Electron-Spin Polarization in Tunnel Junctions in Zero Applied Field with Ferromagnetic EuS Barriers

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An electron-spin polarization P of as much as 80% has been observed in the tunnel current in Au/EuS/Al tunnel junctions. P can be explained by the different heights of the tunnel barriers for the two spin directions. The Zeeman splitting of the Al quasiparticle density of states is greatly enhanced by the exchange interaction at the EuS/Al interface. Spin polarization was even seen in zero applied field. The value of P calculated from the tunneling theory using known barrier heights in EuS is consistent with the measured values.

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The discovery of Zeeman splitting of the quasiparticle density of states[1] of superconducting Al immediately led to the ability to determine the electron-spin polarization P of tunnel currents. With use of this technique, the value of P for electrons tunneling in Al/Al_2O_3 ferromagnetic-metal junctions was extensively studied in Ni, Co, Fe, and $3d$ alloys as well as in some rare-earth metals.[2-4] In these experiments the spin polarization was attributed to the difference in the spin densities of states of the itinerant electrons in the ferromagnets at the Fermi energy.[5] In contrast to these earlier results, the present experiments show for the first time electron-spin polarization of the tunneling current between nonferromagnetic electrodes. This effect can be explained by the different barrier heights for the two spin directions in the ferromagnetic insulator separating the metals. The barrier thus acts as a spin filter.

The Eu chalcogenides have been extensively investigated.[6,7] Several studies of EuS are closely related to the present observations. Esaki, Stiles, and von Molnar[8] reported an internal-field-emission study of junctions having magnetic semiconductors EuS and EuSe as barriers 20 to 60 nm thick. They observed an increase of field-emission current as the temperature was lowered to below the magnetic ordering temperature of the barrier and interpreted it as caused by the decrease of barrier height when spin ordering takes place. Similar results were obtained by Thompson *et al.*[9] with Schottky barriers made on n-type doped semiconducting single-crystal EuS. Field-emission studies[10-12] on EuS-coated tungsten tips showed a high degree of polarization of the field-emitted electrons below the Curie temperature of EuS, $T_C \approx 16.7$ K.[6] These results were explained by the spin-filter effect in EuS below T_C.

In the present study, tunnel junctions were prepared in a conventional way by vacuum deposition on glass slides. Different types of junctions were made and in every case one of the metal electrodes was a 4- to 4.4-nm-thick Al film deposited on a liquid-nitrogen–cooled substrate. The other electrode was a film of Au, Al, or Fe. The tunnel barrier was an EuS film formed by evaporation with use of an electron gun on a pressed pellet of EuS. The average thickness of the EuS barriers used in this work was about 2.5 nm as determined by a rotating sector disk and a quartz-crystal thickness monitor. The best tunneling results were obtained with junction resistances of 1 to 20 kΩ for junction areas $\approx 4\times10^{-4}$ cm^2. The junctions fabricated were Au/EuS/Al, Al/EuS/Al, Al/EuS/Fe, and Fe/EuS/Al, where in each case materials are listed in the order in which they are deposited. Although usually all three materials were deposited on liquid-nitrogen–cooled substrates, in some cases the Au and EuS films were deposited at room temperature. Even though the yield of good tunnel junctions was greater on cold substrates, higher polarizations were found for the higher-temperature depositions. X-ray diffraction of 100-nm EuS control films deposited at 80, 300, and 400 K all indicated the films to be polycrystalline and the line positions agreed with the diffraction pattern taken on an EuS powder sample. Selected junctions were cooled in a ^{3}He refrigerator equipped with a superconducting magnet, and conductance, dI/dV vs V, was measured at 0.4 K as a function of the magnetic field H applied parallel to the film surface. Current-voltage curves with bias up to 1 V were also made at various temperatures, from 1.1 to 20 K. We present here mainly the results from the Au/ EuS/Al junctions; the other types of junctions showed qualitatively similar behavior. Two sets of Au/EuS/ Al junctions were carefully studied. We refer to them as set 1 and set 2. All three materials in junctions of set 1 were deposited onto substrates at $T \approx 80$ K, while in junctions of set 2 the Au and EuS films were deposited onto substrates at $T \approx 300$ K.

Figure 1 shows measurements at 0.4 K of the differential conductance dI/dV versus voltage V of a Au/EuS/ Al junction of set 1. The Al film was superconducting with a transition temperature of 2.33 K. The curve labeled 0 was made before any magnetic field was applied, and the superconducting energy gap of Al, 2Δ, is clearly

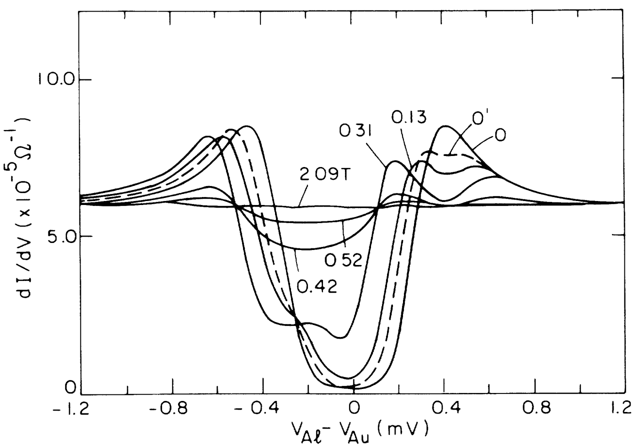

FIG. 1. Conductance vs voltage for a Au/EuS/Al junction from set 1 (deposited at 80 K) at $T = 0.4$ K for various values of applied magnetic field H indicated in teslas. A fit of the theory to the curves gives $P = 55\% \pm 5\%$. The curve $H = 0$ was made before a field was applied. The dashed curve $H = 0'$ was made after having applied a field of 2.09 T and shows Zeeman splitting and polarization on returning to zero field.

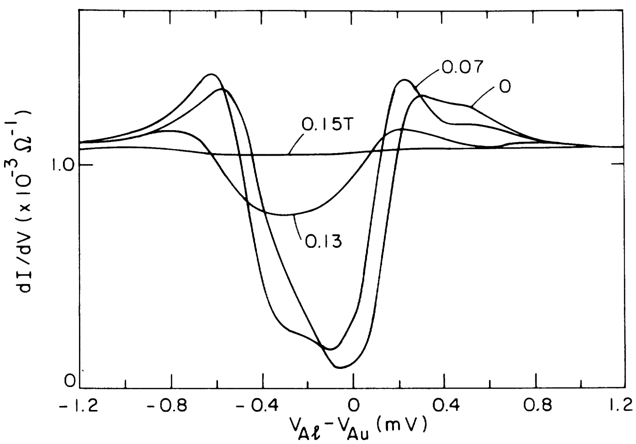

FIG. 2. Conductance vs voltage for a Au/EuS/Al junction from set 2 (Au and EuS deposited at 300 K) at $T = 0.4$ K for various values of H. A fit of theory to the curves gives $P = 80\% \pm 5\%$. Curves were all taken in increasing field. Hysteresis was observed in decreasing H, but is not shown.

seen. dI/dV at $V = 0$ was 1.4% of the normal-state conductance, showing that the conduction process is almost entirely tunneling. As we applied a magnetic field H parallel to the plane of the junction, the conductance peaks were each split by the Zeeman energy because of the magnetic moment of the electron μ. At a value of $H \approx 1.5$ T the paramagnetic limit is reached and the Al film becomes normal. For a tunnel junction with a thin Al electrode, a nonmagnetic barrier such as Al_2O_3, and a normal-metal counterelectrode, the Zeeman splitting in the superconducting quasiparticle density of states is equal to $2\mu H$. However, the splittings shown in Fig. 1 are much greater than those corresponding to the applied field. This is similar to, although more extreme than, the enhanced Zeeman splittings found by Tedrow, Tkaczyk, and Kumar[13] when Al films are in contact with various rare-earth oxides. In this situation the conduction electrons of the thin Al film are subjected to an effective internal field B caused by exchange scattering with the rare-earth ions in the insulator. The critical field of the Al was reached when $H \approx 1.5$ T which corresponds to a value of $B \approx 5$ T, the paramagnetic critical field H_{cp} for Al films of this thickness.[14] When H was reduced to zero (curve labeled 0′ in Fig. 1) the Zeeman splitting persisted, corresponding to an effective internal field $B \approx 1.6$ T. This effect was seen in all the EuS junctions studied, but had never been observed previously with spin-polarized tunneling measurements. This hysteresis perhaps implies a remanent magnetization of the EuS as will be discussed below. A hysteresis in the resistivity of highly doped EuS reported by Shapiro and Reed[15] may be closely related. A striking feature of the data of Fig. 1 is the pronounced asymmetry which implies a large value of the electron-spin polarization of the tunneling current. A simple analysis neglecting spin-orbit scattering in the Al film gives a value of $P = 74\%$. To obtain accurate values of P we used the complete theory[16,17] to fit the curves; the best fit gave values of $P = 55\% \pm 5\%$ and the spin-orbit scattering parameter $b = \hbar/3\tau_{s.o.}\Delta_0 = 0.05$.[17,18]

Figure 2 shows a characteristic junction from set 2 (Au and EuS deposited at 300 K). Zeeman splitting of the quasiparticle density of states in the superconducting Al film and polarization of the tunnel current were present even before any magnetic field (other than the ambient field ≈ 1 Oe) had been applied. In this case, in an applied magnetic field as small as 0.15 T, H_{cp} was already reached. The polarization obtained from a fit to the curves in Fig. 2 is $P = 80\% \pm 5\%$. This is the largest value of P obtained to date for a tunnel junction. The uncertainty in P comes from the fact that the effective internal field B is only known from the Zeeman splitting which is also affected by spin-orbit scattering, unlike previous experiments in which the internal field B is essentially equal to the applied field H. This situation is made worse in the present situation by the small range of B available before the H_{cp} of the Al film is reached. Other types of junctions gave qualitatively similar results. For Al/EuS/Al, $P \approx 20\%$, for 80-K deposited Al/EuS/Fe, $P \approx 40\%$, and $P \approx 65\%$ for Fe/EuS/Al with Fe and EuS deposited at elevated temperature. For the Fe electrode the degree to which the Fe determined P is not yet known and is subject to further study.

The increase in P for the Au and EuS deposited at 300 K presumably results from the greater crystallite and domain size of the EuS when it is formed on Au of large crystalline size. A 30-nm-thick Au film deposited on SiO_2 at room temperature has grains as large as 600 nm in size.[19] Conversely, Au deposited at low temperatures has a smaller grain size, and Al and Fe are known to have an even smaller average crystallite size than Au when deposited at 80 K; these films probably introduce more disorder into the EuS film. For $T \ll T_C$ of EuS the spin-filter effect polarizes the tunnel current through each EuS domain even at $H=0$, although the direction of the magnetization **M** may be different in each domain. If the characteristic domain size is L and the superconducting coherence distance is ξ, then for $L \ll \xi$ contributions of the exchange field to the effective field in the Al film from differently oriented domains will tend to cancel over an Al film area $\approx \xi^2$, leading to zero Zeeman splitting, and consequently polarization cannot be detected. On the other hand, for $L \gtrsim \xi$ the B field in each area ξ^2 of the Al film has a uniform direction and will lead to a large Zeeman splitting even when the direction of **M** varies between domains; in this case, polarization will be seen at $H=0$. The hysteresis effect observed could be caused by remanent orientation of the domains. However, the present experiments cannot rule out a model in which the size of the domains is increased, even if their orientation is disordered when H returns to zero.

Independent of a fit to the asymmetry of the superconducting tunnel conductance, we can estimate the expected value of P from known properties of EuS and from I vs V for voltages above the superconducting gap. For this analysis we assume for Au/EuS/Al junctions a barrier of the form shown in Fig. 3. Above the bulk EuS Curie temperature $T_C = 16.7$ K,[6] the barrier height

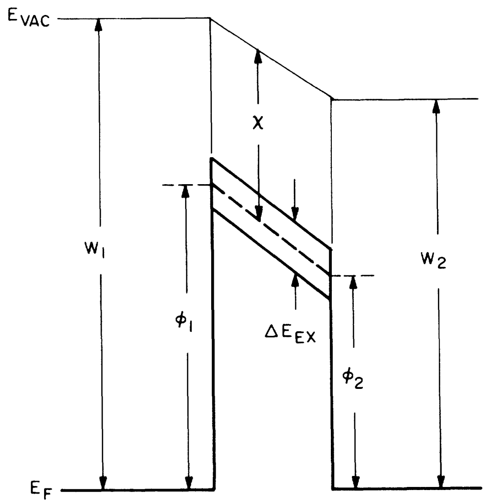

FIG. 3. Schematic representation of the tunnel barrier of a Au/EuS/Al junction. W_1 and W_2 are the work functions of Au and Al, respectively. χ is the electron affinity of EuS. The barrier heights at the Au and Al interfaces are shown as ϕ_1 and ϕ_2 at the bottom of the EuS conduction band (dashed line) at $T > 16.7$ K. The bottom of the two bands shown at $T \ll T_C$ by the solid lines separated by ΔE_{ex} are the barriers seen by the two spin directions.

(shown by the dashed line) is determined by the position of the bottom of the EuS conduction band with respect to the Fermi level of the metals. Using the values for the work functions $W_{Al} = 4.1$ eV and $W_{Au} = 5.0$ eV,[20] and the electron affinity for EuS,[21] $\chi_{EuS} = 2.5$ eV, we infer values of the barrier heights at the interfaces of $\phi_1 = 2.5$ eV and $\phi_2 = 1.6$ eV. The average barrier height is thus $\phi = (\phi_1 + \phi_2)/2 = 2.05$ eV. The tunnel current for such a barrier according to Simmons[22] is given approximately by

$$J = J_0(\phi - \tfrac{1}{2}eV)\exp[-A(\phi - \tfrac{1}{2}eV)^{1/2}] - J_0(\phi + \tfrac{1}{2}eV)\exp[-A(\phi + \tfrac{1}{2}eV)^{1/2}], \quad (1)$$

where $J_0 = (e/2\pi h)S^{-2}$ and $A = (4\pi S/h)(2m)^{1/2}$, with S being the thickness of the barrier and m the electron effective mass in the conduction band. At 20 K, which is above T_C, we can calculate values of ϕ and S which fit the measured values of $J(V)$. For two junctions of set 1 we obtained the values $S = 1.76$ nm, $\phi = 2.15$ eV and $S = 1.89$ nm, $\phi = 2.015$ eV. These values for ϕ differ by less than 5% from that obtained from the work function. The values of S are reasonable for the effective tunneling thickness in a junction of average thickness $= 2.5$ nm and are close to what one expects by comparison to Al_2O_3 barriers. The difference in m from the free-electron mass is small and is assumed to be absorbed in an effective value of S. Below T_C, the conduction band of EuS is split by the ferromagnetic exchange interaction and the barrier is split in height for the two spin directions as shown by the continuous lines in Fig. 3. With use of the bulk value of the exchange splitting in EuS, $\Delta E_{ex} = 0.36$ eV,[6] the calculated average barrier heights for the above junctions are $\phi\downarrow = 2.33$ eV, $\phi\uparrow = 1.97$ eV for $S = 1.76$ nm and $\phi\downarrow = 2.20$ eV, $\phi\uparrow = 1.84$ eV for $S = 1.89$ nm. Using Eq. (1) for each spin direction to calculate $J\uparrow$ and $J\downarrow$ we find that $J = J\uparrow + J\downarrow$ agrees with the current measured at 1.1 K from 2×10^{-3} to 0.4 V within about 10% in these two junctions. Values obtained for the polarization of the tunnel current $P = (J\uparrow - J\downarrow)/(J\uparrow + J\downarrow)$ were 79% and 83%, respectively, with an uncertainty of 5%. These are maximum values for these junction parameters and assume that there is no spin scattering or other degrading effects. Since this measurement only relies on the normal-state properties of the Al electrode it is unnecessary for L to be larger than ξ to obtain the full spin-filter effect at $H=0$. The agreement of this calculation with the value of $80\% \pm 5\%$ for the set 2 junctions is strong evidence for the correctness of the spin-filter model. Also, the temperature dependence of tunnel junction resistance is consistent with that expected from the temperature dependence of exchange splitting in the bulk EuS crystal.[23]

Very recently tunneling in $EuS/Al/Al_2O_3/Ag$ junctions has been investigated[24] in our laboratory: Enhanced Zeeman splitting was observed in Al but no polarization was detected. This shows that the polarization observed with the EuS barrier is due to spin filtering; the exchange-induced splitting in Al is a separate consequence of the ferromagnetic ordering of the EuS.

The high value of polarization obtained with EuS should be useful as a source of spin-polarized electrons in tunneling. Even higher values of P can probably be obtained with other ferromagnetic insulators or semiconductors. The method also provides a way of measuring the exchange splitting for thin films of such substances. The ability to do spin-polarized tunneling studies in zero applied field will allow many new types of measurements of magnetic and superconducting materials.

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