Spin-Dependent Tunneling into Ferromagnetic Nickel

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Tunneling measurements on junctions between very thin superconducting aluminum films and ferromagnetic nickel films in a high magnetic field show that the tunneling current is spin dependent. The effective tunneling density of states in Ni determined by this means implies a polarization of the magnetic moments of the Ni current carriers parallel to the applied field. The technique offers a new method for investigating spin-dependent states in magnetic materials.

Recent tunneling experiments[1] have shown that the quasiparticle energy states in thin superconducting Al films are split in a high magnetic field by the interaction of the field with the quasiparticle spin magnetic moments. Because of the sharpness of the density of states at the edge of the superconducting energy gap, this splitting allows the selection of tunneling electrons of a particular spin direction and permits tunneling investigations of spin-dependent properties of the metals forming the junction. Here we report the use of such a polarized tunnel current to investigate the polarization of the current carriers in ferromagnetic nickel.

Ni is an interesting metal to study because it is believed[2-4] to have a partially filled *d* band with spin up (electron magnetic moment antiparallel to the magnetic induction) and, below the Fermi surface, a full *d* band with spin down. In addition, a partially filled *s* band provides the majority of the conduction electrons. Because of interactions between the unfilled *s* and *d* bands it is believed that the conduction electrons are partially polarized, although the amount and even the sign of the polarization have been a subject of recent controversy.[3,5] A polarized tunneling current should reveal a polarization of the carriers in Ni by causing asymmetry in the tunneling characteristic.

The experiment was performed by measuring the tunneling conductance $\sigma \equiv (dI/dV)_s/(dI/dV)_n$ as a function of voltage for Al-Al_2O_3-Ni junctions in magnetic fields up to 50 kOe. The Al films were about 50 Å thick and carefully aligned with

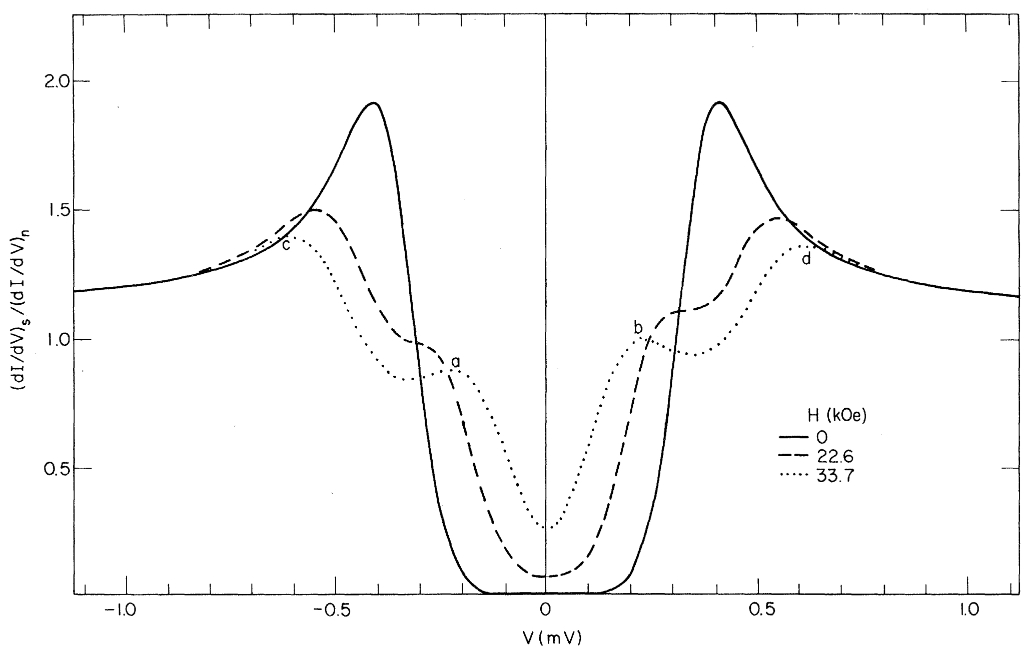

FIG. 1. Normalized conductance of an Al-Al_2O_3-Ni junction measured as a function of the voltage applied to the Al film for three values of applied magnetic field. The asymmetry of the conductance peaks *a*, *b*, *c*, and *d* (H = 33.7 kOe) results from polarization of the Ni carriers.

the magnetic field so that their critical fields were about 45-50 kOe and limited by Pauli paramagnetism.[6] The Ni films were 500 Å thick, and in the high fields used here, the ferromagnetic domains were completely aligned.[7] The superconducting transition temperatures of the Al films were close to 2.5 K and the experiment was performed at 0.4 K. Figure 1 shows measurements of the conductance of a junction as a function of the voltage applied to the Al film for a few selected values of magnetic field. The maxima, which appear at voltages $V=\pm(\Delta\pm\mu H)/e$, are characteristic of the previously reported[1] magnetic splitting of the Al quasiparticle states. The new feature added by the Ni is the asymmetry with respect to voltage: $\sigma_b>\sigma_a$ and $\sigma_c>\sigma_d$, where a, b, c, and d denote the conductance maxima for the 33.7-kOe curve in Fig. 1. This result has a straightforward interpretation in the polarization of the effective tunneling density of states in the Ni.

This interpretation is most simply understood in terms of the analysis of Giaever and Megerle[8] in which the normalized conductance of a superconductor-insulator-metal junction is

$$\sigma(V)=\int_{-\infty}^{\infty}\rho_s(E)\frac{\beta\exp[\beta(E+eV)]}{\{1+\exp[\beta(E+eV)]\}^2}dE. \qquad (1)$$

Here $\rho_s(E)$ is the superconducting density of states, $\beta=1/kT$, E is the quasiparticle energy, and V is the voltage applied to the superconductor. When $T=0$, the second factor under the integral becomes a delta function and $\sigma(V)$ is proportional to $\rho_s(E)$.

In a thin superconducting Al film in a high magnetic field the quasiparticle energies are shifted by $\pm\mu H$, where μ is the absolute value of the magnetic moment of the electron.[6] Figure 2 (upper part) shows a superconducting density of states for $H=0$ (solid line) split by a high magnetic field into spin-up (dashed) and spin-down (dotted) density-of-states curves. For a BCS[9] type of density of states with an energy gap 2Δ

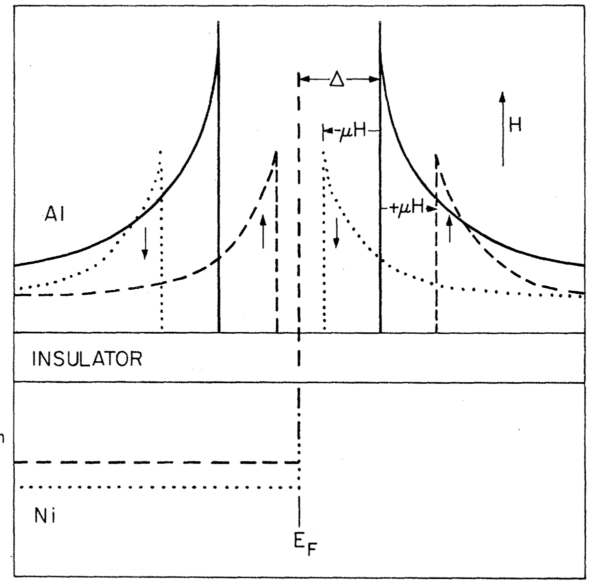

FIG. 2. Tunneling density-of-states diagram showing for the Al a BCS density-of-states split into spin-up (increased in energy by μH) and spin-down (decreased in energy by μH) parts and an assumed predominance of spin-down (magnetic moment parallel to the field) carriers at the Fermi surface of Ni. Arrows on density of states refer to spin direction.

the total density of states which should appear in Eq. (1) is

$$\rho_s(E)=\rho_\downarrow(E)+\rho_\uparrow(E)$$
$$=\tfrac{1}{2}[\rho_s(E+\mu H)+\rho_s(E-\mu H)], \qquad (2)$$

where $\rho_s(E)=|E|/(E^2-\Delta^2)^{1/2}$ for $|E|\geq\Delta$ and $\rho_s(E)=0$ for $|E|<\Delta$. The lower portion of Fig. 2 represents the density of states of a ferromagnetic metal in which the effective tunneling density of states is polarized so that spin-down states (dotted) are more numerous than the spin-up states (dashed). In this case a simple generalization of Eq. (1), assuming no spin-flip tunneling, is

$$\sigma=\int_{-\infty}^{\infty}x\rho_s(E+\mu H)\frac{\beta\exp[\beta(E+eV)]}{\{1+\exp[\beta(E+eV)]\}^2}dE+\int_{-\infty}^{\infty}(1-x)\rho_s(E-\mu H)\frac{\beta\exp[\beta(E+eV)]}{\{1+\exp[\beta(E+eV)]\}^2}dE, \qquad (3)$$

where $x\equiv\rho_{n\downarrow}/(\rho_{n\downarrow}+\rho_{n\uparrow})$ is the fraction of tunneling electrons in the nickel whose magnetic moments are parallel to the applied field. At a sufficiently low temperature the second factor in each integral can be represented by a delta function and the normalized conductance reduces to the simple form

$$\sigma=[x\rho_s(eV+\mu H)+(1-x)\rho_s(eV-\mu H)], \qquad (4)$$

where ρ_s is the same superconducting density of states as in Eq. (1).

From Fig. 2 we can qualitatively understand the experimental result. As the Al voltage increases, the Ni Fermi surface is displaced to the right. Assuming there is no flipping of electron spins during tunneling we expect first a peak (b, Fig. 1) at $eV=\Delta-\mu H$ because of the coincidence of the spin-down Al density-of-states peak with the Fermi surface of the Ni. The corresponding negative voltage peak (a), $eV=-(\Delta-\mu H)$, will be smaller in magnitude because of the smaller spin-up density of states at the Fermi surface assumed for Ni. From Eq. (4), the ratio of the peaks, σ_b/σ_a, is just $x/(1-x)$. In a similar manner σ_c should be somewhat larger than σ_d, although because the conductance at these voltages has contributions from both spin-up and spin-down density of states in the aluminum, the difference should be less. In this case, Eq. (4) shows that the ratio σ_c/σ_d should be

$$\frac{x\rho_s(\Delta)+(1-x)\rho_s(\Delta+2\mu H)}{(1-x)\rho_s(\Delta)+x\rho_s(\Delta+2\mu H)}.$$

The experimental curves agree qualitatively with this interpretation. From the ratio of the heights of the inner peaks σ_b/σ_a we can obtain a value of the effective carrier polarization in the Ni. The polarization $p\equiv(\rho_{n\downarrow}-\rho_{n\uparrow})/(\rho_{n\downarrow}+\rho_{n\uparrow})=2x+1$ is shown in Fig. 3 for various values of the magnetic field as determined from curves like those of Fig. 1. From this figure, $p=0.075$, implying a majority of spin-down (magnetic moment parallel to H) carriers in the Ni.

This analysis assumes that the overlap of the outer peaks onto the inner peaks is negligible even though the conductance curves are considerably broadened from BCS curves. This assumption is supported empirically by the fact (Fig. 3) that the difference in σ at the peaks a and b is not a function of H except near the critical field where the difference must disappear. The assumption that Eq. (3) may be approximated by Eq. (4) probably causes little error at $T/T_c=0.16$.

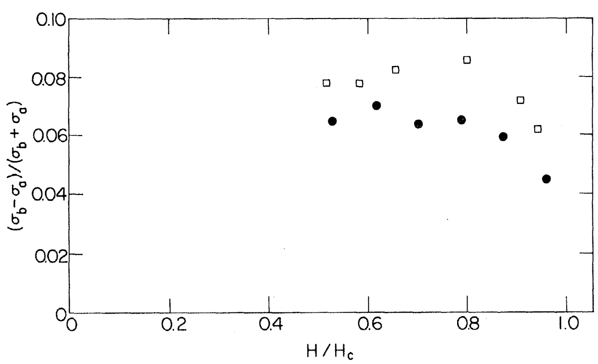

FIG. 3. The apparent polarization as determined from the inner maxima (σ_b,σ_a) in curves like that shown in Fig. 1 as a function of the magnetic field. The value obtained for the polarization $(\sigma_b-\sigma_a)/(\sigma_b+\sigma_a)$ for the carriers in Ni is ≈ 0.075 except near the critical field of the Al. The circles and squares denote different films.

The above interpretation also depends on the assumption of no spin flips in the tunneling process. To test this assumption we have measured the conductance of Al-Al_2O_3-Al junctions in a magnetic field. If there were no spin flips, conductance maxima should appear only at $V=\pm\Delta/e$. If there were spin flips, additional maxima should occur at $V=\pm(\Delta\pm 2\mu H)/e$. In fact only two maxima were observed, implying that spin-flip tunneling is at most responsible for only a very small fraction of the total current. In the case of Al-Al_2O_3-Ni it is possible that Ni atoms diffused into the Al_2O_3 could act as spin-scattering centers. According to Wyatt[10] a minimum voltage $V=\mu_i H/e$ is required to flip the impurity spin, μ_i being the magnetic moment of the impurity center. At the fields used here the minimum voltage should be well above the voltage of first conductance peaks from which the value of p is determined.

The above results imply that the carriers in Ni are partially polarized with their magnetic moments in the field direction. This result is not what one might expect from other knowledge of the band structure. At the Fermi surface, the density of states of minority spins is generally accepted as being much larger than that of majority spins, when we consider both s and d electrons. However, it is unclear how much the d electrons contribute to the tunneling. On the other hand, according to the band-structure calculations by Hodges, Ehrenreich, and Lang,[3] the s electrons have a 1% polarization in the majority direction. Although this is qualitatively in agreement with the present result, the polarizations are not directly comparable since our results apply only to the Fermi surface.

Experimentally Belson,[11] using a method originated by Walmsley,[12] set a lower limit of 8.5% on the carrier polarization in Ni which is consistent with the present result. Recently Bänninger and co-workers[13] have obtained a result very similar to ours by measuring the spin polarization of photoelectrons ejected from Ni. They find electron polarization from 9.4 to 15.5% in the direction of magnetization depending on the method of film preparation. This agrees with our result in sign and approximately in magnitude. The difference may be attributed to film preparation or to the fact that our measurement is localized to the Fermi surface (±0.001 eV) whereas theirs is an average over an energy region of 0.4 or 0.8 eV.

It is certainly premature to take completely seriously the numerical value for the polarization of the carriers of Ni obtained by this simple analysis. Tunneling into strongly interacting *s* and *d* bands is unlikely to fit such a simple model. However, it appears that this method of spin-dependent tunneling should develop into a useful tool for studying spin-dependent energy states in magnetic materials.

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Magnetic Monopole Transitions in Electron Scattering

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The electroexcitation cross section for the second-order magnetic monopole transition $0^+ \to 0^-$ has been calculated in ^{16}O. It is assumed that the spectrum of intermediate excited states is complete and that energy loss may be neglected. If these approximations are valid, it should be possible to observe this transition.

The subject of dispersion corrections in electron scattering has recently been treated in several theoretical and experimental papers,[1-14] mainly for elastic scattering. Such effects are described by diagrams in which two or more virtual photons are exchanged between the electron and the nucleus, with the intermediate nucleus virtually excited. Due to the increased accuracy possible in electron-scattering experiments, such effects are, in principle, observable, possibly showing up as an energy dependence in the charge-density parameters. However, the calculations have indicated that at least for the case of high-energy electrons the dispersion effects are predominantly a function of momentum transfer.[6] This makes it possible to describe the dispersion effect as a small, complex, energy-independent correction to the scattering potential.[7] The situation is not so clear at lower electron energies where the calculations are strongly model dependent. Furthermore, it is difficult to justify neglecting the nuclear excitation energy, which has been done in most of the previous work.

An attempt has been made to interpret the results obtained at low and high energies in terms of dispersion corrections.[10,12-14] However, because of the extremely crude approximations made in the theoretical analysis it is not entirely clear at the present time whether such an interpretation is valid. For this reason it would be of interest to have an experiment the results of which could be unambiguously interpreted as due to dispersive effects. Such an experiment