Competition between proximity-induced superconductivity and pair breaking: Ag sandwiched between Nb and Fe

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The magnetization of superconductor and/or normal-metal (Nb/Ag) double layers is investigated in dependence on temperature T and magnetic field B. Screening currents in the normal-metal induced by the proximity effect give rise to a diamagnetic transition in a weak magnetic field at a temperature T_b. The phase transition is suppressed when Fe is in direct contact with Ag. Surprisingly, the diamagnetic signal of Ag is recovered for small Ag film thickness d_{Ag}. These findings are qualitatively explained by the competition in the Ag layer between proximity-induced superconductivity by the Nb layer and pair breaking by the ferromagnetic Fe layer.

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Heterostructures with superconducting elements, exploiting the macroscopic quantum coherence of the superconductive wave function, have become of increasing interest in recent years. The integration of superconducting materials (S) in electronics leads to interface contacts with normal metals (N) and also with ferromagnets (F) playing an important role in spintronics. The "proximity effect," i.e., the penetration of the superconductive pair amplitude of S into the adjacent N or F metal, is mediated by the microscopic process of Andreev reflection. At the interface, incident electrons and retroreflected holes form correlated but mutually independent Andreev pairs over a coherence length $\xi_N(T)$. The relevant length scales for a S/N system, where S is considered as infinitely thick, are the electron mean free path l_N, the thickness d_N, and $\xi_N(T)$.

Early investigations of the proximity effect in thin S/N double layers have focused on measurements of their transition temperatures. These experiments could be well described by the quasiclassical approach[1] in the "dirty limit," i.e., for $l_N \ll (d_N, \xi_N)$. The magnetic properties, e.g., the diamagnetic response, of S/N systems in an external magnetic field B have been investigated theoretically and experimentally.[2–7] For commercial sheets and wires, the data clearly deviate from the dirty-limit behavior.[4,7,8] In particular, data obtained on well-annealed coaxial Nb/Ag wires[9] are better described by taking into account the large l_N, i.e., considering the "ballistic regime." Besides, a paramagnetic reentrance behavior was observed at low temperatures.[10,11] The dirty or clean limits are characterized by different temperature dependences of ξ_N, i.e., $\xi_N^d(T)=(\hbar v_F l_N/6\pi k_B T)^{1/2}$ and $\xi_N^c(T)=\hbar v_F/2\pi k_B T$, respectively.[3] Measurements of the magnetization $M(T,B)$ allow the determination of the critical field $B_b(T)$ and the characteristic Andreev temperature $T_A=\hbar v_F/2\pi k_B d_N$ of the N layer, defined by $\xi_N(T_A)=d_N$. B_b and T_A can be considered as parameters reflecting the stability of the proximity-induced superconductivity (PIS) in N.

While previous experiments focused on commercial S/N sheets and wires, little is known about the magnetic response of clean S/N double layers with thickness d_N in the submicrometer range in contact with a ferromagnetic layer. In S/F contacts, the pair amplitude in F decays exponentially with distance from the interface, superposed with a periodic modulation due to the ferromagnetic exchange interaction I_{ex}.[12–14] In particular, for strong ferromagnets with $I_{ex}\tau > h$ (τ, elastic-scattering time), the pair-condensate amplitude decays on a length scale of the order of the electron mean free path l_F in F.[15] Cladding of micrometer-thick coaxial Nb/Cu wires with Fe results in a strong depression of the proximity effect.[5] In S/N systems the diamagnetic transition of the N layer shifts to higher temperatures with decreasing d_N indicating an enhanced stability of PIS, whereas the cladding of the outer N surface by a ferromagnetic metal gives rise to additional pair breaking. Hence, in $S/N/F$ systems with appropriate N layer thickness, a concurrent influence of PIS and pair breaking by F on N should be observed, as will be demonstrated in this paper. We report on magnetization measurement on high-quality Nb/Ag double layers with $d_{Ag} < 1$ μm and with a finite but long l_{Ag} of the order of d_{Ag}. We observe a diamagnetic phase transition of the Ag layer at a temperature T_b depending on B and d_{Ag} from which we determine B_b and T_A. In particular, we focus on Nb/Ag/Fe triple layers and find that for thick Ag layers, the diamagnetic screening in Ag is suppressed by the proximity of Fe. On the other hand, for small d_{Ag}, the diamagnetic signal in Ag reappears due to a delicate balance between PIS in Ag and pair breaking in contact with the ferromagnetic Fe.

The samples were epitaxially grown by electron-beam evaporation in ultrahigh vacuum (base pressure 1×10^{-10} mbar). Nb was deposited with a fixed thickness of 200 nm on $(11\bar{2}0)$-oriented sapphire substrates (width w =0.7–5 mm, length L=4–8 mm) at a substrate temperature T_S=920 K and covered by 35–550 nm Ag at T_S=470 K. The crystalline quality was checked by *in situ* reflection high-energy electron diffraction (RHEED) and *ex situ* by x-ray diffraction, indicating growth directions of Ag[111]‖Nb[110] with mosaic spreads of 0.65° and 0.78° for Nb and Ag, respectively. 40 nm Fe were deposited onto the Ag layer at room temperature without further annealing. For some films, a SiO_2 barrier was introduced between the Ag and Fe layers. The observed RHEED streaks indicate a smooth growth of Fe along [110] on Ag(111). The in-plane magnetization of the Fe layer was checked by vibrating-sample magnetometry yielding a Fe moment of $m_{Fe}=(2.1\pm0.1)\mu_B$ in agreement with that of bulk Fe, see Fig. 1 (inset). A coercivity of

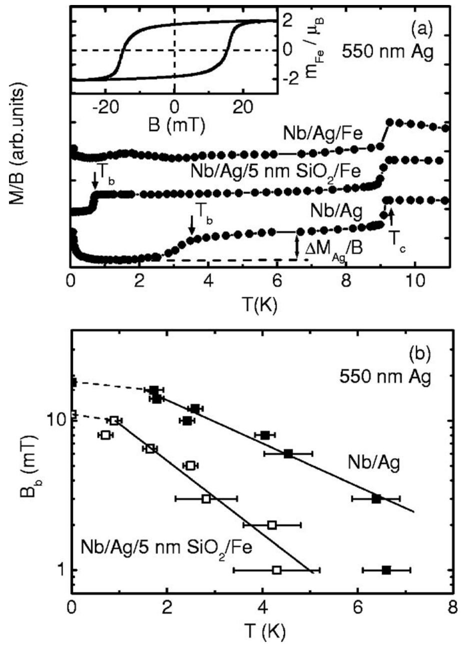

FIG. 1. (a) $M(T)/B$ for samples with $d_{\mathrm{Ag}}=550$ nm and $d_{\mathrm{Fe}}=40$ nm in a field of $B=8$ mT. T_c and T_b indicate the diamagnetic transition of the Nb and Ag layers, respectively. Inset shows the magnetization curve of the Nb/Ag/Fe sample taken at $T=10$ K. (b) Semilogarithmic plot of $B_b(T)$. Solid lines indicate a behavior $\ln B_b \propto -T/T_A$. Dashed lines serve as guides to the eye toward $B_b(0)$.

≈ 15 mT was obtained for all Nb/Ag/Fe triple layers. The samples were, finally, covered by 5 nm SiO_2 or Si to protect them from oxidation in ambient air.

Four-point measurements of the residual resistivity ρ_{Nb} and of the upper critical field on a single 200 nm Nb film yield an electron mean free path $l_{\mathrm{Nb}}=27$ nm using[16] $\rho_{\mathrm{Nb}} l_{\mathrm{Nb}} = 3.75 \times 10^{-16}\ \Omega\,\mathrm{cm}^2$ and an upper critical field $B_{c2}(2\ \mathrm{K}) = 0.65$ T. The following superconducting parameters were obtained from standard BCS relations and material parameters of bulk Nb:[16,17] coherence length $\xi_{\mathrm{Nb}}=19$ nm; penetration depth $\lambda_{\mathrm{Nb}} \approx 42\ \mathrm{nm} < d_{\mathrm{Nb}}$; lower critical field $B_{c1}(2\ \mathrm{K}) = 64$ mT. Since $d_{\mathrm{Nb}} \gg (\lambda_{\mathrm{Nb}}, \xi_{\mathrm{Nb}})$, the S layer can be regarded as infinitely thick. From resistance measurements on several Nb/Ag double layers with the current in plane, a lower limit $l_{\mathrm{Ag}}^{min} \approx d_{\mathrm{Ag}}$ was estimated in comparison with the single Nb film. The magnetization M at constant B was measured as a function of T in a coaxial dB_z/dz gradiometer coupled to a superconducting quantum interference device (SQUID). After cooling down in zero magnetic field to temperatures of about 60 mK, magnetization signals $M(T)$ were recorded during warm-up. A homogeneous and stable magnetic field was applied nearly parallel to the sample surface by magnetic-flux enclosure of an external magnetic field, using a superconducting Pb or NbTi/Nb/Cu cylinder[18] surrounding the sample holder. Although care was taken to measure all samples at similar positions in the gradiometer, an absolute measurement of the magnetization was not possible due to the strong dependence of the Nb signal from a possible tilt angle between film plane and field, which could be controlled only to $\pm 0.5°$.[19] Therefore, the M/B data are given in arbitrary units. After each $M(T)$ cycle at constant field, the superconducting cylinder was heated to well above its transition temperature for complete expulsion of trapped magnetic flux.

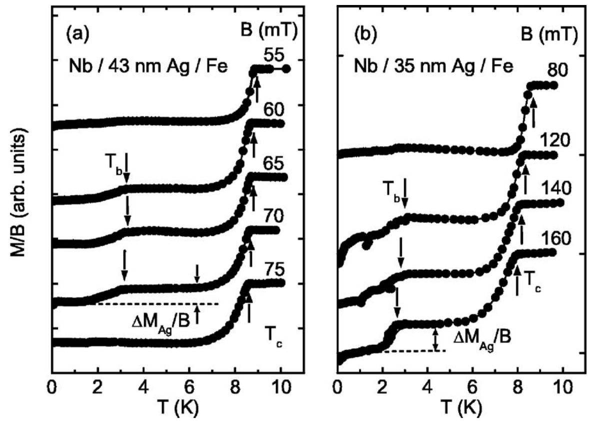

FIG. 2. $M(T)/B$ of Nb/Ag/Fe samples with $d_{\mathrm{Fe}}=40$ nm for different d_{Ag} and applied fields B. See text for details.

Figure 1(a) shows $M(T)/B$ of a Nb/Ag double layer with $d_{\mathrm{Ag}}=550$ nm. The sharp diamagnetic signal at $T_c \approx 9.1$ K is due to the superconducting transition of the Nb layer. At a lower temperature T_b, a further diamagnetic transition of height $\Delta M_{\mathrm{Ag}}/B$ occurs which is attributed to the proximity-induced diamagnetic screening currents in the Ag layer. [The paramagnetic signal at $T < 300$ mK is due to the oxidized surface of the copper sample holder. This signal is absent in subsequent measurements made with a silver sample holder, see, e.g., Fig. 1(a) (middle curve) or Fig. 2]. We mention that with increasing magnetic field, the transition at T_b becomes sharper and shifts toward lower temperatures. This sharpening is observed for all samples studied. For the present samples with small d_{Ag}, the transition occurs at a few Kelvin and is, therefore, broadened by the temperature-dependent penetration depth $\lambda_{\mathrm{Ag}}(T)$.[20] The sharpening of the transition in increasing fields is also inferred from the calculated nonlinear susceptibility of S/N double layers.[3]

The discrete energy levels of Andreev bound states in N are given by[2]

$$E_n = \frac{\hbar v_x}{4 d_N}\left[(2n+1)\pi - \frac{2\pi}{\phi_0}\oint \vec{A}(\vec{r})d\vec{r}\right] \qquad (1)$$

(v_x, component of Fermi velocity perpendicular to the interface; $\vec{A}$, vector potential; ϕ_0, superconducting flux quantum). For $T \ll T_A$ and $B \ll B_b$, each Andreev pair contributes coherently to the macroscopic screening current because only the lowest Andreev level is occupied. At temperatures above T_A, the coherence is continuously destroyed by inelastic-scattering events and thermal excitations, changing the population of the Andreev states. In addition, in a magnetic field, the Andreev pairs acquire an additional phase shift by the vector potential according to Eq. (1) leading to a randomization of the Andreev currents and the destruction of the coherence at the critical field $B_b(T)$.

The $B_b(T)$ phase diagram in Fig. 1(b) is in qualitative agreement with thermodynamic calculations for the clean limit, as previously reported for coaxial Nb/Ag wires.[20,21] The $B_b(T)$ dependence for temperatures $T \geqslant T_A$, i.e., when $\xi_{\mathrm{Ag}} < d_{\mathrm{Ag}}$, nicely obeys a behavior $\ln B_b \propto -T/T_A$, in contrast to the dirty limit where $\ln B_b \propto -\sqrt{T}$. For $d_{\mathrm{Ag}} = 550$ nm, the $B_b(T)$ behavior can be described only partly by the dirty limit by using a very long $l_{\mathrm{Ag}} = 105$ nm violating the condition $l_{\mathrm{Ag}} \ll \xi_{\mathrm{Ag}}$. The characteristic temperature T_A obtained from the slope of $B_b(T)$ (Ref. 20) is $T_A = 3.02$ K, in very good agreement with the theoretical value of 3.06 K estimated from $T_A = 1680\ \mathrm{K\,nm}/d_{\mathrm{Ag}}$. While our data show semiquantitative agreement with the thermodynamic phase diagram, we should point out that we actually measure the superheated field $B_{sh}(T)$, which may be somewhat larger than the thermodynamic critical field for PIS in Ag. In summary, the Nb/Ag data are quite well described by theory for the clean limit, although the smooth transitions are not expected for first-order transitions. Hence, the films should be classified to fall in the ballistic regime.

In what follows, we focus on the pair-breaking effect by a ferromagnetic Fe layer on the PIS. Deposition of a 40 nm thick Fe layer directly onto Ag with $d_{\mathrm{Ag}} = 550$ nm suppresses the diamagnetic signal down to below the lowest temperature of ≈ 60 mK, see Fig. 1(a), even in a weak external field of 0.5 mT (not shown). The small signal variations are caused by thermal instabilities of the SQUID system during that measurement. As already mentioned, in S/F contacts, the pair amplitude in F decays exponentially with distance from the interface. This is also expected for the Andreev pairs penetrating into the ferromagnetic layer from the "normal conducting" N layer in a $S/N/F$ structure. The Andreev pairs experience an additional phase shift in Fe, which destroys the phase coherence in Ag. This has also been reported earlier for coaxial (165 μm Nb/27 μm Cu/0.09 μm Fe) wires.[5] An alternative explanation might be the presence of a magnetic stray field from domain walls, which is minimized only for fields above the coercive field.

For another sample, a 5 nm thick insulating SiO_2 layer was first deposited on top of the Ag layer before deposition of 40 nm Fe. As expected, the influence of the Fe layer is considerably weakened and a diamagnetic signal of Ag reappears. However, as Fig. 1(a) shows, the diamagnetic transition of Ag appears at a much lower T_b when compared to the Nb/Ag double layer. The destructive effect of Fe on the proximity effect in Ag also gives rise to a lower $T_A = 1.86$ K and $B_b(0) = 11$ mT [Fig. 1(b)] compared to $T_A = 3.02$ K and $B_b(0) = 18$ mT obtained for the Nb/Ag double layer. The Andreev pairs have a finite probability to tunnel into Fe via SiO_2 and back again, so that their phase coherence is compromised by I_{ex} of Fe. In addition, pinholes in the oxide barrier may play a role. Both effects lead to a reduced stability of PIS against magnetic and thermal perturbations. In other words, for a fixed temperature, smaller external fields are sufficient for the destruction of coherence[22] in comparison with Nb/Ag double layers.

Surprisingly, diamagnetic screening by Ag *without* a SiO_2 barrier reappears in Nb/Ag/Fe samples with much smaller $d_{\mathrm{Ag}} = 35$ and 43 nm in a certain range of magnetic field. Figure 2 clearly shows transitions around 3 K, which shift only

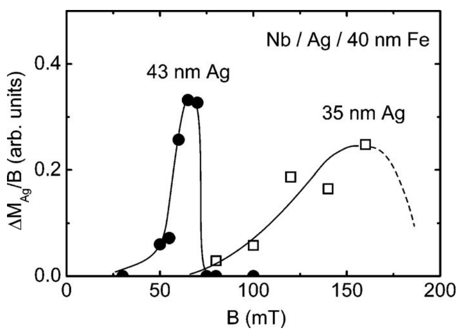

FIG. 3. $\Delta M_{\mathrm{Ag}}/B$ vs B of Nb/Ag/Fe samples with $d_{\mathrm{Fe}} = 40$ nm and different d_{Ag}. Lines are guides to the eye.

slightly to lower temperatures with increasing field B together with an increase of the jump $\Delta M_{\mathrm{Ag}}/B$. Moreover, the fields where the transitions are observed are much larger than the upper limit for the Nb/Ag samples discussed above. At these large values, T_c of the Nb layer is already reduced. Furthermore, a broadening of the transition is observed due to the flux penetration for fields exceeding the lower critical field B_{c1}. Figure 3 displays the height of the diamagnetic jump $\Delta M_{\mathrm{Ag}}/B$ vs magnetic field B for the different samples in order to illustrate the reappearance of the diamagnetic signal of Ag for certain magnetic fields.

One could argue that the effect is due to the presence of a magnetic stray field from the Fe layer. Although a thin magnetic layer does not exhibit a stray field close to the surface plane, a stray field arising from surface roughness[23] or generated by the domain structure of the ferromagnetic layer[24] can play a role. At this point, we cannot conclusively dismiss the possibility that the reappearance of the PIS in a higher applied magnetic field is due to the disappearance of the domains leading to suppression of the stray field. We also cannot exclude effects due to the flux trapped by the Nb layer cooled in the presence of such stray fields. However, the effect of a magnetic stray field from the Fe layer on the diamagnetic screening in Ag is considered to be negligible, as confirmed by zero-field measurements with a magnetized Fe layer; i.e., after complete demagnetization of the field coil, no diamagnetic signal of Nb or Ag was observed. We also mention that the Ag layer shows a complete Meissner effect (not shown) and, therefore, cannot contribute to any flux pinning.

The observed diamagnetic screening of Nb/Ag/Fe triple layers with thin d_{Ag} cannot be explained by present theories for semi-infinite S/N bilayers, since some initial assumptions are not valid. For instance, the variation of the superconducting energy gap Δ of S across the S/N interface cannot be approximated anymore by a step function because $d_{\mathrm{Ag}} \approx \xi_{\mathrm{Nb}}$. Moreover, the effect of pair breaking due to the contact with a F layer is not considered. However, the result can be at least qualitatively explained in the following way. For $d_{\mathrm{Ag}} = 35$ and 43 nm, the Andreev energy corresponds to $T_A = 48$ and 39 K, respectively, so that the coherence in the Ag layer will not be destroyed in contact with the ferromagnetic Fe layer. Theoretically, the lowest-energy state ($n=0$) at perpendicular incidence (maximum v_x) with respect to the interface would lie above the Fermi energies E_F of 21 and

17 meV, respectively [Eq. (1)], and well above $\Delta=1.2$ meV of Nb. No bound Andreev levels exist for trajectories with small incident angles with respect to the interface normal. The finite spin polarization in Fe and the external field B cause a shift of the Andreev levels to lower energies. The occupation of these levels gives rise to diamagnetic screening currents by correlated phase-coherent Andreev pairs. The magnitude of the diamagnetic signal is also determined by the density of Andreev pairs. Higher densities can be reached by increasing the total number of states between E_F and $E_F+\Delta$ with increasing B. The height of the jump $\Delta M_{\mathrm{Ag}}/B$ also increases with increasing magnetic field until it vanishes above fields, which completely destroy the phase coherence. Roughly speaking, the PIS in Ag is stabilized for thinner d_{Ag} (higher T_A) but at the same time, it is weakened by the pair breaking due to the contact with Fe. The balance between these effects can lead to the observation of a diamagnetic transition in Ag for certain d_{Ag} and B. This behavior is visualized in Fig. 3. The external magnetic field B acts on both Nb and Ag layers, whereas the pair breaking of Fe only acts on the Ag layer. In this picture, it is clear that for a constant thickness d_{Ag}, the transition temperature T_b becomes almost independent of B due to the strong internal magnetic field in Fe which is some orders of magnitude larger than the external field B.

In strong ferromagnets with $I_{ex}\tau > h$, such as Fe, the condensation amplitude decays on length scales of the order of l_{Fe}.[15] This suggests that for $l_{\mathrm{Fe}} \geqslant d_{\mathrm{Fe}}$, the coherence can be maintained over the whole layer thickness. Indeed, comparison of the resistivities of different Nb/Ag/Fe films yields lower limit for the mean free path $l_{\mathrm{Fe}}^{min} \approx d_{\mathrm{Fe}}$. Therefore, the coherence of the Andreev pairs is not destroyed despite the large thickness d_{Fe}. Pair breaking by spin-flip scattering can be neglected, since at low temperatures single Fe spins can hardly be flipped against the exchange field of their surrounding.[25]

In conclusion, we have observed a diamagnetic screening of the normal metal in high-quality Nb/Ag double layers with large electron mean free paths l_{Ag}. For thick Ag layers with $d_{\mathrm{Ag}} \geqslant (\xi_{\mathrm{Nb}}, \lambda_{\mathrm{Nb}})$, an additional Fe layer on top of Ag destroys the coherence of Andreev pairs. However, the diamagnetic signal of Ag is recovered if the Ag layer thickness is strongly reduced. This is due to the competition of induced superconductivity by Nb and pair breaking by Fe. This qualitative interpretation requires further theoretical investigations. Finally, we wish to point out that the proximity effect in $S/N/F/S$ contacts may be used in order to realize a tunable π contact by application of an external magnetic field.

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