Proximity-induced density-of-states oscillations in a superconductor/strong-ferromagnet system

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We have measured the evolution of the tunneling density of states (DOS) in superconductor/ferromagnet (S/F) bilayers with increasing F-layer thickness, where F in our experiment is the strong ferromagnet Ni. As a function of increasing Ni thickness, we detect multiple oscillations in the DOS at the Fermi energy from differential conductance measurements. The features in the DOS associated with the proximity effect change from normal to inverted twice as the Ni thickness increases from 1 to 5 nm.

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Hybrid systems consisting of superconducting (S) and ferromagnetic (F) materials have attracted substantial attention due to their interesting properties and potential for applications.1 The superconducting proximity effect in such systems is normally short ranged, due to the large exchange energy in the F material. When a Cooper pair crosses the S/F interface, the spin-up and spin-down electrons enter into different spin bands, and the center-of-mass coordinate picks up an oscillatory factor.2 The physical manifestations of this oscillation can be observed as a series of transitions between “0” and “π” states in S/F/S Josephson junctions as a function of increasing F-layer thickness,3,4 or as oscillations between “normal” and “inverted” proximity features in the tunneling density of states (DOS) of S/F/I/N tunnel junctions.5 (Here I is an insulator and N is a normal metal.)

With substantial experimental effort in S/F/S Josephson junctions, the 0-π transition has been confirmed by many experimental groups.3,4,6–12 What is surprising is that, unlike in the Josephson geometry, the oscillatory behavior of the DOS in S/F/I/N structures has been observed convincingly only once, as a single normal-inverted transition in samples with a weakly ferromagnetic alloy for F.5 In experiments using strong ferromagnets, the results have been less clear.13,14 At this time, to the best of our knowledge, there is no definitive experimental answer to the question of whether the DOS in S/F/I/N structures oscillates as a function of F-layer thickness when F is a strong ferromagnet. The primary goal of this Rapid Communication is to answer this question.

Our S/F/I/N tunnel junctions are fabricated by thermal evaporation and sputtering, using a series of mechanical masks (Fig. 1). We first evaporate a 150-nm strip of Al (N), then we immediately backfill the chamber with 300 Torr of a 10% O₂, 90% Ar mixture. Exposing the freshly evaporated Al to the O₂ quickly (while the Al is still hot) provides good conditions for oxide growth. The O₂ exposure continues for ≈12 h to produce a robust layer of Al₂O₃ (I) on the Al surface. Next, we change masks and evaporate a thick layer (200 nm) of SiO₂ to define the junction geometry. When using mechanical masks for the top leads, shadow effects can cause unwanted regions at the edges of the junctions where the Ni thickness is not well defined. The SiO₂ is in place to avoid the appearance of edge effects in our data. Finally, we sputter a Cu(5 nm)/Ni(5 nm)/Cu(10 nm)/Nb(150 nm)/Au(15 nm) multilayer. The choice of Ni and Cu is beneficial because our Ni has a relatively long spin-diffusion length 21 ± 2 nm, as compared to our maximum Ni thickness, along with a low resistivity ρNi = 33 ± 3 Ω m.15 Ni also provides weak asymmetry of spin-dependent scattering in the bulk and at Ni/Cu interfaces, and a low average Ni/Cu interface resistance.15 These attributes should simplify theoretical analysis. The Cu layer adjacent to the Al₂O₃ has been found to increase the effectiveness of the tunnel barrier. The Au deters oxidation of the Nb layer. Throughout the process we must break vacuum, but the consistency and reproducibility of our results suggest that this has little effect on the quality of our junctions. Due to a high level of oxygenation of our Al, its Tc ≈ 1.9 K. Thus, we performed our measurements at 2.1 K, with the Al in the normal state.

Using a four-terminal lock-in technique, we measure the voltage-dependent differential conductance of our samples dI/dV(V), which approximates the DOS of our S/F bilayer. (The true DOS would be attained if measured at T = 0 K when there is no zero-bias anomaly; we will use “DOS” to refer to our nonideal differential conductance measurements.) We normalize dI/dV by multiplying by the normal-state resistance RN, determined from the inverse of the differential conductance at large bias voltages. Our junction area-resistance products, ARN ≈ 2 × 10⁵ Ωμm², are substantially higher than those of

![Figure 1](image_url)
FIG. 2. (Color online) The blue dotted-dashed curve shows
\( dI/dV \) vs \( V \) for a \( d_{\text{Ni}} = 4.5 \text{ nm} \) junction, where the data are slightly
smoothed and normalized. The blue dotted line is a linear fit to large
bias voltage for the previous blue dotted-dashed curve. The red solid
curve shows the same \( dI/dV \) data after subtracting off the linear
background feature. Inset: Normalized \( dI/dV \) curve shows the same
bias voltage for the previous blue dotted-dashed curve. The red solid
smoothed and normalized. The blue dotted line is a linear fit to large
vs \( dI/dV \)

other groups using passive oxidation,\textsuperscript{16,17} but much lower than
those produced by specialized oxidation techniques.\textsuperscript{5,14} From
the data, it can be seen that this has little to no effect on our
measurements.

Our \( dI/dV \) data exhibit a small negatively sloped, linear
background, which appears to be a component of the normal
state of our junctions. Figure 2 shows a plot of \( dI/dV \) from
our 4.5-nm sample before and after subtraction of the linear
background, along with the linear fit to \( dI/dV \) for \( |V| \gg \Delta \),
where \( \Delta \approx 1.4 \text{ mV} \) is the gap parameter for Nb. There is
another normal-state characteristic which we do not correct
for in our measurements. A slight V-shaped feature centered
at 0 V becomes apparent at large Ni thicknesses. (It is visible
on the red solid curve of Fig. 2 at \( |V| > 2.5 \text{ mV} \)) We did not
have a magnet on our apparatus to force the top Nb layer
into the normal state; nevertheless, we emphasize that the
slight negative slope and V-shaped feature in our background
are both much smaller than background features observed in
S/F/I/N tunnel junctions measured by other groups.\textsuperscript{18,19}

The inset of Fig. 2 shows a plot of \( dI/dV \) for an N/I/S
junction with a 15-nm Cu buffer layer between S and I. All data were
taken at 2.1 K.

FIG. 3. (Color online) Normalized differential conductance vs
voltage for S/F/I/N junctions with several different Ni thicknesses.
The Ni thickness \( d_{\text{Ni}} \) is labeled under each curve at the right-hand side.
As one moves up the figure, each panel has an increasing expansion
of the vertical scale where only the lowest trace is normalized to 1
and the others are displaced upward for clarity. All data are taken at
\( T = 2.1 \text{ K} \).

Ni(\( d_{\text{Ni}} \))/Cu(5 nm) multilayers, for \( 1 \text{ nm} < d_{\text{Ni}} < 5 \text{ nm} \). The
data show an extrapolated nonmagnetic "dead-layer" Ni
thickness of 0.25 ± 0.05 nm at each Ni/Cu interface. Thus
we show data only for \( d_{\text{Ni}} \geq 1 \text{ nm} \). In the 1-nm sample one
clearly sees the Nb gap, but with a significant suppression of
the bulk Nb features due to the proximity effect in the strong
ferromagnet. As we increase the Ni thickness, the zero-bias
dip in the DOS quickly decreases in magnitude. At \( d_{\text{Ni}} = 1.5 \text{ nm} \), we observe the first sign of an inversion in the differ-
ential conductance at zero bias, followed by a maximum inversion
at \( d_{\text{Ni}} = 1.75 \text{ nm} \). The features in \( dI/dV \) at \( |V| = \Delta \approx 1.4 \text{ mV} \) have also been inverted but occur at the Nb gap voltage
in all the samples measured. The inversion cycles quickly and
by \( d_{\text{Ni}} = 2.5 \text{ nm} \), the samples return to the noninverted regime.
The dip in the DOS reaches its maximum at \( d_{\text{Ni}} = 3 \text{ nm} \), then a
second inversion occurs starting at \( d_{\text{Ni}} \approx 4.25 \text{ nm} \). (The second
inversion is more apparent in the expanded scale of Fig. 2.)
This second inverted state looks as though it might extend past
5 nm.

The oscillation is best illustrated in Fig. 4 with a plot of the
normalized differential conductance at both \( V = 0 \) and \( V = 1.4 \text{ mV} \) versus Ni thickness. The oscillation period is irregular;
With a finite ferromagnet, the oscillation is regained even in the pure ballistic limit due to specular\textsuperscript{23} or diffuse\textsuperscript{24} scattering from the film boundaries. In constrast, Sun et al. solved the Bogoliubov–de Gennes equations and claimed that oscillations should occur in either a finite or semi-infinite F layer.\textsuperscript{25}

Because our observed DOS variations are not periodic in $d_{\text{Ni}}$, we do not attempt to fit our data with Usadel theory. We believe that our samples most closely match the assumptions in the papers by Zareyan et al.\textsuperscript{24} From earlier work on S/Ni systems, we expect that $E_{\text{ex}} \approx 100 \text{ mV,}$\textsuperscript{7,8,11} and $v_F = 2.8 \times 10^5 \text{ m/s}$ for Ni,\textsuperscript{26} while $\Delta = 1.5 \text{ mV}$ for Nb. From our measured Ni resistivity $\rho_{\text{Ni}} = 33 \text{ n}\Omega \text{ m}$, we deduce $l_{\text{Ni}} = 45 \text{ nm}$. This puts our samples in the “intermediate” regime with $E_{\text{ex}} \gg \hbar/\tau_F \gg \Delta$. By using Cu buffer layers next to the Ni, we limit the scattering events in our junctions considerably. As stated earlier, we find a spin diffusion length of $21 \pm 2 \text{ nm}$, low overall spin-scattering asymmetry, and very low Cu/Ni interface specific resistance in our multilayers: $AR_{\text{CuNi}} = 0.18 \pm 0.03 \text{ \Omega m}^2.\textsuperscript{15}$ This low interface resistance corresponds to a probability of scattering of only $\sim 15\%$ at each Ni/Cu interface.\textsuperscript{25} The Cu/Nb interface is “rough” in the sense that there is significant diffusive scattering at this interface, as determined from its measured interface specific resistance of $AR_{\text{CuNb}} = 1.1 \pm 0.15 \text{ \Omega m}^2.\textsuperscript{29}$ This value of $AR_{\text{CuNb}}$ is larger than the total $AR = 2AR_{\text{CuNi}} + \rho_{\text{Ni}}l_{\text{Ni}} + \rho_{\text{Cu}}d_{\text{Cu}} = 0.54$ and 0.67 $\Omega m^2$ of the Cu/Ni/Cu region for $d_{\text{Ni}} = 1$ and 5 nm, respectively. We also expect there to be diffuse scattering at the Cu/Al$_2$O$_3$ (tunneling) interface.

Plots of the energy dependence of the DOS shown in the papers by Zareyan et al.\textsuperscript{24} agree qualitatively with our data. Performing a quantitative fit of the theory to our data, however, is problematic. The theory predicts that the first $0-\pi$ transition should occur at very small $d_{\text{Ni}}$, a flaw that may be correctable by adding spin-dependent interfacial phase shifts to the theory.\textsuperscript{30} (One could also argue that, because of the 0.25-nm dead layers at the two Cu/Ni interfaces, one should subtract 0.5 nm from our nominal sample thickness before fitting to the Zareyan theory, but that is not nearly enough to bring theory into agreement with experiment.) The theory also predicts large oscillations in the normalized DOS (i.e., large deviations from 1) at zero energy—much larger than what we observe in the experiment. The amplitude of the theoretical oscillations can be reduced by assuming a very small transparency $T$ of the Nb/Cu interface; such an assumption, however, is incompatible with the measured boundary resistance $AR_{\text{CuNb}} = 1.1 \pm 0.15 \text{ \Omega m}^2$, which implies that $T \approx 0.5.\textsuperscript{27}$ Strong spin-flip scattering would also reduce the amplitude of the DOS variations; the long measured spin memory length in our Ni films, however, precludes that explanation for these samples. One could assume that the variation in F-layer thickness over the junction area is very large, thereby smearing out the oscillations; we believe that such an assumption is unrealistic.

A previous measurement of the Nb/Ni system\textsuperscript{14} did detect signs of one $0-\pi$ transition, but the data contained additional low-energy features, which were later interpreted as signs of $p$-wave spin-triplet pairs.\textsuperscript{24} We do not observe such low-energy features in our data.

In conclusion, we have observed multiple oscillations in the DOS of S/F bilayers as a function of F-layer thickness, where F is a strong ferromagnet (Ni). The oscillations can be

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**FIG. 4.** (Color online) Black squares represent the normalized $dI/dV$ at $V = 0$ for several Ni thicknesses. Red circles represent the normalized $dI/dV$ at $V = 1.4 \text{ mV}$. The black and red lines link the average values of each thickness for $V = 0$ and $V = 1.4 \text{ mV}$, respectively. Inset: Data for $d_{\text{Ni}} \geq 4 \text{ nm}$ on an expanded vertical scale.

The first inverted region persists for $d_{\text{Ni}} \approx 1.3-2.4 \text{ nm}$, while the second noninverted region lasts much longer, for $d_{\text{Ni}} \approx 2.4-4.3 \text{ nm}$. One can see in the inset that the oscillation occurs about a value $dI/dV \approx 0.9996$. This offset in the oscillation is due to the aforementioned, weak $V$-shaped zero-bias anomaly.

The transitions between normal and inverted DOS have been predicted to occur at F-layer thicknesses exactly half of where $0-\pi$ transitions occur in S/F/S Josephson junctions.\textsuperscript{1}

After reviewing the literature on S/Ni/S junctions, we see that our junctions are most similar to those of Blum et al.\textsuperscript{7} and Shelukhin et al.,\textsuperscript{9} where a Cu buffer layer is placed on each side of the Ni layer. In contrast, Robinson et al.\textsuperscript{11} have Nb in direct contact with Ni, with a Ni “dead-layer” thickness of $\approx 0.8 \text{ nm}$ at each Ni/Nb interface in comparison to our Ni dead-layer thickness of only $0.25 \pm 0.05 \text{ nm}$ at each Cu/Ni interface. The Ni thickness at which the first $0-\pi$ transition is observed by these groups varies quite a bit — $2.6$, $1.7$ nm (extrapolated value), and $3.8$ nm for Refs. 7,9 and 11, respectively. Since our first $0-\pi$- and $\pi$-0 transitions occur at 1.3 and 2.5 nm, respectively, we would expect the first two transitions in S/Ni/S junctions to occur at 2.6 and 5 nm, in reasonable agreement with the values observed by Blum et al.\textsuperscript{7}

Theoretical calculations of the DOS in S/F bilayers cover several regimes, defined by the relative strengths of the ferromagnetic exchange $E_{\text{ex}}$, the superconducting gap parameter $\Delta$, and the impurity scattering $\hbar/\tau_F$, as well as the relative sizes of $d_F$ and the mean free path $l_e = v_F/\tau_e$. In the dirty limit, the Usadel equations provide a clearcut prediction of oscillation of the tunneling DOS, with period of order $\xi_F = (\hbar D/E_{\text{ex}})^{1/2}$, where $D = v_F l_e/3$.\textsuperscript{26} In the clean limit, the predictions are less straightforward. Solving the Eilenberger equation in the ballistic limit leads to the conclusion that the DOS does not oscillate in a semi-infinite ferromagnet.\textsuperscript{21,22} Oscillations are predicted to occur, however, in the presence of weak disorder, with an amplitude proportional to $\hbar/ \eta_{\text{ex}} \tau_F$.\textsuperscript{21,22}
described qualitatively, but not quantitatively, by the theory of Zareyan et al. Discrepancies between theory and experiment may be due to the extra Cu layers in our samples, which are not present in the theoretical calculation, or to the absence of spin-dependent interfacial phase shifts in the theory.

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