Spin-dependent band structure effects and measurement of the spin polarization in the candidate half-metal CoS_2

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CoS_2 has recently been predicted to be highly spin-polarized, but very little is known about its magnetotransport properties. We find that this system exhibits a large anomaly in the resistivity at the Curie temperature (T_C), which shows a pronounced shift with applied field, resulting in negative magnetoresistance below T_C and positive magnetoresistance above it. Magnetization measurements indicate an almost first-order character to the ferromagnetic transition, indicating that CoS_2 is close to a tricritical point. Analyzing the field dependence of the magnetization and resistivity, we are able to prove that the resistivity near T_C is controlled only by the magnetization, regardless of whether the ferromagnetic state is induced by cooling or by application of a field. We propose that the increase in resistivity on entering the ferromagnetic state is due to a spin-dependent band structure effect where exchange splitting results in a distinct decrease in the density of states at the Fermi level for the minority spins. Point-contact Andréev reflection measurements on sulfur-stoichiometric polycrystals give a spin polarization of 56%, consistent with indications of electron-magnon scattering in our transport measurements as well as our proposed spin-dependent band structure mechanism for the magnetotransport anomalies.

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The essence of the current focus area termed spin-electronics or "spintronics," [1] is to utilize the electron's spin, as well as its charge, in creating new devices or enhancing the functionality of current ones. Many spin-electronic devices, such as giant magnetoresistance multilayers,[2] spin valves,[3] magnetic tunnel junctions,[4] efficient spin injection ferromagnet/semiconductor bilayers,[5] and magnetic random access memory,[6] could benefit from the use of ferromagnets with high-spin polarization. In certain devices (e.g., "Ohmic" spin injectors) it has even been suggested that this is a necessity.[7] It is therefore of great importance that we realize ferromagnets with 100% spin polarization (P) at the Fermi level (E_F), which are referred to as "half-metallic" ferromagnets. Such ferromagnets have been predicted from electronic structure calculations for a number of years,[8] but only one material, CrO_2, has been unambiguously determined to be half-metallic, both from Andréev reflection[9–11] and tunneling.[11] Some of the key materials issues include the difficulty of preparing CrO_2 by conventional vacuum deposition,[12,13] the anomalously low tunneling magnetoresistance[14] (TMR), and the general issues regarding the spin polarization of the surface compared to that of the bulk. Experiments on other predicted half-metals have also raised serious issues. $La_{1-x}Sr_xMnO_3$ was found to be half-metallic only at low temperatures,[15] while Heusler alloys such as NiMnSb (Ref. 9) and Co_2MnSi (Ref. 16) have disappointing polarizations of 50%–60%, which was interpreted in terms of a reduction in polarization due to antisite disorder.[16,17]

In this paper we present novel magnetotransport effects in the pyrite structure sulfide CoS_2, which has been recently predicted to be either highly,[18,19] or completely,[20] spin polarized. Despite the low Curie temperature ($T_C \sim 120$ K), which rules out applications, this system offers some key advantages for fundamental studies of half-metallic ferromagnetism. First, by avoiding the use of alloys such as Heuslers or the double perovskite Sr_2FeMoO_6,[21] we avoid the problem of reduced polarization by antisite disorder.[16,17,21] Second, it has been suggested that CoS_2/semiconductor interfaces can generate fully polarized currents even in the absence of full polarization in CoS_2.[22] (On this note it is also worth mentioning that CoS_2 is lattice matched to Si.) Finally and most importantly, the simple band structure of CoS_2 (where ferromagnetism occurs in a narrow band of e_g electrons) along with the fact that it can be alloyed with the narrow band-gap semiconductor FeS_2 opens up the possibility of "tuning" the position of the Fermi level in $Co_{1-x}Fe_xS_2$ to obtain an itinerant ferromagnet with $1\mu_B$ per formula unit. Moreover, Mazin[23] has predicted that the half-metallicity in $Co_{1-x}Fe_xS_2$ should be particularly robust; i.e., it should occur over a wide composition range, be insensitive to defects and crystallographic disorder, and could even be expected to have fewer problems with retaining full spin polarization at surfaces.[23] Experimentally it has been found that CoS_2 has a magnetization of $0.9\mu_B$/Co,[24,25] consistent with large, but $<100\%$, polarization.

Relatively little experimental data are available on the magnetic and transport properties of this material. NMR,[26] magnetic anisotropy,[26] and heat capacity[27] have been measured, along with the composition dependence in $Co_{1-x}Fe_xS_2$ (Refs. 24, 25, 28, and 29), $Co_{1-x}Ni_xS_2$ (Refs. 24 and 28), and $CoS_{2-y}Se_y$ (Refs. 28 and 30) alloys. Basic transport measurements have been made,[24,25,30,31] but we are not aware of any report of magnetotransport measurements. In this paper we present detailed high-field magnetotransport data in addition to magnetic characterization. The results show intriguing behavior in the vicinity of T_C, which we interpret in terms of spin-dependent band structure effects. A direct measurement of a spin polarization of 56% shows that CoS_2 is not completely polarized, but provides encouragement for further work on the Fe-doped system $Co_{1-x}Fe_xS_2$.

CoS_2 polycrystals were prepared by sintering CoS_2 powder in a S atmosphere. 99.9%-pure CoS_2 powder was pressed under 20 000 psi into 150-mg pellets and sealed in evacuated ($<1\times10^{-6}$ Torr) quartz tubes with 100 mg of 99.999%-purity S. The material was then reacted at 700–900 °C for 24 h and furnace cooled over 12 h. The quartz tube was arranged in the furnace such that the S vapor condensed at the opposite end of the tube to the sample on cooling. As can be understood on inspection of the Co-S phase diagram, the outcome is critically dependent on S vapor pressure and temperature. Too little S vapor pressure results in $CoS_{1-\delta}$ or Co_9S_8 when the CoS_2 dissociates on heating, whereas an excess of S vapor is required for stoichiometric CoS_2. Samples were characterized by x-ray diffraction (XRD), scanning electron microscopy, and energy dispersive analysis of x rays (EDAX), confirming that the material is single-phase polycrystalline CoS_2 with a grain size of ~10 μm. XRD and EDAX both confirm Sulfur stoichiometry within the experimental resolution.

Electronic transport measurements were performed with the standard four-probe method, employing an ac excitation at 13.7 Hz. Measurements were made from 325 to 1.5 K in magnetic fields up to 17 T. Magnetic characterization was performed in a commercial magnetometer down to 5 K in fields up to 9 T. dc magnetization was measured by the extraction method, while ac susceptibility was measured at 100 Hz using conventional "pickup coil" detection. Point-contact Andréev reflection (PCAR) employed a Pb tip. Conductance-voltage $[G(V)]$ curves were measured using an ac modulation technique and the resulting curves were fitted to a modified Blonder-Tinkham-Klapwijk (BTK) model, as discussed later. Further experimental details are provided in Ref. 10.

Figure 1 shows the temperature dependence of the resistivity (ρ) showing a linear dependence from 300 K down to about 125 K, followed by a large anomaly in the vicinity of T_C. At temperatures below 60 K the resistivity follows T^2 very well, as demonstrated in the inset of Fig. 1. This T^2 dependence has been observed before in potential half-metal systems[32] and can be interpreted in terms of $P=100\%$ (by electron-electron interactions) or in terms of $P<100\%$ (by electron-magnon scattering). In the absence of the predicted temperature dependence for double-magnon-scattering processes[32,33] or the distinctive dependence seen in CrO_2,[34] it is ambiguous to attempt to determine half-metallicity from these $\rho(T)$ data.

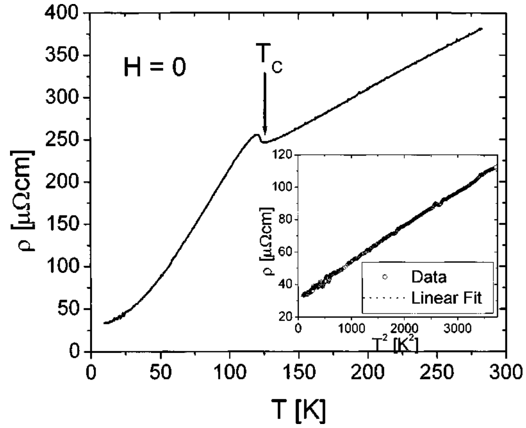

FIG. 1. Temperature dependence of the zero-field resistivity of CoS_2. The arrow marks the Curie temperature. Inset: resistivity plotted vs T^2 for $T<60$ K, along with a linear fit.

Clearly the central question that arises from Fig. 1 is the origin of the anomaly near T_C. This effect has been observed before,[24,25,30,31] where the possibility of critical scattering was ruled out on the basis that (a) the effect is too large and (b) fitting to the scaling laws for critical scattering produces unphysical parameters.[31] To shed light on this issue we made measurements of ρ, magnetization (M), and ac susceptibility (χ_{ac}) as a function of temperature at various magnetic fields. Figure 2 shows the T dependence of M and χ_{ac} measured in magnetic fields from 0 to 9 T. The remarkable aspect of these data is the large shift in apparent T_C with magnetic field, along with the unusually sharp vanishing of M ($\mu_0 H$

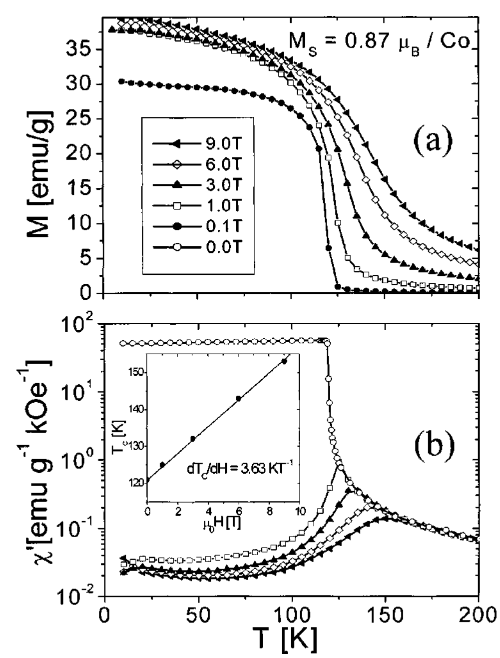

FIG. 2. Temperature dependence of (a) the magnetization and (b) the in-phase component of the ac susceptibility ($\omega=100$ Hz) in dc measuring fields of 0.0, 0.1, 1, 3, 6, and 9 T. In both cases the sample was zero-field cooled. Inset: magnetic field dependence of the Curie temperature, as determined from the ac susceptibility peak.

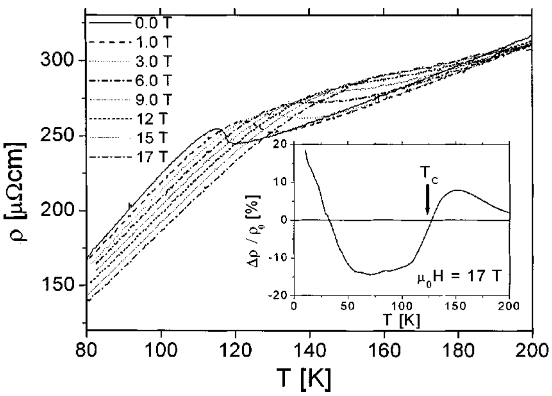

FIG. 3. Temperature dependence of the resistivity, in the vicinity of T_C, for magnetic fields of 0, 1, 3, 6, 9, 12, 15, and 17 T. Inset: magnetoresistance ratio as a function of temperature, for a magnetic field of 17 T. $\Delta\rho/\rho_0 = \rho(H) - \rho(0)/\rho(0)$.

$=0.1$ T) as $T \rightarrow T_C$. The apparent shift in T_C with field can be quantified through χ_{ac}, which has a peak at $T_C(H)$. This is plotted in the inset to Fig. 2(b), where it can be seen that the apparent T_C increases linearly with applied field. Broadening of the transition from ferromagnet to paramagnet in high fields is expected in any conventional system (in fact, the phase transition is only well defined in the zero-field limit), but in this case there are a number of clear indications that the transition is not conventional and that a shift in T_C with applied external field truly takes place. Specifically, given the sharp vanishing of M near T_C and the fact that doping only 5% of Se onto the S site or 12% of Ni onto the Co site leads to a first-order ferromagnetic transition,[24,28,30] we suggest that CoS_2 is remarkably close to exhibiting a first-order phase transition. As discussed later, $M(H)$ curves above T_C provide the essential evidence as they exhibit distinct upward curvature and a peak in dM/dH, indicating field-induced ferromagnetism at $T > T_C$. This is consistent with our argument that a true shift in T_C occurs with increasing external field.

As shown in Fig. 3, we also measured $\rho(T)$ in the extended field region up to 17 T. The anomaly in ρ near T_C tracks the apparent shift in T_C with applied field and becomes less pronounced with increasing field, but persists to >10 T. This is inconsistent with simple expectations for critical scattering. The net result of this shift in the resistivity anomaly with increasing field is the existence of a negative magnetoresistance (MR) below T_C and a positive MR above T_C. This is shown more clearly in the inset to Fig. 3, which plots the magnetoresistance ratio $\Delta\rho/\rho_0(T)$. The MR at $T < T_C$ achieves a maximum negative value of -15% and crosses zero at $T = 125$ K [i.e., very close to $T_C(H=0)$], eventually reaching a maximum positive value of 8%. At higher T the MR vanishes. (Note that positive MR effects are observed at low T, probably of conventional "Lorentz force" origin.)

The negative MR at $T < T_C$ is most simply interpreted in terms of the field suppression of electron-magnon scattering, which exists in all conventional ferromagnets.[35] The temperature dependence is qualitatively consistent with this interpretation (i.e., it is small at low temperatures where few

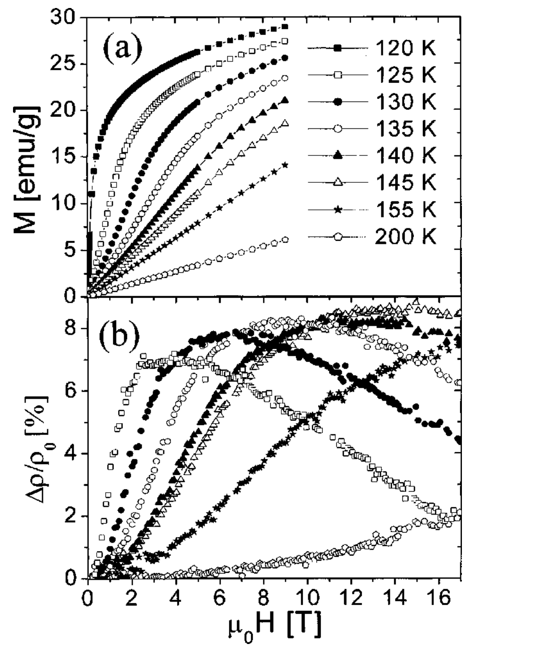

FIG. 4. Magnetic field dependence of (a) the magnetization and (b) the magnetoresistance for T $(>T_C) = 120$, 125, 130, 135, 140, 145, 155, and 200 K. Data were taken at both positive and negative fields to rule out contributions from the Hall effect.

magnons are excited, reaches a maximum at some intermediate temperature, and vanishes at T_C where spin-wave excitations cease to exist), as is the field dependence. Obviously, if we are truly observing electron-magnon (spin-flip) scattering, then the system must have $P < 100\%$,[36] implying that CoS_2 is not half-metallic or that the spin polarization is reduced at these elevated temperatures. This point will be directly addressed later by a measurement of the conduction-electron spin polarization by PCAR. We note that the general situation *below* T_C (i.e., positive MR at low T, crossing over to negative MR at higher T due to the onset of spin-flip scattering) is similar to CrO_2.[34]

The positive MR at $T > T_C$ is of greater interest. Isothermal field sweeps are shown in Fig. 4, along with the corresponding $M(H)$ curves. The general behavior of the MR is independent of T; the MR increases with increasing field, reaching a maximum at about 8%, followed by a decrease at higher fields. The field required to reach the maximum positive MR is temperature dependent, but the value of that MR (7%–8%) is not. The corresponding $M(H)$ curves show a rapid increase in M for $T \approx T_C$ and the expected linear dependence at $T \gg T_C$. However, in the intermediate region (T just greater than T_C) we observe a small but distinct upward curvature in $M(H)$, indicating metamagnetism, where the ferromagnetic (FM) state is induced by the field. This is more clearly seen in Fig. 5 where dM/dH is plotted as a function of H, revealing clear increases with increasing field when T is just above T_C. The peaks in dM/dH mark the entry into the field-induced ferromagnetic phase.[37] As mentioned earlier, this is consistent with the sharp vanishing of M near T_C, the large field-induced shift in the apparent T_C, and the crossover to a first-order transition with very low

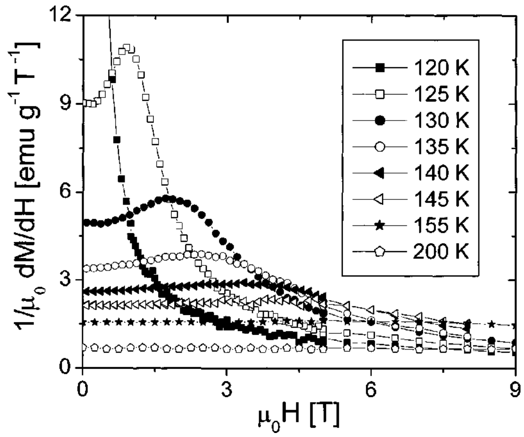

FIG. 5. Field dependence of the derivative of the magnetization with respect to field ($dM/d\mu_0H=1/\mu_0 dM/dH$), at temperatures of 120, 125, 130, 135, 140, 145, 155, and 200 K.

doping.[24,28,30] We observe no hysteresis in $M(H)$ above the critical field and no hysteresis (on cooling and warming) in $M(T)$, indicating that CoS_2 is close to the tricritical point,[37] but is not actually first order.

Taking the $M(H)$ and MR(H) data together, we see that the resistivity obeys a simple universal behavior. As the FM state is approached, the resistivity increases, reaches a maximum $\Delta\rho/\rho_0$ of 8% (when FM order sets in), and then shows a decrease with further increase of magnetic field. Consistent with our earlier interpretation of the negative MR at $T<T_C$, we explain the high-field negative MR (i.e., after FM order sets in) in terms of field suppression of electron-magnon scattering, despite the fact that $T>T_C(H=0)$. The key observation required to understand the positive MR above T_C is that the temperature-independent maximum positive MR value (~8%) is very close to the fractional increase in resistivity obtained when cooling through T_C in zero field. Simple extrapolation of the linear $\rho(T)$ behavior observed above T_C (Fig. 1) to temperatures slightly below T_C results in a calculated increase in resistivity on cooling of 7.5%. This suggests that an ~8% resistivity increase is obtained whenever the FM state is entered, whether by cooling through T_C in zero field *or* by applying large magnetic fields at $T>T_C(H=0)$. Our argument is further strengthened by the analysis shown in Fig. 6 where the MR(H) curves of Fig. 4(b) are replotted as a function of *M*. The data show a very reasonable collapse to a single universal curve, indicating that ρ is controlled only by *M*. The horizontal dotted line shows the fractional increase in resistivity on cooling in $H=0$, demonstrating the close agreement with the maximum MR(H) values. Note that this scaling is only possible for the positive MR component. At higher fields, where ρ decreases due to the suppression of electron-magnon scattering in the field-induced FM state, the scaling can no longer be performed.

We suggest that the increase in resistivity on entry into the FM state is the consequence of a simple spin-dependent band structure effect. Specifically, we propose that as FM order sets in and the narrow CoS_2 conduction-band exchange splits, the total (spin-averaged) density of states (DOS) at the

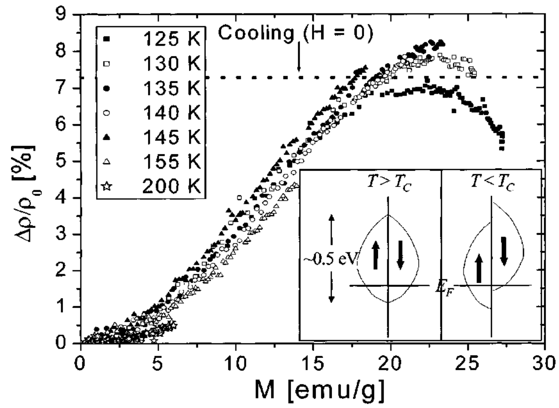

FIG. 6. Magnetoresistance data of Fig. 4 plotted vs the magnetization. The curve was obtained by taking the MR(H) data of Fig. 4(b) and using the known relationship between *M* and *H* at these temperatures [Fig. 4(a)] to plot MR vs *M*. The dotted horizontal line signifies the fractional resistance change obtained on cooling in zero field (see Fig. 1). Inset: simple schematic depiction of the spin-resolved DOS above and below T_C.

Fermi level [$N(E_F)$] is reduced. As a specific example, if the Fermi level is positioned near the bottom of the band at $T>T_C$, then the exchange splitting will result in a slightly increased majority-spin DOS, but a drastic reduction in the minority-spin DOS, as illustrated schematically in the inset to Fig. 6. It is worth pointing out that obtaining highly spin-polarized currents does not require that the exchange splitting force E_F into a gap for the minority spins, but only that the splitting be sufficient to push E_F below the mobility edge, where the minority-spin electronic states become localized and hence do not contribute significantly to transport. We can also advance an alternative explanation of a similar nature based on the calculated density of states due to Mazin.[23] These calculations used the full-potential linear augmented plane-wave method and predict that the Fermi level in *paramagnetic* CoS_2 is situated near a sharp peak in the density of states and that, as a consequence, $dN(E)/dE$ is very large at E_F. Entry into the ferromagnetic phase and the ensuing exchange splitting of the energy bands is therefore likely to result in large changes in $N(E_F)$. Such a situation already prompted Mazin to predict that "interesting" transport properties are to be expected in this material.

It seems that our transport measurements and the value of the saturation magnetization ($0.87\,\mu_B$/Co) are consistent with high, but not complete, spin polarization in CoS_2. To shed further light on this we performed PCAR measurements[38–40] (using a Pb tip at 4.2 K) on the same polycrystals used in this study to directly measure *P*. The results are shown in Fig. 7. Following the method of Ji *et al.*,[10] we extracted the spin polarization and *Z* parameter (which quantifies the interfacial barrier strength) from fits to the conductance-voltage curves using the modified BTK model.[41] These data are then collected as a function of the parameter *Z* to determine the spin polarization in the limit of a perfectly transmissive interface. Further details of the experimental technique and fitting procedure are given in Ref. 10. Figure 7 displays the $G(V)$ curves at the low and high

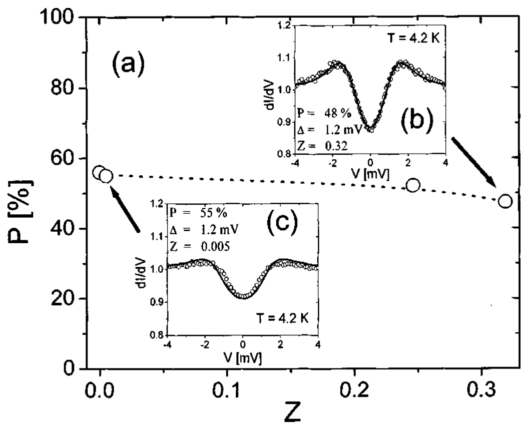

FIG. 7. (a) Spin polarization (P) as a function of the parameter (Z) (explained in the text). Insets (b) and (c) show representative conductance-voltage curves in the high- and low-Z limits, respectively. $T=4.2$ K.

ends of the range of Z values, as well as the dependence of P on Z. The extracted P values are weakly dependent on Z, the $Z=0$ limit being a spin polarization of 56%. This clearly demonstrates that CoS_2 is not completely polarized. This is consistent with our observation of conventional electron-magnon scattering magnetoresistivity and, moreover, a simple calculation shows that this value of spin polarization is consistent with our explanation for the increase in resistivity on entrance into the ferromagnetic phase. Assuming rigid free-electron-like bands [i.e., $N(E)\propto E^{1/2}$] and a conductivity directly proportional to the spin-averaged DOS at E_F (i.e., $N_{\rm av}(E_F)=[N_\uparrow(E_F)+N_\downarrow(E_F)]/2$), we can relate the parameter $P=[N_\uparrow(E_F)-N_\downarrow(E_F)]/N_\uparrow(E_F)+N_\downarrow(E_F)$ to the relative conductivity decrease likely to take place on entering the ferromagnetic phase. Using the measured spin polarization of 56%, we estimate a 13% increase in resistivity on entry into the ferromagnetic phase. Although the calculation is crude (it assumes free-electron-like behavior and ignores the effects due to the variation of v_F^2, where v_F is the Fermi velocity), the agreement with our measured value of 7%–8% is reasonable, adding further weight to our proposed band structure mechanism.

In summary, we have presented the magnetic and magnetotransport properties of CoS_2. We have determined that the system is close to the tricritical point separating first- and second-order magnetic transitions. The Curie temperature shifts dramatically with applied field, as does the resistivity anomaly. This leads to a negative magnetoresistance below the Curie temperature (which is interpreted in terms of electron-magnon scattering) and a positive magnetoresistance above it. This increase in resistivity was proved to be controlled only by the magnetization, regardless of whether the ferromagnetic state is induced by cooling or by application of a large field. We interpret the increase in resistivity in terms of a simple spin-dependent band structure effect due to the reduction in the minority-spin density of states at the Fermi level. The conduction-electron spin polarization was found to be 56% from point-contact Andréev reflection and, moreover, this was shown to be consistent with our proposed spin-dependent band structure effect in the transport. Although we have demonstrated that stoichiometric polycrystalline CoS_2 is not completely polarized, the relatively large spin polarization is encouraging for further work in the $Co_{1-x}Fe_xS_2$ system.

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