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Observation of spin-polarized tunneling in thin proximity-effect sandwiches

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Use of a tunneling Hamiltonian model for thin proximity-effect sandwiches in high parallel fields has indicated the possibility of spin splitting the quasiparticle density of states past the point where the up- and down-spin densities of states will cross. This crossing is signified by a large zero-bias peak in the tunneling conductance. In this paper we review the theory leading to this prediction and present the results of spin-polarized tunneling experiments in Al-oxide-Mg-Al junctions. Measurements at 0.4 K on a sandwich with 40 Å of Al over 25 Å of Mg at fields from zero to 3.4 T (where the sandwich went into the normal state) were in qualitative agreement with the theoretical predictions, including the peak at zero bias at the higher field values.

I. INTRODUCTION

Thin-film superconductors in high parallel fields are a unique system in which the spin effects of the field alignment of the superconductor's quasiparticles can be observed unobscured by the diamagnetic properties of superconductors. Such thin films are known to have a variety of interesting properties, including a spin-split density of states[1] and a phase diagram displaying a tricritical point at which the order of the phase transition into the normal state changes from first to second as the temperature is raised.[2] The change in order of the phase transition was first observed experimentally in resistivity measurements[3] and later in tunneling experiments.[4] The tunneling experiments also showed, for the first time, the spin splitting that occurs in the quasiparticle states due to the either parallel or antiparallel alignment of the quasiparticle magnetic moments with the magnetic field. Subsequent to the initial observation of this spin splitting, the experimental and theoretical techniques for its measurement and analysis have been considerably refined. The spin-polarized portions of tunneling characteristics are now used to give information about spin-orbit scattering times in superconductors[5] as well as to measure the spin polarization at the Fermi surface of ferromagnets into which the polarized electrons can be made to tunnel.[6]

Although the observation of the change in order of the phase transition antedates the observation of spin splitting of the density of states, the characteristics of the tricritical point where the change in order occurs have still not been fully explored.[7] This is in spite of the fact that there is much current interest in systems that display tricritical points in their phase diagrams. In the prototypical example of a tricritical point, that occurring in certain metamagnets[8] such as $FeCl_2$, Griffiths[9] has pointed out the usefulness of theoretically introducing a staggered magnetic field. The staggered field couples to the antiferromagnetic order parameter and aids the ordering. The problem with this theoretical construct is its difficulty of experimental realization. It has been suggested, however, that for the analogous phase diagram in thin superconducting films, the proximity effect affords an *experimentally realizable and controllable* analog of the staggered field.[2]

The suggestion that proximity-effect sandwiches in high parallel fields would have interesting critical properties led to a more detailed theoretical investigation of the microscopic properties of such sandwiches.[10] The theory predicted that the sandwiches should show some remarkable structure in the density of states near the Fermi energy when the sandwiches are at low temperatures and high fields as well as unique magnetic properties in the same temperature-field regime. The structure predicted to occur in the density of states results in a corresponding structure that should be readily observable at zero bias in the conductance of a tunneling junction made with a proximity-effect sandwich as one electrode.

In this paper we report on the first experimental study of thin proximity-effect sandwiches in high parallel fields and compare the results with the recent theoretical prediction. Our spin-polarized tunneling measurements verify the existence of the distinctive zero-bias structure predicted to occur in the conductance (dI/dV) in the low-temperature high-field regime. In Sec. II we review the theory that leads to the predicted structure in the density of states and calculate representative conductance characteristics. We then discuss the fabrication of suitable proximity-effect junctions in Sec. III and describe measurements carried out at low temperatures in Sec. IV. Finally, in Sec. V we give a summary and discuss some implications of this work.

II. THEORY OF THE CONDUCTANCE CHARACTERISTICS

In spin-polarized tunneling experiments in type-I superconductors it is necessary that the superconducting films used be very much thinner than the penetration depth to ensure that there is a fairly uniform penetration of a parallel field and that orbital depairing effects arising from diamagnetic screening currents are minimized. Films used in these experiments typically have thicknesses of 100 Å or less. Such films are also thinner than the superconducting coherence length. We therefore consider a bimetalic proximity-effect sandwich in which each metal film is thin compared to both the penetration depth and the coherence length. The separate pair potentials in each film should be approximately uniform over the thickness of each film. At the sandwich interface, the effective electron-electron interaction changes over a few atomic layers and one may reasonably take the resulting pair potential as having a discontinuous jump. In the presence of a field, the pair potential will vary with field but still be uniform within each film.

The detailed properties of such a sandwich structure can be qualitatively understood using a tunneling model first introduced for the proximity effect by McMillan.[11] The bimetallic sandwich is taken to consist of two well-understood constant-gap BCS superconductors described by Hamiltonians H_n^{BCS} and H_s^{BCS} for the weaker (n-side) and stronger (s-side) superconductors, respectively. The coupling between these films is taken into account by a tunneling Hamiltonian H_T which transfers electrons between all states on opposite sides of the sandwich with equal amplitude. The total Hamiltonian H_{tot} of the films is thus

$$H_{\mathrm{tot}} = H_n^{\mathrm{BCS}} + H_s^{\mathrm{BCS}} + H_T \ . \qquad (1)$$

Gallagher, Frota-Pessoa, and Schwartz[10] included the paramagnetic effects of the magnetic field H on the electron magnetic moments in the model by adding to H_n^{BCS} and H_s^{BCS} terms proportional to $\pm\mu_B H$, where μ_B is the Bohr magneton. This model is solved by taking the well-known Green's functions for H_n^{BCS} and H_s^{BCS} and treating H_T as a weak perturbation in a self-consistent second-order perturbation theory.[10,11] Knowing the Green's functions, it is possible to calculate all of the properties of the sandwich.

In Figs. 1(a) and 2(a) we plot (at zero field) the tunneling-model predictions for the sum of the up- and down-spin local densities of states on the n and s sides, respectively, of a proximity sandwich. We have normalized the energies to the zero-field zero-temperature (half) energy gap Δ_s of an isolated s-metal superconductor. The n metal used in this calculation would have an energy gap of $\Delta_n = 0.001\Delta_s$ if there were no proximity effect acting to enhance its superconductivity. The decay widths for tunneling out of the n and s sides are $\Gamma_n = 0.46\Delta_s$ and $\Gamma_s = 0.184\Delta_s$, respectively. These decay widths correspond to a sandwich in which the n film's thickness is 0.4 times that of the s film (for n and s metals with equal bulk density of states at the Fermi energy level when in the normal state). The Debye temperature of both metals is $152\Delta_s$. In Fig. 1(a) it can be seen that the n-side density of states has a substantial energy gap Ω. This gap is due to the proximity effect and in the tunneling model results primarily from electrons tunneling from the n side into the s side and enjoying the s side's (larger) attractive BCS interaction to an appreciable extent. Just above the energy gap, the rounded n-side density of states resembles that of a partially depaired superconductor. At higher energies, those on the order of Δ_s, there is an appreciable drop in the n-side density of states reflecting the increasing number of s-side states into which n-side electrons may tunnel. The s-side density of states shown in Fig. 2(a) have the same energy gap as that on the n side (indeed the states are now

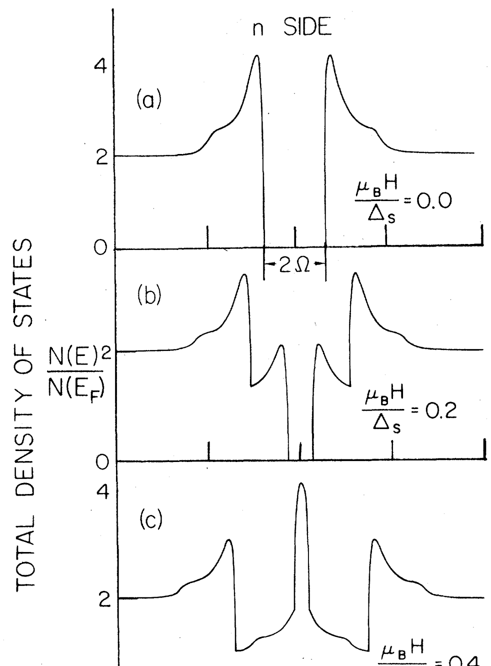

FIG. 1. Predicted sum of the up- and down-spin densities of states on the n side of a thin n-s proximity-effect sandwich for various parallel magnetic field strengths. The density of states has been normalized to the *single-spin* density of states of the n side when the sandwich is in the normal state.

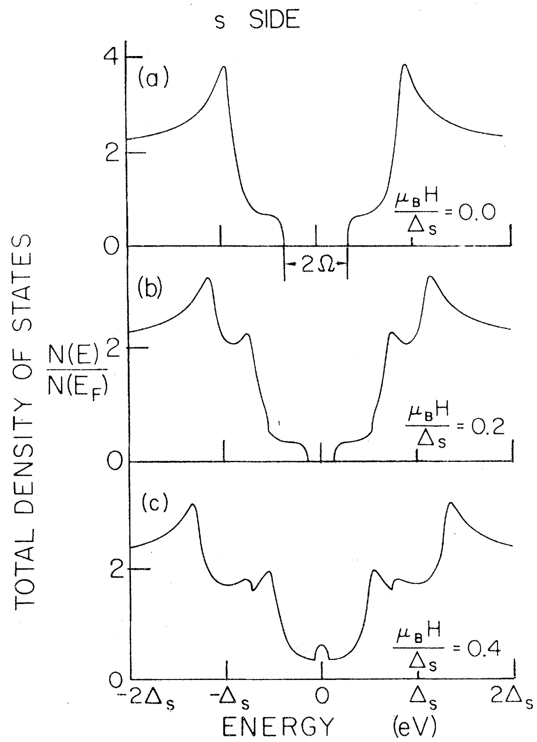

FIG. 2. Predicted sum of the up- and down-spin densities of states on the s side of a thin n-s proximity-effect sandwich for various parallel magnetic field strengths. The density of states has been normalized to the *single-spin* density of states of the s side when the sandwich is in the normal state.

not localized on either side of the sandwich) but there is not an appreciable density for energies much below Δ_s.

As the field is applied [Figs. 1(b) and 2(b)], a splitting of the up- and down-spin densities of states is observed. This splitting results from the either parallel or antiparallel alignment of quasiparticle magnetic moments with the magnetic field.

For an *isolated* s-metal thin-film superconductor at zero temperature, the field which induces a transition to the normal state is the Pauli paramagnetic critical field of $H_P=\Delta_s/(\sqrt{2}\mu_B)$. The Pauli field is determined when the paramagnetic energy lowering of the normal state, $-\frac{1}{2}\chi_P H^2$, equals the superconducting condensation energy of $-\frac{1}{2}N(E_F)\Delta_s^2$, where $\chi_P=2\mu_B^2 N(E_F)$ is the Pauli susceptibility and $N(E_F)$ is the single-spin density of states at the Fermi level when the metal is in the normal state. (A BCS superconductor with a 1 K transition temperature has a Pauli critical field of 1.86 T.) This Pauli critical field is approximately 30% less than the field $H_{sh}=\Delta_s/\mu_B(H_P=H_{sh}/\sqrt{2})$ which would be required to split the spin densities of states to the point where they cross at the Fermi level. The field H_{sh}, superheating field, is the field at which it becomes favorable for a single pair to break up and both electrons align with the field, i.e., the field at which the gain in Zeeman energy $2\mu_B H$ begins to exceed the loss in pairing energy $2\Delta_s$.

In the *proximity-effect* sandwich, Ω/μ_B is the field necessary to split the spin densities of states past the point where they cross at the Fermi level. The field that drives the sandwich normal is now a little smaller than the isolated s-metal Pauli critical field of $H_P=\Delta_s/(\sqrt{2}\mu_B)$. Since the energy gap Ω is much smaller than $\Delta_s/\sqrt{2}$, there is a range of fields for which the spin-split density of states can cross the Fermi energy level. This situation is illustrated in Figs. 1(c) and 2(c). For the n side, the crossing involves a substantial number of states and results in a

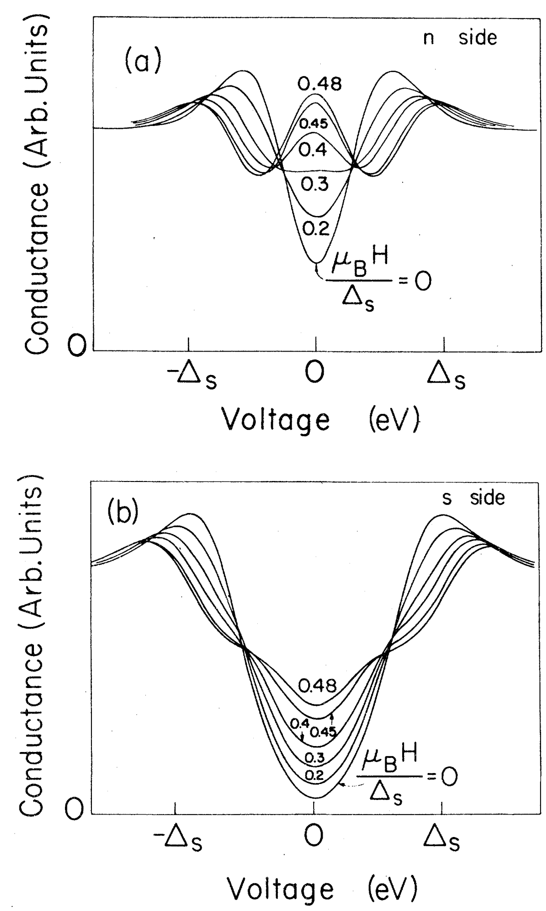

FIG. 3. Thermally smeared versions of the predicted densities of states (Ref. 11) displayed in Figs. 1 and 2. The amount of thermal smearing corresponds to that appropriate for a sandwich at a temperature which is 0.3 times the transition temperature of a bulk superconductor made of s-side material.

significant peak in the total density of states at the Fermi level. As Gallagher, Frota-Pessoa, and Schwartz[10] have discussed, this crossing of the density of states is accompanied by a partial depairing and consequent appearance of a nonzero paramagnetic susceptibility (at zero temperature) and reduction of the self-consistent pair potentials.

In a tunneling measurement between such a proximity-effect structure and a normal-metal counter electrode, one sees in the conductance (dI/dV) versus voltage V characteristics a thermally smeared reflection of the density of states. In Fig. 3 we have calculated thermally smeared versions of the total densities of states[12] shown in Figs. 1 and 2. The amount of thermal smearing corresponds to that appropriate for a sandwich at a temperature which is 0.3 times the transition temperature of an isolated superconductor composed of the s metal. It can be seen that with this amount of thermal smearing the spin splitting at moderate field values cannot be resolved. At higher field values, however, the crossing of the spin-split densities of states does result in a large conductance peak near zero bias in the n-side characteristics. In the s-side characteristics, the corresponding peak in the density of states is so small that it is obscured by the thermal smearing which enters in the conductance calculation. We therefore limited our tunneling measurements to the n side of a proximity-effect sandwich in an attempt to observe the predicted zero-bias peak which is evidence of the spin-split density of states crossing the Fermi energy level.

III. PREPARATION OF THE PROXIMITY-EFFECT SANDWICHES

Several considerations are important for choosing suitable n and s metals in order to observe this effect. It is known from spin-polarized tunneling work on isolated superconductors that the large spin-orbit interaction in higher-atomic-number superconductors suppresses the spin-splitting effects of the magnetic field. It is therefore necessary to use low-atomic-number metals for both the n and s sides of the sandwich. In fact, spin splitting in single thin films has only been resolved in four elemental superconductors: Be, Al, V, and Ga.[13] Al was chosen for the s side of the proximity-effect sandwich studied here since it is by far the least difficult to use. Mg was chosen as the n-side metal because of its low atomic number and because Mg forms good thin films. In order to get a sharp interface between the two metals comprising the proximity-effect sandwich, it is necessary that the metals have low solubilities in each other. The solubilities of Mg and Al in each other were not known, but their dissolution into each other was minimized by deposition onto a cooled substrate.

The proximity-effect tunnel junctions were prepared by vacuum evaporation onto a glass substrate maintained at 77 K. The vacuum before the evaporation was better than 10^{-7} Torr. First the normal metal-counter electrode consisting of a 300-Å Al film was deposited. (Because of the significant flux expulsion from a superconducting film of this thickness, such a film is in the normal state for parallel fields in excess of about 0.15 T.) The film was allowed to thermally oxidize after deposition in order to form the tunneling barrier.

Next the proximity-effect sandwich was formed. The thicknesses of the two metal films comprising the sandwich had to be very carefully controlled. Below a certain minimum thickness (about 40 Å for Al films), the films are agglomerated to an extent that they are not electrically continuous. However, for thicknesses which are even a small fraction of the penetration depth, there is significant orbital depairing resulting in a broadenend density of states and a critical field significantly below the Pauli field, both effects being detrimental to the zero-bias structure we sought to observe. We therefore attempted to make the thinnest possible electrically continuous films.

Below we report on conductance measurements made on two sandwiches. The first consisted of 40 Å of Mg covered by 50 Å of Al and the second of 25 Å of Mg covered by 40 Å of Al. At the time of the tunneling measurements, however, the top Al layer was somewhat thinner than its deposited thickness since it oxidized in air at room temperature for approximately $\frac{1}{2}$ h while the sample was being installed in the cryostat.

IV. MEASUREMENTS AT LOW TEMPERATURES

Isolated Al films with thicknesses on the order of 50 Å have superconducting transition temperatures T_c^s near 2.5 K.[14] (Using the BCS weak-coupling relation $\Delta_s = 1.76\, k_B T_c^s$, we have $\Delta_s = 0.38$ meV.) The proximity-effect structures described here had transition temperatures T_c of 1.7 and 2.3 K for the 40-Å Mg–50-Å Al and the 25-Å Mg–40-Å Al films, respectively. The tunneling measurements were performed in a cryostat maintained at 0.4 K by pumping on liquid ^{3}He. The expected thermal smearing was therefore about half of that included in the calculation of Fig. 3, but the additional thermal smearing put into the calculation accounts approximately for additional broadening mechanisms, such as that due to orbital depairing, which were not included in the calculation. The cryostat was located in an 8-T superconducting magnet and the sample could be rotated until aligned with the field. The dynamic junction conductance dI/dV was measured using a modulation technique with a modulation voltage of 18 μV (rms).

Initial concern focused on having two continuous films; therefore the thicker sandwich (40-Å Mg–50-Å Al) was first fabricated and studied. Upon application of the magnetic field, the conductance characteristics of the junction formed on this sandwich showed some evidence of spin splitting as well as significant field-dependent broadening due to diamagnetic screening (the so-called "orbital depairing"). This relatively thick proximity-effect sandwich underwent a first-order transition into the normal state before a field could be applied which would have been great enough to cause the crossing at the Fermi level. This lowering of the transition field appeared to be due to the significant diamagnetic effects in this relatively thick film.

When the conductance of the thinner film (25-Å Mg–40-Å Al) was measured, there was less evidence of diamagnetic broadening of the density of states. Indeed, the spin splitting of the conductance peaks could just be resolved, and, at fields just below the sandwich's critical field, the zero-bias maximum was observed. Figure 4 shows the experimentally observed tunneling characteristics of this film. (It is at $\mu_B H/\Delta_s = 0.38$ that the spin splitting can just be resolved.) The conductance characteristics and their field dependence are qualitatively as predicted in Fig. 3(a). In particular, there is a significant peak at zero bias for $\mu_B H/\Delta_s = 0.48$ ($H = 3.2$ T). This field is slightly below that where the film underwent a *first-order* transition into the normal state.

It would not be difficult to adjust the parameters of the theory until the experimental curves are quantitatively reproduced. Such a detailed comparison would

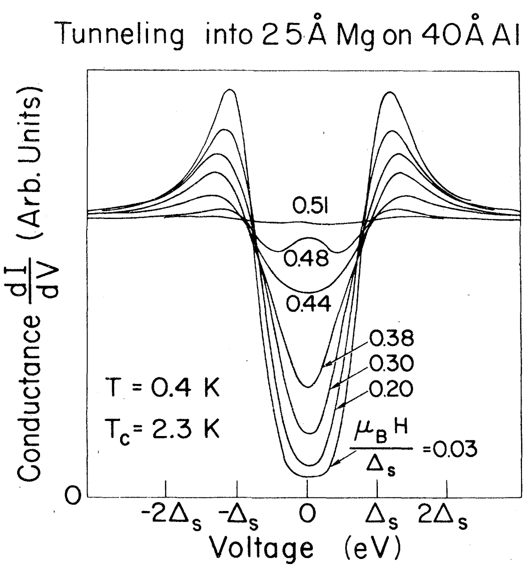

FIG. 4. Conductance of a thin $Al-Al_2O_3-Mg-Al$ proximity-effect junction in various parallel magnetic fields. The unit of energy is $\Delta_s = 0.38$ meV, the low-temperature energy gap of an isolated thin-film Al superconductor ($\Delta_s/\mu_B = 6.6$ T).

not be meaningful, however, unless the experimental characteristics were less broadened and the theory could be trusted quantitatively. To get less broadened experimental characteristics, one would have to go to lower temperatures than are available to us at present. Even if the broadening were thereby reduced, numerous studies have shown that, even in the simpler case at lower fields, the McMillan tunneling model gives only qualitatively correct densities of states.[15]

V. SUMMARY AND DISCUSSION

In summary, we have observed spin splitting of the densities of states in thin proximity-effect sandwiches in high parallel fields including the crossing at the Fermi energy level. Our conductance measurements on the normal-metal side of the proximity sandwich were shown to be in agreement with the qualitative tunneling theory of the proximity effect. It would be interesting to tunnel into the superconducting side of an identical sandwich, necessarily at lower temperature, and look for the corresponding but smaller predicted structure there. However, other effects, such as orbital depairing, may preclude the observation of this structure even at lower temperatures.

Associated with the crossing of the spin-split density of states at the Fermi energy level, there should be an abrupt increase in the paramagnetic susceptibility.[10] Direct experimental verification of this onset would also be interesting. This onset should be observable in a susceptibility measurement and in a Knight-shift measurement. The latter could yield separately the spin susceptibilities on the *n* and *s* sides of the sandwich.

Finally, we also mention that a conductance characteristic similar to that found in Fig. 4 has recently been observed for tunneling into a V-Ti alloy type-II superconductor in both parallel and *perpendicular* fields.[16] It is likely that in this system one is seeing a superposition of the *n*- and *s*-side characteristics calculated in Fig. 3. This superposition comes from the sum of tunneling into the normal-metal-like regions arising from the vortex cores (and perhaps also from a second phase possibly present in the alloy films) and into the surrounding superconducting regions. Although the fractional area of the normal-like vortex cores is small, there is a substantial area near the cores where there is a large screening supercurrent. The local density of states in such a region should resemble that of a superconductor carrying a uniform current[17] and, in particular, should not be BCS-like and should have a much reduced gap. No calculations along these lines have been done, but we mention that Fulde[18] has done a calculation for a dirty superconductor which is valid near a second-order phase transition and has predicted a zero-energy peak in the density of states.

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[12]Figures 1 and 2 show the density of states calculated for self-consistent pair potentials (see Refs. 10 and 11) in the *n* and *s* sides which are appropriate for a sandwich at zero temperature. In Fig. 3 we should use self-consistent pair potentials calculated at the appropriate (nonzero) temperature. We instead used the zero-temperature pair potentials, but there is little temperature dependence at low temperatures.

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