Proximity Effects in Superconductor/Insulating-Ferromagnet NbN/GdN Multilayers

John Q. Xiao* and C. L. Chien
Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218
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Superconductor/insulating-ferromagnet NbN/GdN multilayers reveal that the superconducting properties of the NbN layers are dominated by the insulating nature of the GdN layers, which curtail the pair-breaking effect despite the strong ferromagnetism. The effects of ferromagnetic walls on superconducting slabs have been captured. Very large parallel critical fields have also been observed.

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Proximity effects between superconducting and magnetic layers have been some of the most actively studied subjects in the past decades. It has been well established that magnetic moments and magnetic order are highly effective in destroying Cooper pairs and strongly depressing superconductivity. Thus in superconductor/ferromagnet (S/F) multilayers, one observes that the superconducting transition temperature (T_{cs}) decreases monotonically and rapidly as the magnetic layer thickness is increased [1–5]. Recently, the existence of the so-called π-phase state, in which the two adjacent superconducting layers have an opposite phase, has been theoretically predicted in superconductor/ferromagnet multilayers with suitable ferromagnetic layer thicknesses (d_m) [6,7]. A damped oscillatory behavior in T_{cs} as a function of d_m has been predicted, and recently observed in Nb/Gd multilayers [8]. However, in all previous cases, the ferromagnetic layers (e.g., Fe and Gd) in the multilayer systems are always metallic. The fundamental aspects of the low conductivity of the ferromagnetic but *insulating* layer have never been experimentally addressed. Because of the energy gap in the insulator, the proximity effects and the pair-breaking effects of the ferromagnet layers are likely to be strongly modified. To explore these important aspects, we have studied superconductor/insulating ferromagnet NbN/GdN multilayers. Unusual behaviors in superconducting properties and critical fields, very different from those of previous superconductor/metallic ferromagnet multilayers, have been observed.

Most of the common ferromagnets are elemental metals and alloys. All previous studies of S/F bilayers and multilayers have used those elemental metals and alloys. Nonmetallic ferromagnetic thin films (e.g., oxides, ferrites, and garnets) cannot be readily incorporated into multilayered thin films because of the requirement of high-temperature processing, which is incompatible with the fabrication of high-quality multilayers. An exception is GdN, which is a ferromagnetic insulator with a Curie temperature (T_{cm}) of as much as 60 K, depending on stoichiometry [9]. GdN has a resilient fcc NaCl structure ($a \approx 0.5$ nm), which is retained even for off-stoichiometric samples [10]. Thin films of GdN can be readily achieved using reactive sputtering with room temperature substrates. For the superconducting layers, we have chosen NbN, which can be made into thin films by reactive sputtering as well. NbN, also with a NaCl structure ($a = 0.44$ nm), is a type-II superconductor with a high superconducting transition temperature (T_{cs}) of about 15 K and a short superconducting coherence length ($\xi = 5$ nm) [11]. Therefore, even very thin layers of a few nm can still sustain superconductivity.

Multilayered samples have been fabricated by reactive magnetron sputtering onto Si(100) substrates at room temperature, using Nb (99.8% pure) and Gd (99.9% pure) targets sharing the same sputtering gas mixture of Ar and N_2. With a suitable gas mixture, NbN films with high T_{cs} ($\approx$15 K) have been made [10]. Unfortunately, the optimum gas mixtures for NbN films with high T_{cs} and GdN films with high T_{cm} are different. We have chosen a compromised gas mixture of 6.8 mT Ar and 1.2 mT N_2, with which thick NbN films show $T_{cs} = 9.6$ K, and GdN films show $T_{cm} \approx 25$ K, fulfilling the requirements of high values of T_{cs} and T_{cm}, and that T_{cm} be much higher than T_{cs}. Thus for the analyses that follow, the value for the superconducting transition temperature of NbN in the thick film limit is $T_{c0} = 9.6$ K.

The multilayer samples are designated as $(\mathrm{NbN})_{ds}/(\mathrm{GdN})_{dm}$, where the superconducting NbN layer thickness (d_s) and the magnetic GdN layer thickness (d_m) are denoted in nm. Three series of NbN/GdN multilayers have been studied. Two have a fixed superconducting layer thickness of $d_s = 4$ nm and $d_s = 6$ nm, which are close to, below and above, the superconducting coherence length, and the third has a fixed magnetic GdN layer thickness of $d_m = 6$ nm. The electrical resistance of the samples, its temperature and field dependences, have been measured using a four-terminal geometry on samples with known dimensions. The results are expressed in resistivity, defined as $\rho = RWN(d_s + d_m)/L$, and sheet conductance per bilayer, defined as $G_\square = L/RWN$, where R is the total resistance, N the number of bilayers, L and W the length and width, respectively, of the sample. Magnetic properties have been determined using a SQUID magnetometer, and the superconducting critical fields have been measured up to 50 kOe.

Representative low-angle x-ray spectra of $(\mathrm{NbN})_6/(\mathrm{GdN})_{dm}$ are shown in Fig. 1. The appearance of many

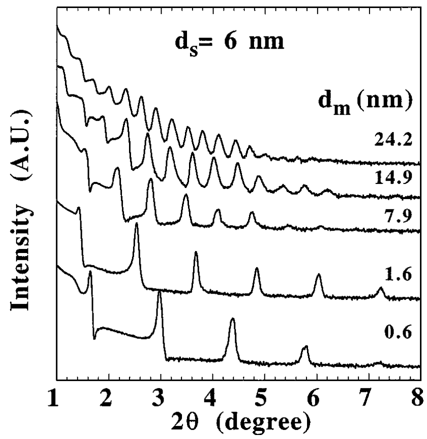

FIG. 1. Small-angle x-ray diffraction data of $(\mathrm{NbN})_{ds}/(\mathrm{GdN})_{dm}$ multilayers with a fixed $d_s = 6$ nm and $d_m = 0.6$, 1, 7.9, 14.9, and 24.2 nm.

diffraction peaks indicates a high-quality layer structure, which is also confirmed by TEM microscopy where flat and smooth layers have been observed [10]. Layer thicknesses, determined from low-angle x-ray diffraction, are all within 5% of the designed values.

Since the insulating nature of the GdN layers is of crucial importance for the present study, we have measured the resistivity of the NbN/GdN multilayers in considerable detail. We first verified the insulating nature of GdN thin films by making electrical measurements *in situ* during deposition using the van der Pauw method. Only open circuit resistance (>10 MΩ) has been noted in all thicknesses of the GdN films. To ensure that the GdN layers remain insulating in the NbN/GdN multilayers, we have analyzed the sheet conductance. Although resistivity (ρ) is commonly used for multilayers, the sheet conductance per bilayer ($G_\square$) defined earlier is the more appropriate quantity. Neglecting the conductance due to interface scattering, in a parallel network [4],

$$G_\square = \sigma_s d_s + \sigma_m d_m \,, \tag{1}$$

where σ_s and σ_m are the conductivity for NbN and GdN, respectively. With the inclusion of interface scattering,

$$G_\square = \sigma_s d_s + \sigma_m d_m - G_{sm} \,. \tag{2}$$

The negative sign in front of the interface conductance per bilayer (G_{sm}) is due to the fact that interface scattering increases resistivity and thus decreases conductivity.

In Fig. 2(a), we plot $G_\square$ vs d_s for $(\mathrm{NbN})_{ds}/(\mathrm{GdN})_6$ multilayers with a fixed GdN layer thickness of $d_m = 6$ nm. As expected from Eq. (1), $G_\square$ is indeed linear in d_s, and the slope gives the conductivity (σ_s) of the NbN layer which is thickness independent. The value of $\rho_s = 1/\sigma_s = 313\ \mu\Omega$ cm obtained is typical for NbN films [11]. In Fig. 2(b) we show $G_\square$ as a function of d_m for $(\mathrm{NbN})_6/(\mathrm{GdN})_{dm}$ and $(\mathrm{NbN})_4/(\mathrm{GdN})_{dm}$, both with a fixed value of d_s. Now, $G_\square$ is constant and independent of d_m, i.e., σ_m is negligibly small, illustrating

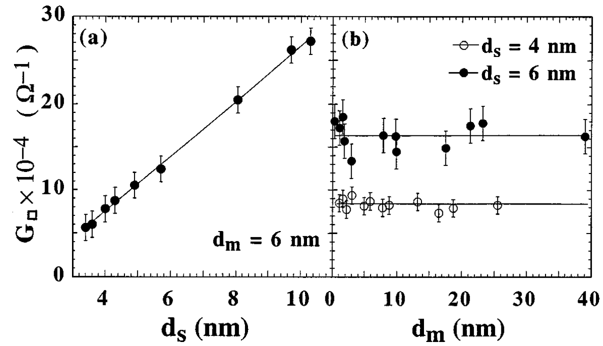

FIG. 2. Sheet conductance per bilayer ($G_\square$) of $(\mathrm{NbN})_{ds}/(\mathrm{GdN})_{dm}$ multilayers: (a) as a function of d_s for a fixed $d_m = 6$ nm, (b) as a function of d_s for a fixed $d_m = 4$ nm, and $d_m = 6$ nm.

that the GdN layer is insulating. Consequently, the sheet conductance per bilayer in Eq. (2) is essentially $G_\square \approx \sigma_s d_s - G_{sm}$. Indeed, one notes from Fig. 2(b) that the value of $G_\square$ for $d_s = 6$ nm is appropriately larger than that for $d_s = 4$ nm, and furthermore, the value of G_{sm} is about $4 \times 10^{-4}\ \Omega^{-1}$. These analyses conclusively show that the resistivity of the NbN layer is a constant (313 $\mu\Omega$ cm), and that the GdN layer is insulating.

For the superconducting properties, we first describe the results of multilayers with a fixed GdN layer thickness. The values of T_{cs} for $(\mathrm{NbN})_{ds}/(\mathrm{GdN})_6$ as a function of d_s are shown in Fig. 3, and can be well described by the well-known relation of [3]

$$T_{cs} = T_{c0}\left[1 - \frac{(0.85)^2 \xi_0 l \pi^2}{d_s^2}\right], \tag{3}$$

where ξ_0 and l are the superconducting coherence length and the electron mean free path, respectively, and $T_{c0} = 9.6$ K is the superconducting transition temperature in the thick-film limit. This relationship, a ramification of the strong magnetic pair breaking effect, has been observed

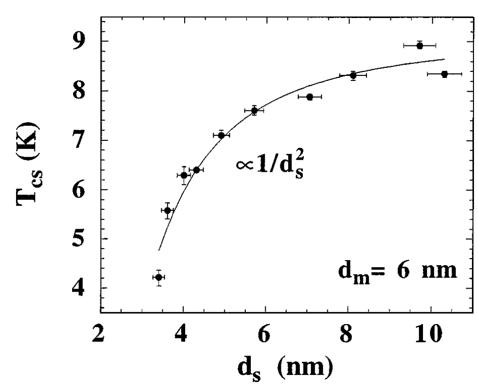

FIG. 3. Superconducting transition temperature (T_{cs}) of $(\mathrm{NbN})_{ds}/(\mathrm{GdN})_{dm}$ multilayers with a fixed $d_m = 6$ nm as a function of d_s. The vertical bars for the data points represent 90% to 10% change in the normal-state resistivity. The solid line is $T_{cs} = 9.6 \times (1 - 5.5/d_s^2)$.

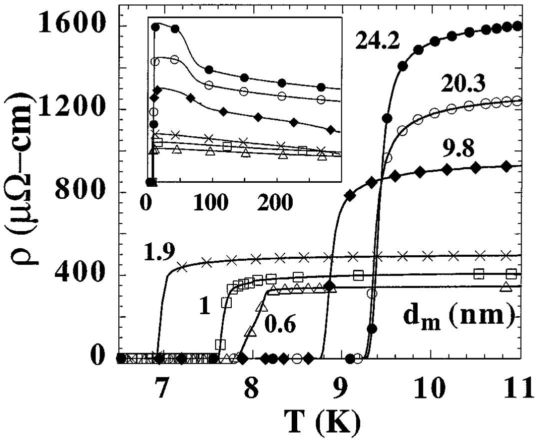

FIG. 4. Temperature dependence of resistivity (ρ) of $(NbN)_{ds}/(GdN)_{dm}$ multilayers with a fixed $d_s = 6$ nm and various d_m's. The inset shows the temperature dependence to 300 K.

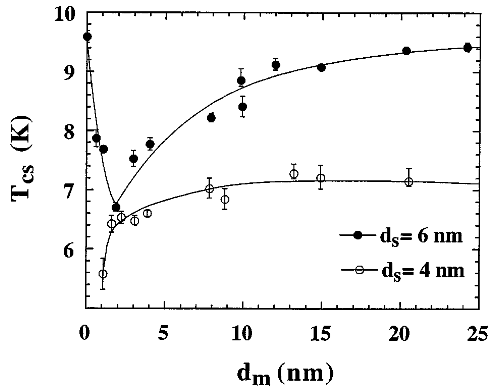

FIG. 5. Superconducting transition temperature (T_{cs}) of $(NbN)_{ds}/(GdN)_{dm}$ as a function of d_m with a fixed $d_s = 6$ nm and $d_s = 4$ nm.

in many superconductor/metallic ferromagnet multilayers [3].

Altogether different behaviors have been found in multilayers when the thickness of GdN is varied. Two series of samples of $(NbN)_4/(GdN)_{dm}$ and $(NbN)_6/(GdN)_{dm}$ have been measured. The resistivity (ρ) results of $(NbN)_6/(GdN)_{dm}$ are shown in Fig. 4, in which the GdN layer thickness (d_m) is labeled next to each curve. The resistivity of the sample increases rapidly and monotonically with increasing GdN layer thickness (d_m); these changes persist to higher temperatures as shown in the inset. This is expected from the insulating nature of the GdN layers. The quantities of ρ and $G_\square$ are related by $\rho = (d_s + d_m)/G_\square$. For a fixed d_s, since $G_\square$ is essentially constant, ρ increases with d_m unabatedly as observed.

The insulating GdN layers play a crucial role in the superconducting properties of $(NbN)_{ds}/(GdN)_{dm}$ multilayers. As shown in Fig. 4, the superconducting temperature varies nonmonotonically with d_m, first decreasing before increasing with d_m. These results for $d_s = 6$ nm and those for $d_s = 4$ nm are summarized in Fig. 5. We first discuss the results for $d_s = 6$ nm. The value of T_{cs} first decreases rapidly with increasing d_m. However, for $d_m > 2$ nm, T_{cs} monotonically *increases* with d_m, before eventually reaching the terminal value of $T_{cs} \approx 9.4$ K for $d_s = 6$ nm. The initial decrease of T_{cs} at low d_m is caused by strong pair-breaking effects of the ferromagnetic layers. This mechanism would assure a continuing reduction of T_{cs} as d_m is increased, as has been experimentally observed in many multilayers of a superconductor with a ferromagnetic metal [3]. The increase of T_{cs} with d_m in S/F multilayers has not been observed previously. The present results indicate that, for $d_m < 2$ nm, the Cooper pairs tunnel across the insulating GdN layers and suffer the detrimental effects of the ferromagnetic layers. However, for $d_m > 2$ nm, the insulating GdN layers severely limit the Cooper pairs from diffusing into GdN layers and thus reducing the strong magnetic pair breaking effect. For a sufficiently thick GdN layer ($d_m \geq 10$ nm) the $(NbN)_{ds}/(GdN)_{dm}$ multilayers behave effectively as isolated NbN layers of thickness d_s. In the thick GdN limit, where tunneling ceases completely, the only pair-breaking effect for the isolated NbN layers is that of ferromagnetic walls. For the samples with $d_s =$ 6 nm, the terminal value of T_c Å 9.4 K in the thick d_s limit is only slightly smaller than $T_{c0} = 9.6$ K, indicating that the effects of the ferromagnetic walls are insignificant.

The superconducting properties of superconductor/insulating ferromagnet multilayers have not been explored experimentally but only theoretically. Tokuyasu, Sauls, and Rainer have studied a superconducting slab sandwiched between magnetic and nonmagnetic insulators [12]. The effects of the ferromagnetic wall are described by a magnetic surface pair-breaking parameter $b = \xi_0 \tan(\theta/2)/(2d_s)$, where ξ_0 is the coherence length, d_s the thickness of the superconducting slab, and θ the spin mixing angle. Strong pair-breaking effects due to the ferromagnetic walls can be expected for large values of b ($b > 0.2$), whereas weak pair-breaking effects with a negligible reduction of T_c can be anticipated for small values of b ($b < 0.1$). The present results of NbN/GdN with $d_s = 6$ nm appear to be in the weak pair-breaking regime. However, since the parameter b is inversely proportional to d_s, the effects of the ferromagnetic walls are larger for thinner d_s, an expectation supported by our results. We have also measured $(NbN)_4/(GdN)_{dm}$ with a smaller value of $d_s = 4$ nm, which is below the coherence length of NbN. Now, as shown in Fig. 5, after the initial rapid decrease of T_c at small d_m, T_c again rebounds. However, the terminal value of T_c is only about 7 K, considerably less than T_{c0}, revealing stronger effects of the magnetic walls. Thus the salient features of the ferromagnetic walls have been observed.

The unusual characteristics of the NbN/GdN multilayers are also reflected in their dimensionality and critical fields. It is well known that in the 2D limit, i.e., no interlayer coupling between adjacent superconducting layers, the parallel critical field ($H_{c\|}$) varies as $(1 - T/T_{cs})^{1/2}$, whereas in the 3D case, $H_{c\|}$ varies as $(1 - T/T_{cs})$ [3,13]. In Fig. 6 we plot the temperature dependence of the critical field for the $(\mathrm{NbN})_6/(\mathrm{GdN})_{dm}$ samples with a fixed $d_s = 6$ nm. As expected, the perpendicular critical fields ($H_{c\perp}$) of all samples depend linearly on T/T_{cs} as shown in the inset. However, both 2D and 3D behaviors are observed in $H_{c\|}$. For thin GdN layers (e.g., $d_m = 1.1$ nm), $H_{c\|}$ shows a quasilinear T dependence, a signature of 3D behavior, corroborating the earlier conclusion that the superconducting electrons tunnel across the magnetic layers. For thick GdN layers ($d_m \geq 9.9$ nm), $H_{c\|}$ shows a distinctive $(1 - T/T_{cs})^{1/2}$ dependence, a 2D behavior expected from isolated thin superconducting layers. There is an indication of a 2D–3D transition for GdN with intermediate thicknesses (e.g., $d_m = 7.8$ nm): a square root of T dependence at low temperatures, followed by a linear T dependence near T_c [13]. Thus the critical field data also confirm that the unusual superconducting behaviors of the GdN/NbN multilayers are due to the insulating nature of the GdN layers.

Another very interesting result is that the values of $H_{c\|}$ are extremely large. With an external field of 50 kOe at our disposal, we could only measure $H_{c\|}$ within 6% of T_{cs} for the sample with $d_m = 1.1$ nm, and within 1% of T_{cs} for samples with $d_m \geq 24.2$ nm. The value of $H_{c\|}$ at lower temperatures would be enormous. These large parallel critical fields arise partly from the presence of the ferromagnetic GdN layers which channel a disproportionately larger fraction of the magnetic flux.

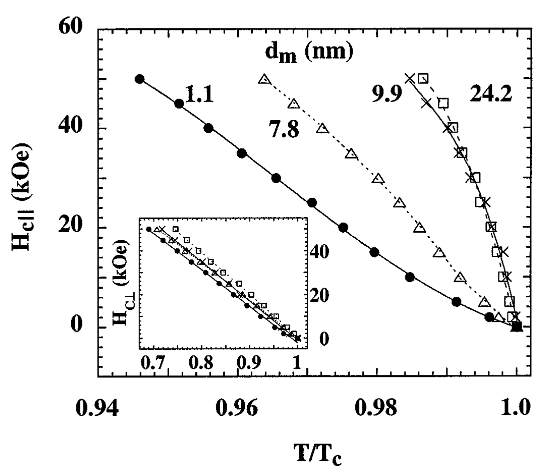

FIG. 6. Temperature dependence of parallel critical field ($H_{c\|}$) of $(\mathrm{NbN})_{ds}/(\mathrm{GdN})_{dm}$ multilayers with a fixed $d_s = 6$ nm. The inset shows the results of the perpendicular critical field ($H_{c\perp}$) of the same samples.

The high resistivity of the GdN also severely curtails the extent of the superconducting wave function into the GdN layer, in which a large magnetic induction is present. The extremely large values of $H_{c\|}$ for these multilayers with large d_m are attractive for technological reasons.

In summary, we have studied superconductor/insulating ferromagnet multilayers, specifically designed to probe the effects on superconductivity due to the insulating nature of the ferromagnetic layer. In contrast to all previously studied superconductor/metallic ferromagnet multilayers, the superconducting transition temperature *increases* with increasing ferromagnetic (GdN) layer thickness. Resistivity and sheet conductance measurements demonstrate conclusively that this unusual behavior is caused by the insulating GdN. Consequently, the Cooper pairs are progressively confined within the NbN layers and raise T_{cs} substantially. Extremely large critical fields and dimensionality effects have been observed.

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*Permanent address: Department of Physics and Astronomy, University of Delaware, Newark, DE 19716.

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