

Measurement of Conduction-Electron Polarization via the Pairing Resonance

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We show that the pairing resonance in the Pauli-limited normal state of ultrathin superconducting Al films provides a spin-resolved probe of conduction-electron polarization in thin magnetic films. A superconductor-insulator-ferromagnet tunneling junction is used to measure the density of states in supercritical parallel magnetic fields that are well beyond the Clogston-Chandrasekhar limit, thus greatly extending the field range of the tunneling density of states technique. The applicability and limitations of using the pairing resonance as a spin probe are discussed.

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The possibility of incorporating spin degrees of freedom into electronic technologies has led to an explosion in research into mechanisms of spin polarization of conduction currents in semiconducting and metallic systems [1,2]. In addition to being technologically important, spin polarization in conducting systems remains a fundamentally interesting many-body problem. Clearly, an accurate determination of the conduction-electron polarization P , particularly in thin-film magnetic structures, is crucial both from the point of view of “spintronic” device development and basic research. Unfortunately, however, there are few direct probes of P . The three most successful techniques, listed in historical order, have been Zeeman-split superconducting tunneling density of states spectroscopy (SCTDOS) [3,4], spin-resolved photoemission spectroscopy (SRPES) [5], and point contact Andreev reflection (PCAR) [6,7]. Each of these techniques has its own unique advantages and limitations. SCTDOS has outstanding resolution $\sim 10 \mu\text{V}$, and is compatible with thin-film geometries, but is limited to a narrow range of magnetic fields and is not easily implemented with bulk samples [4]. SRPES has a relatively low resolution of $\sim 10 \text{ mV}$, and does not discriminate well between itinerant and localized bands. PCAR can be used on bulk samples and thick films, but it requires very low impedance point contacts, and is also incompatible with magnetic fields [7,8]. In this Letter we introduce an extension of the SCTDOS technique that exploits the spin structure of the Pauli-limited normal-state (PLNS) pairing resonance [9,10] to measure electron polarization in magnetic fields well above the superconducting critical field. We show that the technique not only greatly extends the range of fields over which the polarization can be measured, but it can be much less sensitive to field misalignment than SCTDOS.

Tedrow and Meservey [4] pioneered the use of superconducting spin-resolved tunneling to directly measure the electron polarization in magnetic films. The technique utilizes a planar junction geometry consisting of a

superconductor-oxide-ferromagnet (SC-Ox-FM) sandwich in which the superconductor counterelectrode is purposely made very thin. The tunnel junction is then cooled below the superconducting transition temperature and a carefully aligned magnetic field is applied parallel to the junction plane. If the SC film thickness t is much less than the superconducting coherence length ξ , then the Meissner response to the applied field is suppressed, and the critical field of the SC counterelectrode is entirely Zeeman mediated [11]. If the spin-orbit scattering rate of the SC is small, then spin is a good quantum number, and the spin rotation symmetry of the SC can be exploited to provide a spin-resolved probe. In the Tedrow and Meservey technique the polarization is measured in the superconducting phase by applying a subcritical parallel magnetic field to the tunnel junction at low temperature, $T \ll T_c$. This induces a Zeeman splitting of the SC quasiparticle DOS spectrum, and the BCS coherence peaks get split into spin-up and spin-down bands by the parallel field [12]. As will be discussed below, the relative heights of these peaks give a direct measure of the electron polarization in the FM. Here we show that polarization can, in fact, be measured in fields that are several times larger than the parallel critical of the SC by using the PLNS pairing resonance in appropriately designed tunnel junctions.

The SC-Ox-FM tunnel junctions were formed by first depositing a thin Al counterelectrode via e -beam deposition of 99.999% Al stock onto fire polished glass microscope slides held at 84 K. The depositions were made at a rate of $\sim 0.1 \text{ nm/s}$ in a typical vacuum with pressure $< 3 \times 10^{-7} \text{ Torr}$. After deposition, the counterelectrode was exposed to the atmosphere for 10–20 minutes in order to allow a thin native oxide layer to form. Then a 45 Å thick FM film was deposited onto the counterelectrode with the oxide serving as the tunneling barrier. In this study the FM was either CNI_3 or CCO_3 , where the e -beam depositions were made from arc-melted buttons. The counterelectrode thicknesses were chosen so that their in-plane sheet resist-

ance was $\sim 1\text{--}2$ k Ω /sq. This corresponded to thicknesses that were typically 22–24 Å. The low temperature parallel critical fields of the counterelectrodes were ~ 6.5 T. The junction area was about 1 mm \times 1 mm, while the junction resistance ranged from 15–100 k Ω depending on exposure time and other factors. Only junctions with resistances much greater than that of the films were used. Measurements of resistance and tunneling were carried out on an Oxford dilution refrigerator using a standard ac four-probe technique. Magnetic fields of up to 9 T were applied using a superconducting solenoid. A mechanical rotator was employed to orient the sample *in situ* with a precision of $\sim 0.1^\circ$.

In the upper panel of Fig. 1 we plot the 70 mK tunneling conductance of a Al-AIO_x-CNI₃ tunnel junction in a subcritical parallel magnetic field of 4 T. The Zeeman splitting of the BCS DOS spectrum is clearly evident, where we have labeled the spin moment associated with each peak. Normally, the respective spin peaks would be identical on either side of the Fermi energy. The asymmetry arises from the unequal spin populations in the ferromagnetic CNI₃. This is expected since the magnetization properties of the CNI₃ and CCo₃ are very similar to their elemental counterparts [13]. If one assumes that spin is conserved in the tunneling processes, then tunneling currents from the spin-up (down) bands in the Al will only tunnel into corresponding spin-up (down) bands in the ferromagnet. Tedrow and Meservey exploited this conservation property to extract the polarization of a variety of transition-metal FM films [4],

$$P = \left| \frac{\delta_1 - \delta_2}{\delta_1 + \delta_2} \right|, \quad (1)$$

where the peak height differences $\delta_{1,2}$ are defined in Fig. 1(a) and the corresponding polarization of the CNI₃ is $\sim 18\%$.

The zero-field gap energy for the Al counterelectrode used in Fig. 1 was determined to be $\Delta_o \sim 0.49$ meV by fits to the superconducting DOS spectrum. Because the thickness of the Al counterelectrode is much less than the superconducting coherence length $\xi \sim 150$ Å, it undergoes a first-order parallel critical field transition at the Clogston-Chandrasekhar critical field [14,15]

$$H_c^{CC} = \frac{\Delta_o \sqrt{1 + G^0}}{\sqrt{2} \mu_B}, \quad (2)$$

where μ_B is the Bohr magneton and G^0 is the antisymmetric Landau parameter [16]. In panel (b) of Fig. 1 we show the DOS spectrum at the critical field. This spectrum is in the coexistence region between the superconducting phase and the Pauli-limited normal state. The new feature that appears at the transition is a manifestation of the pairing resonance (PR). Finally, in panel (c) we show a normal-state spectrum where the BCS coherence peaks have been extinguished. The remaining structure consists of a broad, symmetric, background with two small satellite

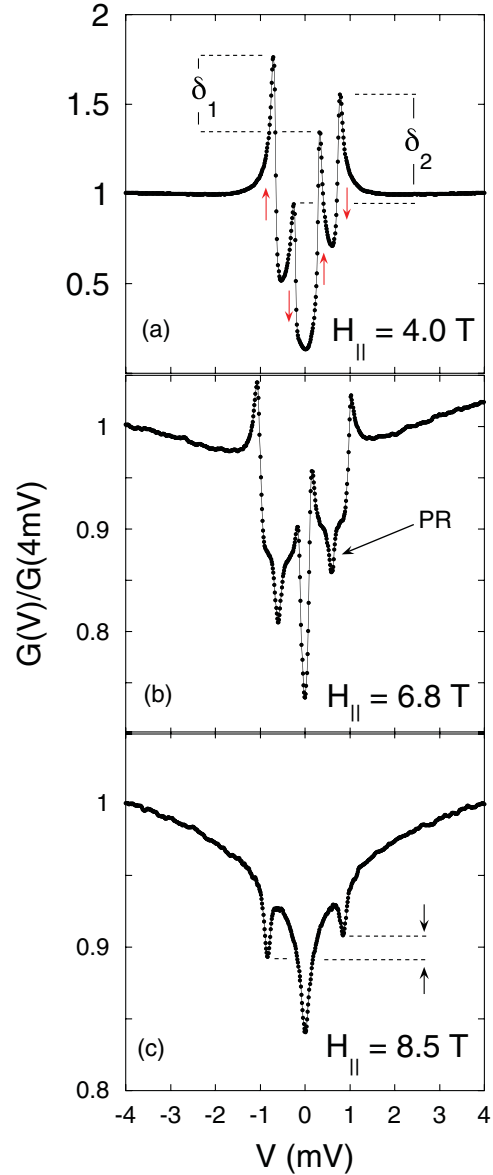


FIG. 1 (color). Evolution of the tunneling conductance of a Al-AIO_x-CNI₃ tunnel junction as the parallel critical field transition is crossed at 70 mK. (a) superconducting phase showing an asymmetric Zeeman-split DOS spectrum. The arrows denote the spin assignment of the coherence peaks. (b) DOS spectrum at the parallel critical field transition in which the normal-state pairing resonance (PR) coexists with superconducting coherence peaks. (c) Pauli-limited normal state in which only the pairing resonance and the zero bias anomaly remain. Note that the positive and negative resonances have different magnitudes.

resonances on either side of $V = 0$. The background feature is often referred to as the zero bias anomaly and is a well documented electron-electron interaction effect [17,18]. In contrast, the satellite features represent incoherent Cooper pairing. The fact that the resonance dips have unequal magnitude in Fig. 1 indicates that they are spin specific. As we show below, they can be used to

extract FM polarization in fields well beyond the Clogston-Chandrasekhar critical field of Eq. (2).

For the purposes of measuring polarization the most important property of the PR is its spin structure. In Fig. 2 we present a graphic representation of the resonance as it is observed in tunneling into a paramagnetic (PM) metal film. Since the PM has no preferred spin direction, the resonance is symmetric about $V = 0$. As is depicted in Fig. 2(a), when a sufficient forward-bias voltage is reached, spin-down electrons in the PM can tunnel across to form doubly occupied levels close to the top of the spin-up band in the PLNS. These spin-singlet states can then mix with the unoccupied states in the near vicinity to form an evanescent Cooper pair [9]. This effectively produces a small depletion of spin-down quasiparticle states due to the fact that they have been consumed by the resonance. By particle-hole symmetry there is a similar depletion of spin-up states at the reverse-bias voltage needed for spin-up electrons lying just above the PLNS doubly occupied sites to tunnel over to the top of the spin-up band in the PM. The precise energy of these resonances is field dependent,

$$eV^* = \frac{1}{2}(E_z + \sqrt{E_z^2 - \Delta_0^2}), \quad (3)$$

where $E_z = (2\mu_B H)/(1 + G^0)$ is the Zeeman energy re-normalized by G^0 .

The zero bias anomaly background in Fig. 1(c) is independent of magnetic field and varies as $\ln V$ for $V \geq k_B T/e$. It can easily be subtracted from the data in order to isolate the resonances as shown in Fig. 3. The red dots are data taken on a Al-AIO_x-CCO₃ tunnel junction at 70 mK in a 6.7 T parallel magnetic field. The arrows depicted the spin assignments of the resonances and δ_{\pm}

refer to their respective amplitudes. The solid black line is a best fit to the resonance profile using a procedure and formalism described elsewhere [19]. As was the case for the superconducting phase, we only need the relative magnitudes of the positive and negative-bias resonances in order to determine the polarization. If we take the PLNS density of states to be N^s , then well away from the PR spin rotation symmetry requires $N_{\uparrow}^s = N_{\downarrow}^s = N^s/2$. On resonance, however, there will be a depletion of one spin component at positive bias and the other at negative bias,

$$N_{+,-}^s = N_{\downarrow,\uparrow}^s + N_{\uparrow,\downarrow}^s(1 - \epsilon), \quad (4)$$

where $0 \leq \epsilon \leq 1$ represents the strength of the resonance. In general, ϵ depends upon a number of factors, including magnetic field, temperature, spin-orbit scattering rate, and the dimensionless normal-state transport conductance, g [9,19]. Spin conservation forbids intra-spin-band tunneling, so if we assume that the ferromagnet has a majority spin density N_{\uparrow}^f and a minority spin density N_{\downarrow}^f , then the tunneling conductance is simply proportional to the product of respective spin-specific DOS on either side of the tunnel junction as is depicted in Fig. 2(b). The magnitudes of the positive- and negative-bias resonance features are

$$\begin{aligned} \delta_{+,-} &= [N_{\downarrow,\uparrow}^s N_{\downarrow,\uparrow}^f + N_{\uparrow,\downarrow}^s N_{\uparrow,\downarrow}^f(1 - \epsilon)] - [N_{\uparrow,\downarrow}^s N_{\uparrow,\downarrow}^f + N_{\downarrow,\uparrow}^s N_{\downarrow,\uparrow}^f] \\ &= \frac{-\epsilon N^s N_{\uparrow,\downarrow}^f}{2}. \end{aligned} \quad (5)$$

From this the polarization follows,

$$P = \left| \frac{N_{\uparrow}^f - N_{\downarrow}^f}{N_{\uparrow}^f + N_{\downarrow}^f} \right| = \left| \frac{\delta_+ - \delta_-}{\delta_+ + \delta_-} \right|. \quad (6)$$

The data in Fig. 3 give a polarization of 39.5% which is comparable to values reported for pure Co by the early work of Tedrow and Meservey [4] and later measurements

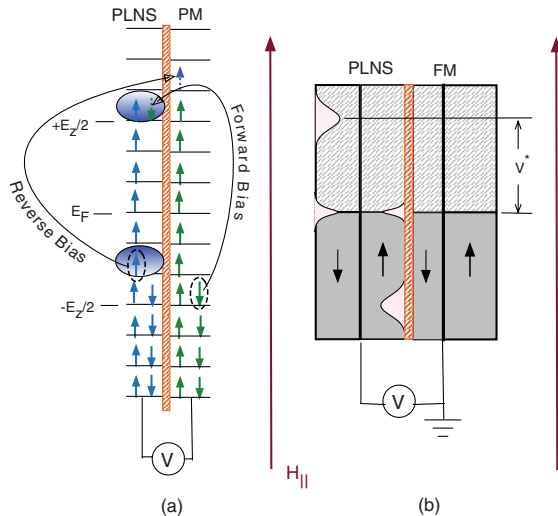


FIG. 2 (color). (a) Schematic of the spin structure of the pairing resonance. (b) DOS profiles of a tunnel junction comprised of an Al film in the Pauli-limited normal state (PLNS) on one side and a ferromagnetic film on the other. Note the depletion of states in the PLNS due to the pairing resonance and the zero bias anomaly.

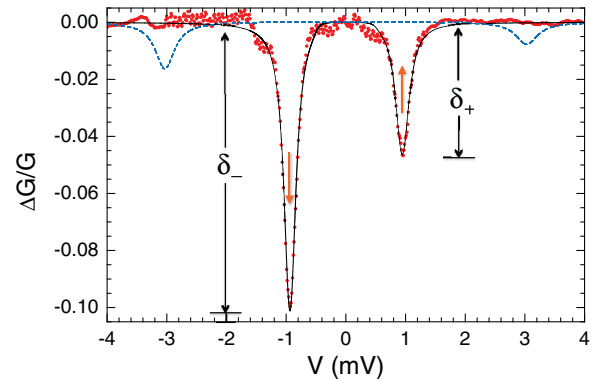


FIG. 3 (color). The red symbols are tunneling spectra taken at 70 mK in a parallel field of 6.7 T, where the zero bias anomaly background has been subtracted off. The orange arrows denote the spin assignment of the occupied and unoccupied resonances. The solid black line represents a best fit to the resonance curve. The dashed blue line is the predicted resonance profile at 18 T, extrapolating from the fitting parameters obtained at 6.7 T.

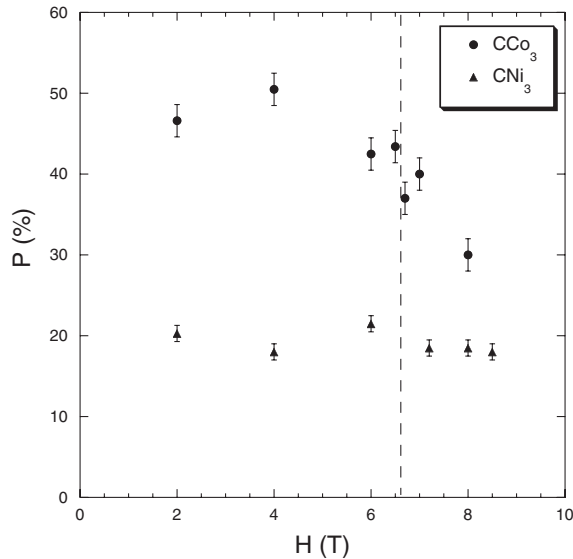


FIG. 4. Electron polarization of 45 Å CNI₃ and CCo₃ films as a function of magnetic field. The dashed line represents the approximate parallel critical field of the Al counterelectrodes used in these measurements.

using PCAR [7]. We note that Eq. (6) is independent of both the PR's strength ϵ and its width. Thus within the limits of signal-to-noise constraints, one should be able to obtain polarization measurements at quite high fields. The dashed line in Fig. 3 represents a prediction for the resonance curve at 18 T obtained by extrapolating the necessary parameters from fitting the data at 6.7 T. Although the PR is significantly attenuated and broadened by the high field, it is still well within the noise level of the data. Since the strength of the resonance grows as the conductance g is reduced, one can use slightly thinner counter-electrodes to increase the visibility of the resonance. Previous studies have shown that the PR in counter-electrodes with $g \leq 6$ remains well defined in perpendicular fields of a few teslas, thus eliminating the need for precise parallel alignment [20].

In Fig. 4 we present electron polarization as a function of magnetic field for CNI₃ and CCo₃ films. The vertical dashed line represents the approximate critical field of the counter-electrodes used in this study. The data show good agreement between polarization values measured just below $H_{c||}$ and normal-state values measured just above $H_{c||}$. The polarization of CNI₃ film is independent of field, as would be expected considering the fact that the Zeeman energies associated with fields of a few teslas are less than 0.1 meV, which is much less than exchange energies associated with the magnetization. Interestingly, though, the polarization of the CCo₃ film exhibits a significant decrease in fields above 3–4 T. This trend can be seen in both the superconducting phase measurements and the PR measurements. The origin of this field dependence is uncertain.

In summary, we have shown that conduction-electron spin polarization can be determined from the relative amplitudes of the occupied and unoccupied pairing resonance features. The technique can be used in fields that are several times higher than the Clogston-Chandrasekhar critical field, thus allowing polarization measurements to be made over a very wide range of magnetic fields. Preliminary polarization measurements in CCo₃ films show a strong suppression of the polarization in fields above a few teslas.

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