Variation of the Electron-Spin Polarization in EuSe Tunnel Junctions from Zero to Near 100% in a Magnetic Field

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A thin film of EuSe was used as a tunnel barrier between the normal metal Ag and superconducting Al. Tunneling characteristics at 0.45 K showed the following: (1) the absence of exchange splitting of the EuSe conduction band in zero magnetic field, where EuSe is antiferromagnetic; (2) field-dependent spin polarization of the tunneling electrons as high as 97% at $H \geq 1.2$ T; (3) enhanced Zeeman splitting of the Al quasiparticle states because of the exchange interaction at the Al-EuSe interface; (4) a decrease in tunnel resistance by as much as 75% in a magnetic field ≥ 1.7 T. The results show that EuSe barrier junctions provide field-tunable resistors and sources of low-energy polarized electrons.

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Among the semiconducting europium chalcogenides, EuS and EuO are ferromagnetic and EuTe is antiferromagnetic [1]. Studies on bulk single crystals of EuSe show a complex magnetic phase diagram; it is an intermediate case between ferromagnetic and antiferromagnetic compounds [2]. In very low magnetic fields, EuSe orders as one antiferromagnetic phase (AF I) at 4.6 K, then as a ferrimagnetic phase at 2.8 K, and finally as a second antiferromagnetic phase (AF II) at 1.8 K. In the AF I phase of EuSe, a small redshift (≈ 0.07 eV) in the optical absorption edge has been reported [3]. The application of a magnetic field changes EuSe into a ferromagnetic phase, leading to a large redshift due to exchange splitting which lowers the bottom of the conduction band. Earlier spin-polarized tunneling studies with ferromagnetic EuS films as tunnel barriers showed that the tunneling electrons can be 90% spin polarized [4]. This phenomenon has been attributed to the spin-filter effect in the EuS barrier caused by the exchange splitting of the EuS conduction band. This exchange splitting ($2\Delta = 0.36$ eV) in EuS lowers the tunnel barrier energy for spin-up electrons and raises it for spin-down electrons, giving rise to the preferential selection of up-spin electrons in the tunneling process. In EuS, this phenomenon is seen even at $H=0$ and the spin-polarization value is independent of the applied magnetic field.

In the present work, thin-film tunnel junctions of Ag/EuSe/Al were prepared on glass substrates by vacuum evaporation of the materials. Metal masks were used to obtain the cross-pattern tunnel junction geometry. Silver cross strip films 40 nm thick were first deposited on room-temperature substrates. These were covered with EuSe films 2.5 to 5.0 nm thick by evaporating bulk EuSe using an electron gun. The junction was completed by depositing long strips of 4.2 nm thick Al film over the above structure while the substrate was cooled to 80 K. After warming to room temperature, the substrate was removed from the vacuum system and mounted on a ^{3}He probe for measurements at low temperature and with a magnetic field applied parallel to the junction surface.

The tunnel junction resistance (R_j) was measured as a function of temperature during cooling from 300 to 0.5 K, with and without applied magnetic field. $R_j(T,H)$ data were taken by the four-terminal method with a measuring current of 10 μA, corresponding to a voltage far above the Al superconducting energy gap at low temperature. Semiconducting behavior was observed, with R_j increasing by a factor of about 4 when cooled down from 300 K to 40–50 K and remained constant in zero or small magnetic fields down to about 10 K. Figure 1 shows R_j for a junction below 24 K. For $H=0$ and 124 Oe there was a gradual decrease of about 1% in R_j below 10 K, giving a shallow minimum at 5 K and then increasing about 4% upon cooling to 0.5 K. However, in a field of 0.2 T, R_j began its decrease at 16–18 K, with a minimum occurring at 3.3 K, below which it increased by about 2.5%.

By fitting to the Simmons' tunneling theory [5], the slight decrease in R_j at $H=0$ is consistent with a decrease of the bottom of the conduction band (and thus the tunnel barrier height) by about 5×10^{-4} eV; likewise

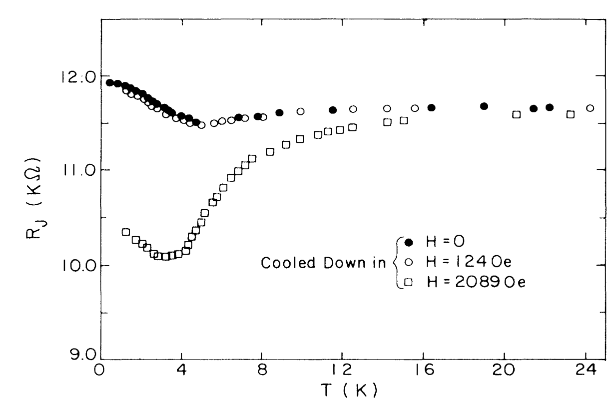

FIG. 1. Resistance variation of a tunnel junction as a function of temperature upon cooling through the magnetic ordering temperature of EuSe in zero field, in 128 Oe and 0.2 T.

the increase in R_j in $H=0$ below about 4.5 K can be interpreted as an increase in the tunnel barrier of about 0.002 eV. This increase in R_j would correspond to a small blueshift in the optical absorption edge. A blueshift has been observed in EuSe and EuTe at the antiferromagnetic ordering temperature, but of much greater magnitude [1]. The much larger dip in resistance with magnetic field is caused by the exchange splitting of the conduction band of EuSe in the ferromagnetic state. At a temperature of 0.5 K the ratio of the resistance at $H=0$ to that at 1.5 T was 2.1 in the junction shown in Fig. 1. An analogous effect was observed by Esaki, Stiles, and von Molnar [6] for the internal field emission through a EuSe barrier at much higher voltages.

To confirm the intrinsic magnetic behavior of EuSe films used as tunnel barriers, magnetization measurements as a function of temperature were carried out on EuSe films 5 and 10 nm thick deposited on sapphire. The magnetic moment versus H and T was measured in a SQUID magnetometer. Susceptibility versus temperature data showed good agreement with bulk behavior.

Tunnel conductance measurements were made at 0.45 K for the junctions as a function of applied magnetic field and are shown in Fig. 2 for one of the junctions. At this temperature the Al film is superconducting, as the film has a T_c of 2.5 K. Referring to Fig. 2, peaks in the conductance curves at $H=0$ show the superconducting energy gap in the Al film. At zero bias the conductance is almost zero, showing negligible leakage current for the junction and a constant background conductance at higher bias, indicating a fairly high barrier. Upon applying a magnetic field, the conductance peaks are each split into two peaks symmetrically spaced on either side of the $H=0$ peaks. This effect is the Zeeman splitting $2\mu H$ of the Al quasiparticle density of states, where μ is the magnetic moment of the electron. As H increases, the splitting increases. However, the observed splitting is much larger than that which is expected from the applied field alone. Similar enhanced splitting has been observed by Tedrow, Tkaczyk, and Kumar [7] and by Tkaczyk and Tedrow [8] in various rare-earth oxide layers and by us in EuS/Al layers [4]. The excess splitting is due to the exchange field of the aligned Eu^{2+} ions acting on the superconductor in an applied field. This interface interaction is extremely sensitive to the interfacial coupling between the Al and the EuSe as earlier seen with EuS [5]. In some cases this extra effective internal field was 4 T, whereas in others the maximum was only 1 T.

In addition, conductance curves are not only Zeeman split, but also are asymmetric in height about $V=0$. The asymmetry is present even at $H=0.104$ T, and increases as H increases. This asymmetry indicates the degree of spin polarization P of tunneling electrons—the larger the asymmetry, the higher P. At $H=0$, the conductance curve is symmetric, indicating no polarization of tunneling electrons or no Zeeman splitting in the Al. An experiment to be described below shows that at $H=0$, P is indeed zero. The H dependence of P is plotted in Fig. 3 for this junction. At low fields, P rapidly increases as H increases, reaching a maximum saturation value of $P=84\%$ at about 0.8 T. The values of P quoted in this Letter have been corrected for spin-orbit scattering in the Al film. This behavior is also reflected in the tunnel conductance values from the curves in Fig. 2 slightly beyond the superconducting gap region where dI/dV is approximately the inverse of the junction resistance R_j. When R_j is plotted as a function of H (Fig. 3), it rapidly decreases at low fields and approaches a constant value about 1 T. The ratio of the resistance at $H=0$ to that at 1.2 T was found to be $R_R=1.7$. Assuming a value of $\Phi_0=1.1$ eV and the exchange splitting in the EuSe film to be $2\Delta=0.26$ eV [3], we calculate from the spin-filter model [4] and Simmons' tunneling theory [5] a tunnel barrier thickness $S=1.9$ nm, $R_R=1.8$, and $P=84\%$ at 1.24 T. The value assumed for 2Δ is the optically measured exchange splitting in bulk EuSe crystals in fields of

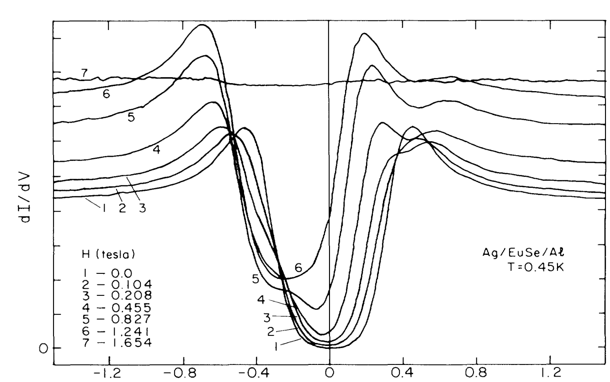

FIG. 2. Tunnel conductance plotted as a function of junction voltage bias in various applied magnetic fields, showing the effects of Zeeman splitting and electron-spin polarization.

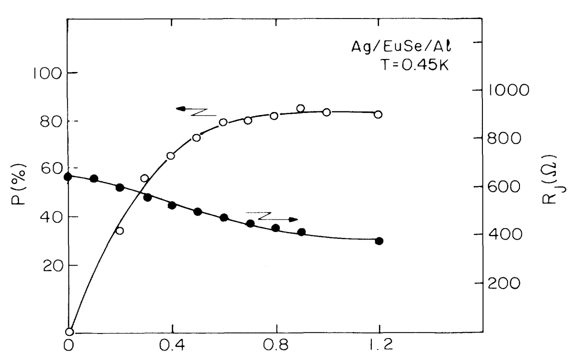

FIG. 3. Calculated electron-spin polarization as a function of the applied magnetic field, from the tunnel junction conductance data in Fig. 2. Also shown is the junction resistance (R_j) variation in the applied magnetic field.

about 1 T. The value taken for $\Phi_0 = 1.1$ eV is considerably less than the average barrier height of 1.47 eV expected from the work functions of Al and Ag, 4.28 and 4.26 eV, respectively, and the electron affinity of EuSe of 2.8 eV. However, this reduction of barrier height is typical of disordered films of semiconductors and insulators formed by vacuum deposition. The calculated values of P and R_R agree well with the measured values.

Some junctions showed even more extreme behavior. For one such junction, Fig. 4 shows a plot of tunnel conductance versus bias voltage for various applied magnetic fields. With increasing H, R_j decreased so significantly that by 1.8 T it had dropped to 25% of its $H=0$ value. In a magnetic field, the conductance peaks both shift towards lower voltage. This shift is due to Zeeman splitting of the conductance peak in the magnetic field, except that only the up-spin peak is present. The results clearly show a much higher value of spin polarization of the tunneling electrons than found in Fig. 2. The value obtained by fitting the Maki-Fulde [9,10] theory is $P=(97\pm 3)\%$. This is the first time that this single peak motion implying very close to 100% polarization has been observed in spin-polarized tunneling. The junctions which show the single-peak feature have a thicker EuSe barrier (higher R_j) than, for example, those junctions that showed both peaks in a field. For example, for the junction of Fig. 2 the calculated barrier thickness is $S=1.9$ nm, whereas for that of Fig. 4, $S=2.4$ nm; this difference is significant because an increase in S by 0.2 nm increases the resistance by a factor of 10. In addition, single-peak junctions also show a weaker exchange interaction with the Al films; the effective exchange field is only 1.1 T even at an applied field of 1.66 T as compared with 4.0 T for a EuSe/Al/Al_2O_3/Ag junction at $H=0.62$ T. By fitting the measured tunnel currents above the superconducting energy gap of the junction shown in Fig. 4 to the spin-filter model and Simmons' tunneling theory, values of the barrier thickness $S=2.42$ nm, $\Phi_0=0.90$ eV, and $2\Delta=0.26$ eV are consistent with the data at $H=0$ and $H=1.2$ T.

The results can be understood in a qualitative way as follows. In zero field at $T=0.45$ K, EuSe being an antiferromagnet, there is no net internal field acting on the Al film. This result is supported by the absence of Zeeman splitting at $H=0$ even after subjecting the junction to a high field; the result also follows from an experiment to be described below. As H increases to 0.1 T and beyond, enhanced Zeeman splitting is seen. This is because, in a field, EuSe starts developing a net magnetic moment along the field direction and eventually becomes fully spin aligned beyond about 1.2 T, showed by the saturation in the enhanced Zeeman splitting. Thus, in an applied field the aligned Eu^{2+} ions act on the Al through an exchange field. The net internal exchange field as deduced from the Zeeman splitting is very sensitive to the condition of the Al/EuSe interface, where a monolayer of an oxide can decrease the exchange coupling dramatically [11]. In addition, the maximum magnetization of the EuSe film is presumably dependent on its disorder.

Both the appearance of spin polarization and the decrease in junction resistance in a magnetic field have a common origin. At $H=0$ the tunnel barrier is schematically shown in Fig. 5. Upon applying a magnetic field, the exchange splitting of the conduction band in EuSe increases with H. This exchange splitting into spin-up and spin-down levels results in the decreasing and increasing of the barrier height for up- and down-spin electrons, respectively, as compared to the $H=0$ case which is shown schematically in Fig. 5. Such a tunnel barrier allows spin-up electrons to tunnel preferentially, thereby creating polarization of the tunneling current. The probability of tunneling for spin-down electrons decreases and that

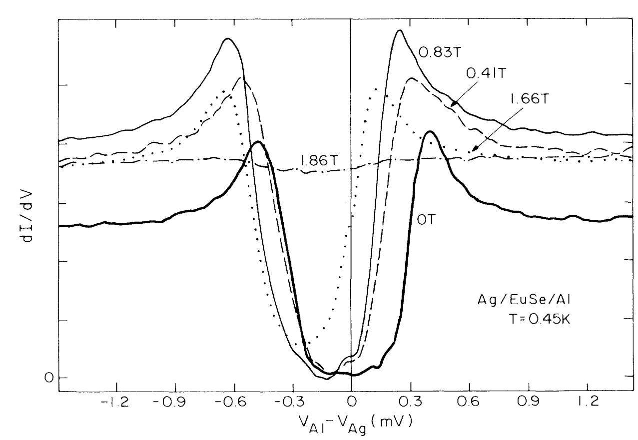

FIG. 4. Tunnel conductance vs junction voltage bias in various applied fields for a high resistance tunnel junction, showing only up-spin peaks indicating nearly 100% electron-spin polarization.

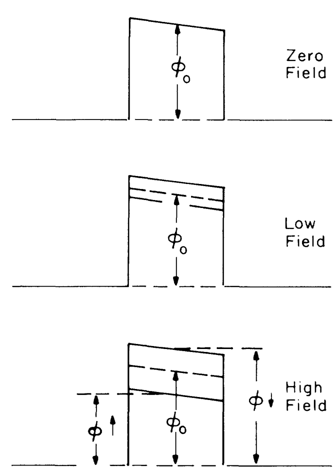

FIG. 5. Schematic of the spin-filter model. Tunnel barrier changing in an applied magnetic field due to the emergence of exchange splitting in EuSe in an applied field.

for spin-up electrons increases as the exchange splitting of the EuSe conduction band increases with H. Beyond an applied field of about 1.2 T the exchange splitting saturates and so the spin polarization reaches a maximum value. In some cases, as seen in Fig. 4, near 100% polarization is reached.

A natural question that arises is whether at $H=0$ or at very low fields spin polarization is absent or, alternatively, there is simply no Zeeman splitting in the Al at $H=0$. To observe polarization, the Al quasiparticle density of states must be Zeeman split and in zero field this splitting is presumably zero for antiferromagnetic EuSe, unlike ferromagnetic EuS. To look for possible exchange splitting in EuSe at $H=0$, EuS was used to provide the internal field for the Al film by making EuS/Al/EuSe/Ag junctions. Here, EuSe is the tunnel barrier between the metal electrodes Al and Ag as before, whereas EuS is in contact with Al on one side to provide the exchange field necessary to create Zeeman splitting in the Al at $H=0$. A polarization less than 1% can be detected in the conductance curves. This value would correspond to an exchange splitting of less than a few meV in the EuSe barrier. At $H=0$ the EuS/Al/EuSe/Ag junctions yielded highly symmetric tunneling curves, indicating that there was no measurable spin polarization caused by the EuSe barrier at $H=0$. However, in an applied field of only 100 Oe, spin polarization of about 25% was observed, increasing to higher values as H increased.

In summary, the present research shows that there is no exchange splitting of the conduction band in EuSe at $H=0$. The exchange splitting is observed only in fields large enough to enter the ferrimagnetic or ferromagnetic phase regions. This exchange splitting of a tunnel barrier leads to a spin-filter effect in which application of a magnetic field varies the polarization of tunneling electrons from 0 to close to 100%. The Zeeman splitting enhancement of Al films in contact with EuSe was found to correspond to an additional effective field as high as 4 T. Finally the tunnel junction resistance is a strong function of magnetic field and can decrease as much as a factor of 4 from $H=0$ to 1.7 T. The magnetoresistance of EuSe tunnel junctions is one of the largest seen to date and is comparable with the giant magnetoresistance recently seen in multilayers of ferromagnetic materials.

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