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Suppression of superconductivity in Nb by IrMn in IrMn/Nb bilayers

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Effect of antiferromagnet on superconductivity has been investigated in IrMn/Nb bilayers. Significant suppression of both transition temperature (T_c) and lower critical field (H_{c1}) of Nb is found in IrMn/Nb bilayers as compared to a single layer Nb of same thickness; the suppression effect is even stronger than that of a ferromagnet in NiFe/Nb bilayers. The addition of an insulating MgO layer at the IrMn-Nb interface nearly restores T_c to that of the single layer Nb, but H_{c1} still remains suppressed. These results suggest that, in addition to proximity effect and magnetic impurity scattering, magnetostatic interaction also plays a role in suppressing superconductivity of Nb in IrMn/Nb bilayers. In addition to reduced T_c and H_{c1} , the IrMn layer also induces broadening in the transition temperature of Nb, which can be accounted for by a finite distribution of stray field from IrMn. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4824891>]

Antiferromagnet/ferromagnet (AFM/FM)^{1,2} and ferromagnet/superconductor (FM/S)^{3–7} heterointerfaces have been investigated intensively in the last few decades due to their importance in both fundamental physics studies and device applications. Compared to the AFM/FM and FM/S interfaces, however, the amount of work on antiferromagnet/superconductor (AFM/S) heterointerfaces is quite limited.^{8–16} Since the classic work of Hauser *et al.* on Pb/Cr bilayers,⁹ several investigations have been carried out on AFM/S junctions using different combinations of antiferromagnets and superconductors, including Nb/Cr multilayer,¹⁰ Cr/V/Cr trilayer,¹¹ and Nb/ γ -FeMn bilayer structures.¹² Key findings reported so far include suppression of critical temperature (T_c) of S by the AFM^{10–12} and existence of Josephson current in S/AFM/S trilayer structures.^{12,14–16} The suppression of superconductivity has been discussed in the context of both proximity effect and de-pairing of Cooper pair by magnetic impurities diffused from the adjacent AFM layer during and/or after the sample preparation.^{9–13} It is worth noting that both Cr and γ -FeMn have a complex spin structure,^{17–21} which may also play a role in determining the nature of proximity effect at the interface with superconductors. In fact, Hübener *et al.* have observed strong suppression of T_c in Cr/V/Cr trilayers at the onset of a spin-density wave in Cr.¹¹ In practice, the situation may be more complex due to the presence of uncompensated spins at the AFM surface as manifested in the strong exchange coupling between AFM and FM,² and formation of domain walls in the AFM layer interacting with a FM.^{22–24} Although it is not straightforward to probe its existence, domain walls may also form in the AFM layer of AFM/S bilayers due to magnetostatic interactions. In a recent work, we have found that magnetostatic interactions between Nb and IrMn/NiFe in a Nb/IrMn/NiFe trilayer induces instability in the exchange bias field at the

IrMn/NiFe interfaces.²⁵ In order to gain further insight of AFM/S interface, in this work, we focus on simple IrMn/Nb bilayers and IrMn/MgO/Nb trilayers and study how IrMn affects the superconductivity of Nb. It was found that IrMn suppresses T_c and lower critical field (H_{c1}) of the Nb more significantly than that of NiFe in NiFe/Nb bilayers. The suppression of T_c is mitigated by the insertion of a thin MgO layer at the interface between IrMn and Nb, while H_{c1} remains to be largely suppressed by IrMn across the MgO layer. These results suggest that both proximity effect (and/or magnetic impurity scattering) and magnetostatic interactions are present at the AFM/S interface; the former is responsible for the suppression of T_c , whereas the latter mainly reduces H_{c1} when the effective magnetostatic field is moderate as compared to the upper critical field of Nb. The experimentally observed broadening of T_c in IrMn/Nb and IrMn/MgO/Nb samples is discussed using an analytical model involving a finite distribution of stray field from the IrMn layer. Our results demonstrate that magnetostatic interaction indeed exists and plays an important role in determining the characteristics of AFM/S junctions.

Three groups of samples have been fabricated by sputtering with a base pressure of 10^{-6} Pa on SiO₂/Si substrates under the same deposition conditions. These samples are Group A: IrMn(10)/Nb(t_{Nb})/Ru(5) and IrMn(t_{IrMn})/Nb(100)/Ru(5), Group B: Nb(t_{Nb})/Ru(5) and NiFe(10)/Nb(100)/Ru(5), and Group C: IrMn(10)/MgO(t_{MgO})/Nb(100)/Ru(5) and NiFe(10)/MgO(3)/Nb(100)/Ru(5). Here, the figures inside the brackets are thicknesses of individual layers in unit of nanometer. The thickness of Nb (t_{Nb}) was set at 20 nm, 50 nm and 100 nm, respectively. The thicknesses of IrMn (t_{IrMn}) and MgO (t_{MgO}) were varied in the range of 0–20 nm and 0–3 nm, respectively. The Ru capping layer was introduced to protect Nb from oxidation. Although we cannot exclude the proximity effect between Ru and Nb, it should not affect the discussion and analysis in the subsequent sections because the same Ru capping layer is used for

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all the samples and we focus on the comparison between samples with/without IrMn or with/without MgO layer. The Group B samples were used as the control to study the effect of IrMn on Nb, whereas the addition of MgO in Group C samples was to differentiate the proximity effect from magnetostatic interactions. The magnetic measurements, including zero-field cooling (ZFC) and initial (M-H) curves, were performed using a superconducting quantum interference device (SQUID) magnetometer. As superconductors are sensitive to the magnetic field and their magnetic history, all the measurements were commenced after nulling the field with oscillatory mode and then resetting the field to the starting value; this could maintain the residual field within a range of 2 Oe. For samples with a NiFe layer, an external field of 50 Oe was applied at 50 K to saturate the NiFe layer in the field direction. The applied field was then oscillated to 0 Oe before the ZFC measurement was conducted.

Figure 1(a) shows the normalized ZFC curves of Nb(100), IrMn(10)/Nb(100), and NiFe(10)/Nb(100) at an applied field of 10 Oe, where the magnetic moment of NiFe is deducted from the original data for NiFe(10)/Nb(100). As shown in the figure, T_c of Nb(100) decreases from 8.98 K in Nb(100) to 8.52 K in IrMn(10)/Nb(100) and 8.63 K in NiFe(10)/Nb(100) bilayers, respectively. Compared to Nb(100) and NiFe(10)/Nb(100), the transition width of IrMn(10)/Nb(100) is notably wider too. Figure 1(b) presents the normalized initial M-H curves of Nb(100), IrMn(10)/Nb(100), and NiFe(10)/Nb(100) at 8.2 K, which is well below T_c for all the three samples. The three samples all show typical Meissner effect of a type-II superconductor. The magnetic moment starts to upturn when the external field exceeds H_{c1} , due to the penetration of magnetic flux into the superconducting layer. H_{c1} is remarkably smaller for the IrMn/Nb and NiFe/Nb samples as compared to the Nb single layer. The influence of IrMn on the superconductivity of Nb is further manifested in the ZFC curves of Nb(20) and IrMn(10)/Nb(20) shown in Fig. 1(c). The ZFC curves were

measured at an applied field of 100 Oe and the inset shows the initial M-H curves at 6 K. Although T_c (~ 7.8 K) for Nb(20) is lower than that of Nb(100), the transition from normal state to superconducting state is clearly observed in Nb(20). In a sharp contrast, for IrMn(10)/Nb(20), the superconducting phase does not appear even after temperature is lowered to 4.2 K and there was no Meissner effect observed at 6 K either [Fig. 1(c)]. Although it is not shown here, similar results have been obtained for Nb(50) and IrMn(10)/Nb(50). Both T_c and H_{c1} of IrMn(10)/Nb(50) were found to be smaller than those of Nb(50).

The three pairs of samples, Nb(100, 50, 20) and IrMn(10)/Nb(100, 50, 20), all exhibit the same trend, *i.e.*, both the T_c and H_{c1} of Nb are suppressed by the IrMn layer, and the suppression is even stronger than that of the NiFe layer. Before we discuss the magnetic origin of these effects, we first examine if this was caused by the difference in crystalline structure of the Nb films grown on different buffers, *i.e.*, SiO₂, IrMn, NiFe, and MgO. To this end, the X-ray diffraction (XRD) patterns of the samples have been measured, which all exhibit clear Nb(110) and Nb(220) peaks, corresponding to the body-centered cubic phase of Nb. The Nb films are polycrystalline; the grain size calculated from the width of XRD peaks is 21.1 nm, 20.7 nm, 18.0 nm, 17.1 nm, and 17.3 nm for IrMn(10)/Nb(100), NiFe(10)/Nb(100), Nb(100), IrMn(10)/MgO(3)/Nb(100), and NiFe(10)/MgO(3)/Nb(100), respectively, *i.e.*, grain size of Nb grown on IrMn (NiFe) underlayer is 16.7% (14.6%) larger than that of Nb film grown on SiO₂ directly. Bose *et al.* have studied the grain size dependence of T_c and superconducting energy gap in Nb nanocrystalline thin films and found that T_c decreases with the average particle size of Nb obtained from the XRD line shape analysis.²⁶ In this context, if the grain size were the only deciding factor in our case, T_c of Nb grown on IrMn and NiFe underlayer would be higher than that of the Nb film grown on SiO₂ directly. However, the results in Fig. 1 just show the opposite, *i.e.*, T_c of Nb in IrMn/Nb and NiFe/Nb bilayers is reduced as compared to the single Nb layer of the same thickness. Apart from the structural factor, other major factors that could affect the superconducting properties of Nb in the hybrid structures are the proximity effect, de-pairing by magnetic impurities in the Nb layer and magnetostatic interactions. Figure 1(d) shows the IrMn thickness dependence of T_c and H_{c1} for IrMn(t_{IrMn})/Nb(100). As can be seen, both T_c and H_{c1} of Nb(100) are suppressed by the IrMn layer even at a thickness of 1 nm, which agrees with the results of Bell *et al.* observed in Nb/ γ -FeMn bilayers.¹² It should be noted that IrMn is found to be still in the antiferromagnetic state at 2 K at a thickness of 1.5 nm, whereas the AFM onset thickness of FeMn at the same temperature is 2 nm.²⁷ The suppression of superconductivity at very small AFM thickness can be attributed to the latter's very short coherence length, 2.4 nm for FeMn¹² and 3.1 nm for IrMn. The latter is calculated by using $v_F = 0.94 \times 10^6$ m/s, $T_N = 690$ K, and $l = 1.39$ nm for the Fermi velocity, Neel temperature, and mean-free path of IrMn, respectively. Since both T_c and H_{c1} drop sharply at 1 nm of IrMn and beyond which they are almost constant, proximity effect presumably dominates over de-pairing by magnetic impurities inside the Nb layer. In order to further elucidate the role of

FIG. 1. (a) Normalized ZFC curves at 10 Oe and (b) normalized M-H curves at 8.2 K for Nb(100), NiFe(10)/Nb(100) and IrMn(10)/Nb(100). (c) ZFC curves at 100 Oe and M-H curves at 6 K (insert) for IrMn(10)/Nb(20) and Nb(20). Open triangulars and squares indicate the results for IrMn(10)/Nb(20) and Nb(20), respectively. (d) IrMn thickness dependences of T_c and H_{c1} for IrMn(t_{IrMn})/Nb(100), which are indicated by the solid and open square, respectively.

FIG. 2. (a) Normalized ZFC curves at 10 Oe and (b) normalized M-H curves at 8.2 K for Nb(100), IrMn(10)/Nb(100), and IrMn(10)/MgO(3)/Nb(100), respectively. (c) Normalized ZFC curves at 10 Oe and (d) normalized M-H curves at 8.2 K for Nb(100), NiFe(10)/Nb(100), and NiFe(10)/MgO(3)/Nb(100).

magnetostatic interaction, we have introduced a 3 nm-thick MgO layer at the Nb/IrMn and Nb/NiFe interface and compared their magnetic properties with the samples without an MgO layer. This is motivated by the fact that a thin MgO layer should be able to suppress both the proximity effect and diffusion of magnetic impurities significantly but not the magnetostatic interactions.

Figures 2(a) and 2(b) show the ZFC and initial M-H curves of IrMn(10)/Nb(100), IrMn(10)/MgO(3)/Nb(100), and Nb(100), respectively. Compared to IrMn(10)/Nb(100), which has a T_c of 8.52 K, T_c of IrMn(10)/MgO(3)/Nb(100) is much higher and is nearly the same as that of Nb(100). Although T_c has increased significantly, the transition becomes much broader with the addition of the MgO layer. Similar increase of T_c was also seen in NiFe(10)/MgO(3)/Nb(100) ($T_c = 8.90$) as compared to NiFe(10)/Nb(100) ($T_c = 8.63$), though it is not as significant as in the case of IrMn/Nb. As shown in Figs. 2(b) (for the case of IrMn) and 2(d) (for the case of NiFe), in addition to T_c , the addition of MgO also improves the upper critical field (H_{c2}), which agrees well with the theoretical dependence of H_{c2} on T_c .²⁸ In a sharp contrast to the significant recovery of T_c and H_{c2} , only a subtle change has been observed in H_{c1} . Figure 3 shows the MgO thickness dependence of T_c and H_{c1} in IrMn(10)/MgO(t_{MgO})/Nb(100) samples. The solid and open squares at the top-left corner denote the T_c and H_{c1} of

FIG. 3. MgO thickness dependences of T_c (solid square with line) and H_{c1} (open square with line) for IrMn(10)/MgO(t_{MgO})/Nb(100). The solid and open squares at the top-left corner denote T_c and H_{c1} of Nb(100) single layer, respectively.

Nb(100), respectively. It can be seen clearly that T_c is gradually restored with increasing the MgO thickness, while H_{c1} is not affected much by increasing the MgO thickness. The proximity effect, scattering by magnetic impurities, and magnetostatic interaction all affect the superconductivity of adjacent Nb layer. While both the proximity effect and magnetic impurity scattering effectively break the Cooper pairs, effect of magnetostatic interaction is reflected more prominently in the lower critical field. Therefore, the rebound of T_c with the insertion of MgO can be attributed to suppression of proximity and scattering effect, while the change in distribution of T_c is presumably caused by the spatial variation of stray field. In samples involving NiFe, the stray field comes from domain walls. As for IrMn, possible sources for the stray field include both the uncompensated surface spins and domain walls.^{24,27} The latter could be developed in the thin IrMn layer through interaction with Nb below T_c . The random distribution of these net surface or volumetric spins^{22,23} naturally leads to a non-uniform stray field across the sample plane. The proximity effect is less sensitive to the random distribution of magnetic domains as it is dominantly a surface effect. Therefore, the widening of transition region can be mainly associated with the finite distribution of stray field.

For a conventional superconductor, the relationship between $T_c(H)$ and magnetic field (H) can be expressed as $T_c(H) = T_c(0)\sqrt{1 - \frac{H}{H_{c2}(0)}}$, where $T_c(0)$ is the transition temperature at zero external field, $H_{c2}(0)$ is the upper critical field at 0 K, ~ 5 T for bulk Nb,²⁹ and H is the external field or stray field in this case. In order to quantify the effect of stray field on T_c , we assume that the stray field from IrMn and NiFe has a log-normal distribution ($\text{Log-N}(\mu, \sigma^2)$) in the film plane (Fig. 4(a)) and then use the equation to calculate the corresponding distribution of T_c (inset of Fig. 4(a)). Based on the simulated T_c distribution, the M-T curves are readily obtained by assuming that all the regions with a T_c higher than the measurement temperature will give a perfect

FIG. 4. (a) Log-normal distributions of stray field in IrMn(10)/Nb(100) (solid-line) and NiFe(10)/Nb(100) (dashed-line); the inset is the distribution of corresponding T_c . Also shown in the figure (dotted line) is the calculated stray field distribution in IrMn(10)/Nb(100) by assuming a Gaussian distribution of the patch size with uncompensated spins. (b) Simulated and experimental ZFC curves of IrMn(10)/Nb(100), IrMn(10)/MgO(3)/Nb(100), NiFe(10)/Nb(100), and NiFe(10)/MgO(3)/Nb(100).

diamagnetism. Although we have also tried to use normal distribution to simulate the stray field distribution, it failed to fit the experimental results. As shown in Fig. 4(b), a good agreement between simulation and experiment has been obtained through optimizing the parameters μ and σ for all the four cases, i.e., IrMn/Nb and NiFe/Nb with or without the 3 nm thick MgO layer. The values of μ and σ which produced the best fits are given in Table I. Also shown in the table are the mean and standard deviation of the stray field. Both the mean value and standard deviation of the stray field in IrMn/Nb are larger than those in NiFe/Nb.

These results can be understood qualitatively by assuming that there is a thin layer of uncompensated spins aligned vertically on the AFM surface. In this case, the spatial frequency spectrum of stray field can be calculated by $H_z(k, z) = \frac{1}{2} \frac{m(k)}{A} k e^{-kz}$, where $m(k)$ is the lateral spatial frequency spectrum of uncompensated spins, A is the surface area, k is the spatial frequency, and z is the vertical distance from the surface of uncompensated spins.³⁰ This equation implies that the larger the spatial frequency the faster the decay of stray field in the vertical direction. The range of spatial frequency is determined by the patch size of local areas with uncompensated spins. For simplicity, we assume that the patches with uncompensated spins have a rectangular shape with a length much larger than the width. We further assume that the width follows a normal distribution. With this assumption, we can simply the two-dimensional problem into a one-dimensional one which can be handled analytically without losing the essential physics. For a patch width of a , the field at the center of and a distance z away from the patch is readily obtained as $H_z(0, z) = 2m_s a / (a^2 + 4z^2)$, which is further simplified to $H_z(0, z) = 2m_s / a$ when $a \gg z$. Here, m_s is the areal density of uncompensated spin moment. Substituting $a = 2m_s / H_z(0, z)$ into the normal distribution of the patch width, we obtained a distribution of stray field which resembles well the log-normal distribution used to fit the experimental data. For comparison, the calculated distribution is shown in Fig. 4(a) by the dotted line for the IrMn/Nb sample, in which the mean and standard deviation of patch width are assumed to be 4.9 nm and 1.2 nm, respectively. As for the uncompensated spin moment density, it is assumed to be the same as that reported for (CoFe/Pd)/IrMn multilayers, i.e., 3.4×10^{-4} A m²/m².³⁰ This gives a mean value of 1735 Oe for the stray field. Although the exact values of the stray field depend on both the patch size and uncompensated spin density, the shape of the distribution will remain closely resembling the log-normal distribution due to the simple fact that the stray field is inversely proportional to the patch size. The large standard deviation of stray field in IrMn/Nb indicates that the uncompensated spins and/or domain walls are distributed more randomly in IrMn as compared to NiFe. With the

addition of MgO, the distribution becomes even wider; this is because, in addition to the spacing effect, the suppression of proximity and scattering effects makes magnetostatic interaction more prominent even the reduction of T_c is small. Before ending this section, we have to point out that, although we have assumed that the uncompensated spins are aligned vertically on the surface, the main results will not change even when we assume that the spins are aligned in-plane because we are mainly interested in the stray field distribution rather than the absolute strength.

In summary, suppression of T_c and H_{c1} of Nb film is observed in IrMn/Nb bilayers; the effect is even stronger than that of a ferromagnetic NiFe layer. Further investigation by inserting an MgO insulating layer between the AFM and S layer revealed that H_{c1} is mainly suppressed by magnetostatic interactions, whereas the proximity and/or spin-dependent scattering effects dominate the reduction of T_c . The experimentally observed broadening of T_c in IrMn/Nb and IrMn/MgO/Nb samples can be fitted well by assuming a log-normal distribution of stray field from the IrMn layer. Our results demonstrate that magnetostatic interaction indeed exists and plays an important role in determining the characteristics of AFM/S junctions.

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TABLE I. Parameters used in fitting the experimental data.

	μ	σ	Mean (Oe)	STD (Oe)
NiFe/Nb	6.87	0.35	1022	369
NiFe/MgO/Nb	6.67	0.5	896	478
IrMn/Nb	7.56	0.5	2177	1160
IrMn/MgO/Nb	7.26	0.7	1815	1444

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