Superconducting transition in Nb/Gd/Nb trilayers

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We report the observation of oscillations in the superconducting transition temperature T_c of Nb/Gd/Nb trilayers as the Gd layer thickness d_{Gd} is varied. The T_c minimum occurs at $d_{\mathrm{Gd}} \approx 13$ Å. The results are consistent with our previous study of Nb/Gd multilayers and are in support of the theoretical prediction of a π-phase junction in superconductor/ferromagnet/superconductor systems. [S0163-1829(96)08633-X]

The possibility of a π-phase coupling was proposed by Bulaevskii *et al.*[1] for junctions containing magnetic impurities in the tunnel barrier. In a π junction, the ground state has an intrinsic phase difference $\Delta\phi = \pi$ between the superconducting order parameters in the neighboring superconductors. These so-called ''π-phase'' states have also been predicted to exist in superconductor- (S-) ferromagnet (F) layered structures.[2–4] These predictions suggest that as the superconducting order parameter penetrates into the F layers via the proximity effect, it decays nonmonotonically with a characteristic length scale $\xi_M = \sqrt{4\hbar D_M / I}$, where D_M and I are the diffusion constant and the exchange interaction in the ferromagnetic layer, respectively. For certain F layer thicknesses d_F, the π-phase state is found to have a higher superconducting transition temperature T_c than the ''0-phase'' state with $\Delta\phi = 0$. As a result, T_c of S-F multilayers oscillates with d_F as the ground state switches between the 0-phase and the π-phase states.

We recently reported the observation of oscillations of the superconducting T_c in Nb/Gd multilayers as the Gd layer thickness d_{Gd} is varied.[5] For constant Nb layer thicknesses d_{Nb} of 500 Å and 600 Å, the T_c oscillation occurs in the thickness range 10 Å $\leqslant d_{\mathrm{Gd}} \leqslant$ 40 Å where the Gd ferromagnetism is well established. These results suggest the existence of the predicted π-phase state in S-F multilayers. ξ_M for Gd was determined to be 13.5 Å.

One manifestation of the π-phase coupling is a spontaneous persistent current and magnetic flux in a ring containing an odd number of π junctions in zero field.[1] Indeed, the observation of a spontaneous supercurrent would be a direct probe of the existence of a π-phase state. This phenomenon has been demonstrated in recent experiments probing the pairing symmetry in high-T_c superconductors.[6–8] π-phase proximity-coupled S-F structures using conventional low-T_c superconductors are another candidate for such an experiment. The simplest implementation would be a trilayer structure in the form of S-F-S with the top and bottom S layers connected to form a superconducting loop. However, in Nb/Gd/Nb trilayers with d_{Nb} in the range of 150–191 Å, Strunk *et al.*[9] observed plateaus in the dependence of T_c on d_{Gd}, but no clear oscillation. This discrepancy raises the question of whether the oscillatory T_c is a consequence of the multilayer geometry upon which the theoretical calculation of Radović *et al.*[2] is based.

In this paper, we present our study of the superconducting transition in sputter-deposited Nb/Gd/Nb trilayers; we have observed a nonmonotonic dependence of T_c on d_{Gd} that is consistent with our previous studies of Nb/Gd multilayers. In particular, the Gd layer thickness at which the minimum in T_c occurs is the same for both the trilayers and the multilayers.

In our previous work, the Nb/Gd multilayers consisted of uniform Nb layers of either 500 Å or 600 Å, and wedge-shaped Gd layers from 10 Å to 40 Å. In the present study of Nb/Gd/Nb trilayers, we used a fixed Nb layer of 250 Å and a wider Gd thickness range of 0–50 Å. Since a Nb layer has interfaces with two magnetic layers in a multilayer and with only one in a trilayer, the pair breaking effect should be approximately twice as strong in the former case. On this basis it was suggested by Strunk *et al.*[9] that the T_c dependence on d_{Nb} should be the same if the Nb layers in the multilayers are twice as thick as that in the trilayers. We therefore chose trilayers with $d_{\mathrm{Nb}} = 250$ Å to facilitate comparison with our multilayers with $d_{\mathrm{Nb}} = 500$ Å.

The deposition of the trilayer films was carried out in a dual-source dc magnetron sputtering chamber. The deposition conditions were similar to that of the multilayers described previously.[5] The layer thicknesses were controlled during growth with quartz-crystal thickness monitors. The wedge-shaped Gd layer was deposited by linear translation of the substrate behind a mask during growth, and its slope was set to be 1 Å/mm. The film was then cut into 2-mm-wide strips perpendicular to the wedge direction, giving a series of samples with varying d_{Gd}. In addition to the trilayer samples, a Nb/Gd multilayer film with $d_{\mathrm{Nb}} = 500$ Å and wedge-shaped Gd layers was also deposited in the *same* run using the *same* wedge Gd layer deposition procedure. The multilayer films had the same configuration as those in the previous study.[5] With the multilayers, we could check the reproducibility of our previous results, and make a direct comparison of superconductivity in the multilayer and trilayer geometries.

Low-angle and high-angle x-ray diffraction of the multilayer samples shows similarly good layering quality as that of the previous study, and the bilayer wavelengths determined from low-angle diffraction agree well with the desired values. Since the trilayers and the multilayers were deposited in the same fashion and at the same time, their quality should be comparable. The Gd layer thickness is determined from the sample position within the series. Because

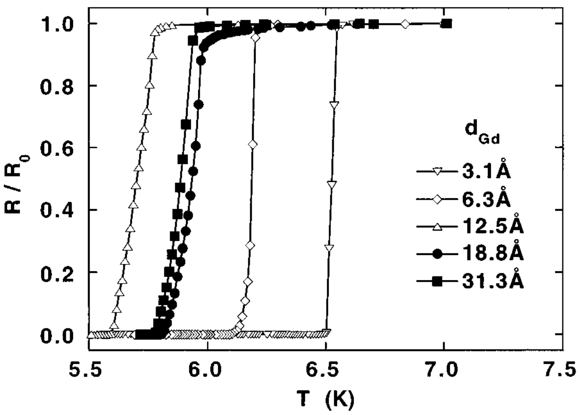

FIG. 1. Resistive superconducting transitions of Nb/Gd/Nb trilayers with $d_{\mathrm{Nb}}=250$ Å and $d_{\mathrm{Gd}}=3.1$ Å, 6.3 Å, 12.5 Å, 18.8 Å, and 31.3 Å.

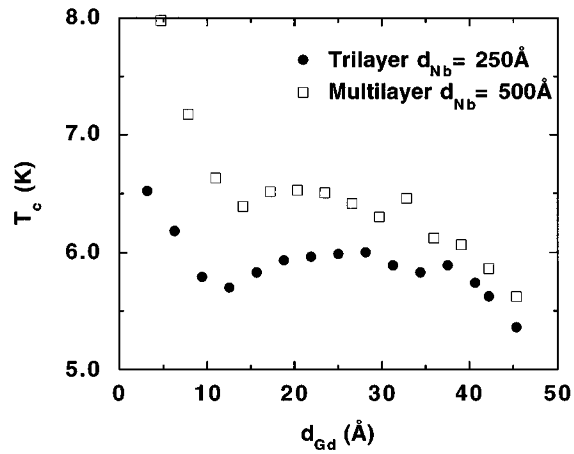

FIG. 2. Superconducting transition temperature T_c as a function of the Gd layer thickness d_{Gd} for Nb/Gd multilayers with $d_{\mathrm{Nb}}=500$ Å and Nb/Gd/Nb trilayers with $d_{\mathrm{Nb}}=250$ Å.

of the built-in thickness gradient of the wedge layer and the finite width of the samples, the thickness determination is accurate to ± 1 Å.

The superconducting transitions of both the trilayers and the multilayers were measured resistively using the standard four-probe technique. Because of the significant difference in the total thicknesses and the cross-sectional areas of the trilayer and multilayer samples, the measurement current for the trilayers was adjusted so that the current density was about the same (10 A/cm^2) when measuring the two systems. Similar to what we observed previously, the resistive transitions of some multilayers were again broad. We previously attributed this to the small volumes of inhomogeneity or damage at the edge when the samples were cut, and indeed, the susceptibility measurements on the same samples showed a sharp transition.[5] On the other hand, the resistive transitions in *all* the trilayers are sharp. Figure 1 shows the resistive transition of some trilayer samples with $d_{\mathrm{Gd}}=3.1$ Å, 6.3 Å, 12.5 Å, 18.8 Å, and 31.3 Å. The resistances are normalized to the residual normal-state values, which are nearly constant (16 $\mu\Omega$ cm) for all samples. The midpoints of the resistive transitions have been taken as the transition temperature T_c for the trilayers. The nonmonotonic dependence of the superconducting transition on d_{Gd} is clearly evident.

The d_{Gd} dependence of the transition temperature is plotted in Fig. 2 for both the trilayers and the multilayers. The oscillatory dependence of the T_c on d_{Gd} is clearly observed. For both the trilayers and the multilayers, the T_c minimum occurs at the same Gd layer thickness $d_{\mathrm{Gd}}\approx 13$ Å. The minimum in T_c signifies the transition from the 0-phase state ($d_{\mathrm{Gd}}<13$ Å) to the π-phase state ($d_{\mathrm{Gd}}>13$ Å). T_c then increases as d_{Gd} is further increased, reaching a maximum at $d_{\mathrm{Gd}}\approx 20$–25 Å before decreasing again. At $d_{\mathrm{Gd}}\approx 35$ Å, T_c of the trilayers shows a small upturn. It is possible that at this Gd layer thickness, the ground state of the S/F structures switches from the π-phase state back to the 0-phase state, but the data are insufficient to make a definitive statement on this point.

It should be noted that, although the T_c's of the multilayers with 500 Å Nb and the trilayers with 250 Å Nb are qualitatively similar, there are noticeable differences. For the same d_{Gd}, the T_c of a trilayer with $d_{\mathrm{Nb}}/2$ is always lower than that of a multilayer with d_{Nb}. The difference is the largest at low d_{Gd}, and diminishes at higher d_{Gd}. Thus, the symmetry argument leading to the simple expectation of trilayers with $d_{\mathrm{Nb}}/2$ being equivalent to multilayers with d_{Nb} is only approximately correct. Also, in the limit of $d_{\mathrm{Gd}}=0$, the multilayer is a bulk superconductor, whereas the trilayer becomes a single Nb film 500 Å in thickness and thus has a lower T_c because of the finite-size effect.[10]

It was suggested by Strunk *et al.*[9] that the penetration depth of Cooper pairs into the Gd layers may be too short for the T_c oscillation to be observed in Nb/Gd/Nb trilayers. This question can be resolved by measuring the parallel upper critical fields $H_{c2\|}$ of the Nb/Gd multilayers. If the Cooper pairs in one Nb layer do not penetrate through the Gd layers into the adjacent Nb layers, $H_{c2\|}$ should exhibit

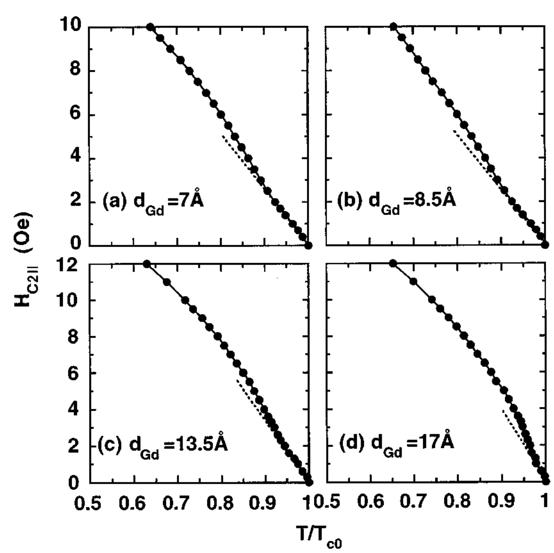

FIG. 3. Temperature dependence of the upper parallel field of Nb/Gd multilayers with $d_{\mathrm{Nb}}=500$ Å and (a) $d_{\mathrm{Gd}}=7$ Å, (b) $d_{\mathrm{Gd}}=8.5$ Å, (c) $d_{\mathrm{Gd}}=13.5$ Å, and (d) $d_{\mathrm{Gd}}=17$ Å. The dashed lines are the linear temperature dependence expected for 3D superconductors.

two-dimensional (2D) behavior, $H_{c2||} \propto \sqrt{1-T/T_c}$. On the other hand, if the Nb layers are coupled, $H_{c2||}$ should exhibit three-dimensional (3D) behavior, $H_{c2||} \propto (1-T/T_c)$. Figure 3 shows the parallel upper critical fields $H_{c2||}$ as a function of T/T_c for Nb/Gd multilayers with $d_{\mathrm{Nb}}=500$ Å and $d_{\mathrm{Gd}}=$ 7 Å, 8.5 Å, 13.5 Å, and 17 Å. Near T_c, $H_{c2||}$ varies linearly with T, indicating 3D behavior; i.e., the Cooper pairs tunnel through the F layer to couple with the neighboring superconductors. As the temperature is reduced, the S layers begin to decouple when the perpendicular coherence length $\xi_\perp$ of an equivalent anisotropic 3D superconductor becomes comparable to the multilayer period.[11] A dimensional crossover from 3D to 2D occurs, resulting a nonlinear temperature dependence. As expected, with increasing d_{Gd}, the dimensional crossover becomes closer to T_c. Similar effects have been observed in Fe/V multilayers.[12] The results conclusively show that in the d_{Gd} range where the T_c oscillation occurs, the Cooper pairs tunnel through the F layers.

In summary, we have observed a nonmonotonic dependence of the superconducting transition temperature T_c in Nb/Gd/Nb trilayers on the Gd layer thickness d_{Gd}. The results are consistent with our previous study of Nb/Gd multilayers. While an oscillatory T_c is a signature of the π-phase coupling, the existence of a π-phase state can only be proved conclusively by experiments that are sensitive to the phase of the superconducting order parameter. Our observation of T_c oscillations in Nb/Gd/Nb trilayers suggests that such an experiment is feasible without resorting to a more complex multilayer structure.

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