Probing Ferromagnets with Andreev Reflection

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High transmissivity, ferromagnet-superconductor thin film nanocontacts are studied experimentally. Compared to nonmagnetic metal-superconductor contacts, Andreev reflection is strongly suppressed due to the spin polarization of conduction electrons in the ferromagnet. This effect is used to measure both the transparency of the interface and spin polarization of the *direct* current in the ferromagnet in contrast to the spin polarization of the *tunneling* current previously measured in ferromagnet-insulator-superconductor systems. [S0031-9007(98)07318-9]

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Scattering at the interface between a ferromagnetic and a nonmagnetic metal holds the key to a more complete understanding of many important effects such as giant magnetoresistance (GMR) [1], and spin injection into metals [2]. When the nonmagnetic metal is a superconductor, novel effects such as the nonmonotonic dependence of the superconducting transition temperature on the film thickness of ferromagnet-superconductor (FS) multilayers [3] and nontrivial magnetoresistance of SFS structures have been observed [4]. Recent experiments of Vas'ko *et al.* [5] also indicate the suppression of pairing in a superconductor by the injection of spin-polarized current. To properly understand such effects we need to better understand the nature of scattering and spin dependence of transport at interfaces between dissimilar metals. However, separating out interface from bulk scattering effects is difficult [6], and measurements of the spin polarization of currents through clean ferromagnetic-nonmagnetic (FN) metal interfaces have not yet been accomplished.

In this Letter we show that Andreev reflection can be used to measure both the spin polarization of the current in a ferromagnet and the transmission probabilities of FS and FN interfaces. To isolate the effect of interfacial scattering from bulk scattering, we have made ballistic nanocontacts with the lateral dimension of the clean interface between the two metals small in comparison to other scattering lengths. The suppression of Andreev reflection then allows the determination of the spin polarization P, of the current in the ferromagnet, and the transmission coefficient T, of the interface. By forming these types of nanocontacts (Pb-Co and Pb-Ni), we have made the first detailed measurements of transport at F-S interfaces. For comparison we have made the same measurements on N-S (Pb-Cu) nanocontacts. In this way we have determined both the spin polarization of the ferromagnet, and the transmission coefficients for Pb-Co, Pb-Ni, and Pb-Cu interfaces. Our experiments are sensitive to the direct current in a ferromagnet as opposed to the tunneling current in the pioneering experiments of Tedrow and Meservey [7].

In the case of a normal metal-superconductor (NS) interface, the electron transport below the gap (Δ) of the superconductor is enabled by Andreev reflection [8], whereby every incident electron is reflected as a hole while a Cooper pair carries the current away in the superconductor. For a clean ballistic nanocontact with a transmission coefficient T (when the superconductor is normal), the Andreev reflection probability A is proportional to T^2, because subgap transport involves a transfer of two electrons across the barrier. As pointed out by Blonder, Tinkham, and Klapwijk [9] (BTK), this makes it possible to characterize the interface between the two metals if the voltage dependent conductance $G(V)$ can be determined in both the normal and the superconducting states. de Jong and Beenakker [10] subsequently pointed out that for an FS contact A should also be sensitive to the polarization of the conduction electrons in the ferromagnet, since not every electron from the up-spin conduction band can find a down-spin electron with which to pair, and thus will not be able to enter into the superconductor. This, they argued, should reduce the Andreev probability A to approximately $A(1 - P)$ where P is the polarization of the current in the ferromagnet and is given by $P = (J_\uparrow - J_\downarrow)/(J_\uparrow - J_\downarrow)$, where $J_\uparrow$ and $J_\downarrow$ are the current densities of the up-spin and down-spin electrons, respectively.

We have fabricated bimetallic nanocontacts by thermal evaporation in a vacuum of low 10^{-7} Torr onto both sides of a silicon-nitride membrane containing a tapered nanohole (3–10 nm minimum diameter) [11]. Either Co, Ni, or Cu was evaporated first onto one side of the membrane, and then Pb was deposited onto the other side to form an interface at the narrowest part of the constriction (see Fig. 1). This method has several advantages over the conventional mechanical point-contact technique [12]. Since the samples are made in high vacuum, there is no oxide barrier at the interface. Pb has a much lower heat of condensation than Co, Ni, and Cu and does not dissolve in these metals at room temperature [13] and thus should yield relatively abrupt, clean interfaces. This is important because substantial intermixing at the FS interface could

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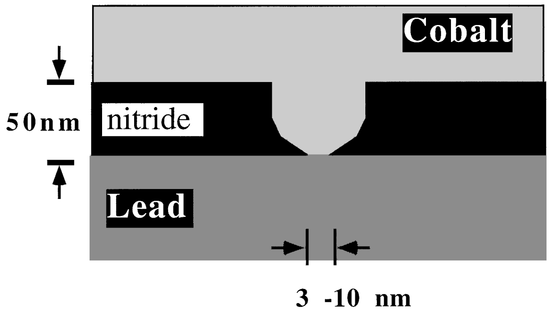

FIG. 1. Schematic of a Pb-Co nanocontact.

suppress the gap and lead to unintended effects. Our Pb-Co and Pb-Ni samples have a T_c of 7.2 K which is the bulk value.

The conductance $G = dI/dV$ was measured as a function of voltage down to 1.4 K using a standard lock-in technique both in the absence of a magnetic field (G_{FS}) and at $2k$ Gauss (G_{FN}). All our experiments are described in terms of the dimensionless function

$$g(V) = \frac{G_{FS}(V) - G_{FN}(V)}{G_{FN}(0)}. \quad (1)$$

$g(V)$ is a measure of the Andreev reflection probability [9]. In the case of a perfectly transmissive contact ($T = 1$) with no spin-polarization of the normal electrons, $g(V) = 1$ for $V < \Delta$ and $g(V) \to 0$ for $V \gg \Delta$. Figure 2(a) shows $g(V)$ for two lead-cobalt samples, with normal state resistances 15.5 and 12.1 Ω. The increase in g below the gap is due to Andreev reflection. However, the maximum value of $g(V)$ is close to 0.15, suggesting that Andreev reflection is significantly reduced for this interface. We have studied twenty-one Pb-Co devices, with normal state (when Pb is normal) resistances varying from 5 to 45 Ω and find that at any voltage $V, g(V)$ varies by less than ± 0.02 from sample to sample. We also studied Pb-Ni devices, where we expect a similar reduction in Andreev reflection and Pb-Cu devices where we do not. Figure 2(b) shows $g(V)$ for a Pb-Ni device (7.3 Ω) at 2.5 K. Here the maximum value of $g(V)s$ is 0.23. For comparison Fig. 2(c) shows $g(V)$ for a Pb-Cu device (6.5 Ω), with $g(V) \approx 0.5$ for eV $< \Delta$ which is higher than both Pb-Ni and Pb-Co.

Andreev reflection can be suppressed by elastic scattering at the interface and, for an FS contact, also by a net spin polarization of the electrons. Another source of a variation in $g(V)$ can be an additional series resistance, e.g., due to scattering in the bulk. We now argue that bulk elastic scattering cannot be responsible for the reduction of $g(V)$. In a nanocontact device the conductance is given by [14], $G = (2e^2/h)(k_F a)^2[T - \Gamma \frac{a}{\ell}]$ where a is the diameter of the hole, ℓ is the bulk scattering length, T is the transmission coefficient of the interface, and $\Gamma \approx 1$. If we assume that the bulk scattering length ℓ is the same for each of these devices, this independence of $g(V)$ on the resistance (5–45 Ω) and hence the diameter

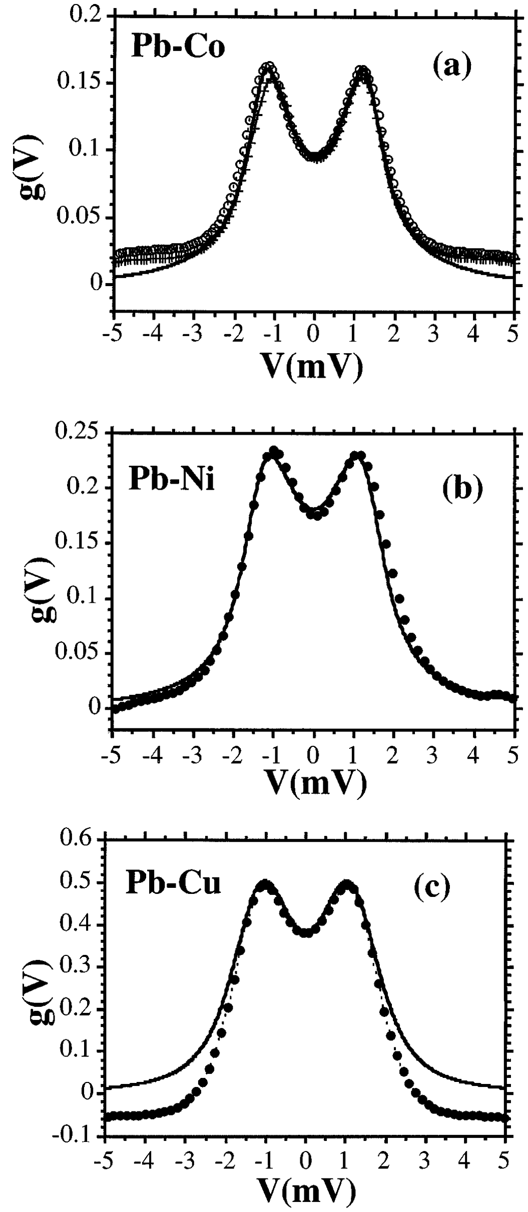

FIG. 2(a). $g(V) = G_{FS}(V) - G_{FN}(V)/G_{FN}(0)$ for two Pb-Co samples with normal-state resistances 15.5 Ω (○) and 12.1 Ω (+) at 1.41 K; (b) $g(V)$ for a Pb-Ni device with normal state resistance of 7.3 Ω at 2.5 K; (c) $g(V)$ for a Pb-Cu device with normal state resistance of 6.5 Ω at 4.2 K. The solid lines are a fit of a modified BTK model (see text) to the data.

of the contact implies that $T\ell \gg a$. This indicates that the nanocontacts' transport properties are indeed dominated by the interface between the metals.

To include the effects of interface scattering and spin polarization, we have generalized the de Jong-Beenakker model [10] to the case of arbitrary scattering at the interface along the lines of BTK theory [9]. For the ferromagnet, we used a Stoner model with momenta $k_\uparrow$ and $k_\downarrow$ for up and down spin electrons given by $k_\uparrow^2 = (2m/\hbar^2)E_F$ and $k_\downarrow^2 = (2m/\hbar^2)(E_F - 2H)$ where E_F is the Fermi Energy and H is the exchange energy.

The transport in the superconductor is described by the Bogoliubov–de Gennes equation. For simplicity, we have assumed that the Fermi wave vector for Pb, $k_F = k_\uparrow$ and that the effective masses are equal in S and F. A difference in effective masses or Fermi wave vectors will result in elastic scattering at the interface. To take this and any additional impurity or defect scattering at the interface into account, we included a delta function potential $V(\vec{r}) = \lambda\delta(z)$ where λ is spin independent. The strength of scattering by the delta function is measured by the dimensionless parameter $Z = (m\lambda/\hbar^2 k_F)$. For the superconducting gap, we have used an abrupt approximation, $\Delta(\vec{r}) = \Delta\Theta(z)$ where Θ is the unit step function. This approximation is justified since the contact-size (3–10 nm) is smaller than the coherence-length of lead (83 nm) [15]. In order to calculate the conductance in the FS case, we used the linear response formula due to Takane and Ebisawa [16]

$$G_{\mathrm{FS}} = \frac{e^2}{h}\sum_{a,b}[\delta_{ab} - |r_{be,ae}|^2 + |r_{bh,ae}|^2], \qquad (2)$$

where $r_{bh,ae}$ is the Andreev reflection coefficient and $r_{be,ae}$ is the normal reflection coefficient. The summation is over all the modes of the point contact and over spin indices a and b. The calculation then follows very closely to that of Ref. [9] and [10], performed for a three-dimensional (3D) geometry. In order to extract G_{FN}, we just set $\Delta = 0$. We then calculate $g(V)$ as a function of V numerically. The temperature dependence is assumed to arise only from the thermal smearing of the Fermi surface and the temperature dependence of a gap. In the case of a nanocontact, the number of modes with spin up and down are given by $N_{\uparrow(\downarrow)} = k_{\uparrow(\downarrow)}^2 A/4\pi$ where A is the area of the contact. Since $J_{\uparrow(\downarrow)} \propto N_{\uparrow(\downarrow)}$ in the ballistic limit, the polarization P can also be written as $P = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow) = (k_\uparrow^2 + k_\downarrow^2)/(k_\uparrow^2 + k_\downarrow^2)$. This modified BTK model has only two independent parameters, P and Z. In the case of an SN contact, this reduces to a three-dimensional BTK model which has only one parameter, Z, since $P = 0$. The details of the calculation will be presented elsewhere [17]. The transmission coefficients $T_\uparrow$ and $T_\downarrow$ for the up and down spins and the average transmission coefficient T can then be determined in terms of P and Z.

The solid lines in Figs. 2(a), 2(b), and 2(c) are the best fits of this model to the data. We used $\Delta(0) = 1.34 \pm 0.01$ mV for all our devices, which is consistent with the bulk gap of lead [15] ($\Delta(0) = 1.36$ mV). Similar quality fits are obtained for the data sets taken on a given sample at several temperatures below the T_c of lead, yielding the same values of P and Z in each case. These fits are obtained without the necessity of invoking a "gap-smearing" parameter to explain a broadening of the transition from the low V to high V behavior. For Pb-Co samples, we obtain $P = 0.371 \pm 0.002$ and $T = 0.666 \pm 0.002$. For the Pb-Ni sample [Fig. 2(b)], $P = 0.327 \pm 0.002$ and $T = 0.682 \pm 0.002$. The Pb-Cu data [Fig. 2(c)] were best fitted with $P = 0.000 \pm 0.002$ and $T = 0.794 \pm 0.002$. Table I summarizes our measurements of P and T for a number of samples. For each of the different contact types, i.e., Pb-Co, Pb-Ni, and Pb-Cu, the sample-to-sample variation δP is less than ± 0.02 while δT is less than ± 0.03. This small variation is attributable to sample-to-sample differences in the nature of the interface, e.g., interfaces between different crystal faces of lead and cobalt may give rise to different scattering rates at the interface.

While our 3D modified BTK model fits the data very well for $V < 3$ mV, it deviates from the measured $g(V)$ for large biases. This deviation is most visible in the case of Pb-Cu [Fig. 2(c)]. The deviations are either due to the strong coupling effects in lead or the high current densities achieved in our samples (10^9 A/cm^2 at 10 mV for a 10 Ω sample) which can lead to partial gap suppression [18]. Figure 3(a) shows the point-contact spectra (dR/dV vs V) for the same Pb-Cu sample both in the normal and the superconducting state. The phonon peaks of lead [19] are very clearly visible in both, the peaks being shifted to higher energies in the superconducting case. Figure 3(b) shows dR/dV vs V for a Pb-Co sample. Our ability to see lead phonon peaks in the point-contact spectra of all our samples also gives us confidence in the relative cleanliness of our devices (negligible bulk scattering).

Our experiments show that the reduction of Andreev scattering at FS interfaces can be used as a probe to study transport in the ferromagnet. Of course our analysis is based on a very simple model. Cobalt and nickel are not Stoner ferromagnets; their band structure is quite complicated and a complete theory should take the s-band electrons, the d-band electrons, and their hybridization into account. While the remarkable success in fitting the data suggests that the simple model captures the essential physics, certainly a more complete theory taking the band structure of Pb and Co (or Ni) into account is needed to fully establish its validity.

TABLE I. Spin polarization and transmission coefficients of direct FS interface currents as measured by Andreev reflection, and in comparison to previously measured polarization of FIS tunnel currents.

Metal	P (tunneling) [7]	P (Andreev)	$T_\uparrow$ (with Pb)	$T_\downarrow$ (with Pb)	T (average)
Co	$+0.35 \pm 0.03$	0.37 ± 0.02	0.95 ± 0.01	0.38 ± 0.01	0.67 ± 0.03
Ni	$+0.23 \pm 0.03$	0.32 ± 0.02	0.94 ± 0.01	0.43 ± 0.01	0.68 ± 0.03
Cu		0.00 ± 0.01	0.79 ± 0.01	0.79 ± 0.01	0.79 ± 0.03

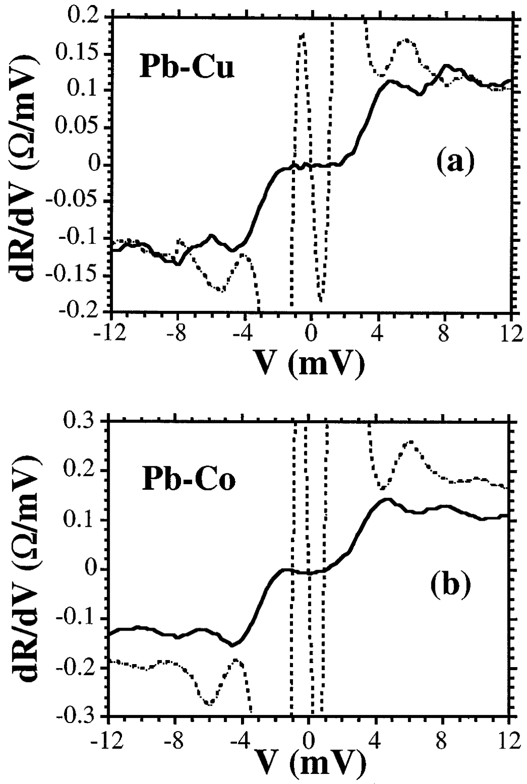

FIG. 3(a). dR/dV for the Pb-Cu sample of Fig. 2(c) at 4.2 K, when the Pb is superconducting (dashed line) and when the Pb is normal (solid line); (b) dR/dV for a Pb-Co sample (15.5 Ω) of Fig. 2(a) at 1.4 K, when the Pb is superconducting (dashed line) and when the Pb is normal (solid line).

In the past Meservey and Tedrow [7] used ferromagnet-insulator-superconductor junctions to study the spin polarization of tunneling electrons. As shown in Table I, the polarization measured by them differs from our results. Their results for Co and Ni are +35% and +23%, respectively, while ours are 37% and 32%, respectively. As pointed out by Stearns [20], the tunneling measurement is sensitive to the density of states of the tunneling electrons (the itinerant d electrons). Recent work also suggests that the tunneling experiments are sensitive to the nature of electronic states within the first few monolayers of the ferromagnetic electrode [21]. Our experiments, on the other hand, are performed in the high-transparency limit and hence are sensitive to the polarization of the incident electrons from within a volume of radius 3–10 nm (the size of the nanocontact). The independence of the measured polarization on contact size also indicates that we are sensitive to the spin polarization of the bulk current and are not just measuring an interface effect. In terms of a Stearns model (the modified BTK model in the low transparency limit), we measure the quantity $(k_\uparrow^2 - k_\downarrow^2)/(k_\uparrow^2 + k_\downarrow^2)$ while the tunneling experiments measure the quantity $(k_\uparrow - k_\downarrow)/(k_\uparrow + k_\downarrow)$. Although we can measure the magnitude of the polarization, our experiments are not sensitive to its sign.

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Note added in proof.—A similar, Andreev reflection study of magnetic-superconducting interfaces, carried out with mechanical point contacts, has very recently been reported by Soulen *et al.* [22].

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