For example, starting from zero bias under positive voltage, the area below the dI/dV curve for the AP state is larger than that for the P state, indicating the integrated conductance (resistance) for the AP state would be larger (smaller) than that for the P state, which results in a negative TMR. The calculation shows that the TMR changes its sign as the voltage polarity is reversed, as in our experimental observations. Similarly, estimating TMR(V) dependence from the measured dI/dV curves, we get the same bias dependence [see Fig. 3(d)]. The experimental bias dependence [Fig. 2(c)] follows the calculation reasonably well. This confirms that the TMR results from the spin sensitive conductance variation due to spin-split quasiparticle DOS.

Above we have shown that a TMR obtained from a S/I/FM junction is due to the spin sensitive conductance variation. A similar mechanism can also lead to a TMR in a S/I/S junction and EuS(1.5)/Al(4)/Al₂O₃/Al(3.5) forms such a structure. As shown in Figs. 4(a) and 4(b), we obtain a positive TMR of 6% at 1 K when the top and bottom Al electrodes were superconducting. The TMR does not change sign with the voltage polarity reversed, where the voltage is defined as ($V_{\text{top}} - V_{\text{bottom}}$). Similar to the previous case, we measured the tunneling conductance as a function of the bias voltage.

When the magnetization of the EuS is aligned in the presence of a small external field $H = 200$ Oe, we can clearly observe the splitting of the quasiparticle DOS [see Fig. 4(c)], with an exchange field as large as 1.9 T. In the vicinity of the EuS coercive field ($H = -40$ Oe) where the exchange field nearly cancels out, we obtained unsplit quasiparticle DOS [see Fig. 4(d)]. After integration, the conductance variation gives rise to different junction resistances, leading to the observed TMR. Because of the symmetry of the dI/dV curves, the sign of the TMR is independent of the voltage polarity.

In summary, we utilize the exchange interaction at the F/I/S interface to tune the spin DOS in a superconductor, demonstrating a magnetization controlled TMR coming from the spin-split superconductor quasiparticle DOS tunneling, with an observed value as large as 36%. Our results could potentially offer a new direction to the development of cryogenic superconducting spintronic devices.

We thank Badih A. Assaf for the help with SQUID measurements. We also thank Igor Zutic for helpful discussions. This work was supported by NSF Grant No. DMR-1207469 and ONR Grant No. N00014-13-1-0301, and the Center for Excitronics at MIT, an Energy Frontier Research Center funded by US Department of Energy, Office of Science and Office of Basic Energy Sciences, under Award No. DE-SC0001088.

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