Enhancement of superconductivity by decreased magnetic spin-flip scattering: Nonmonotonic T_C dependence with enhanced magnetic ordering

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Fe/Cu superlattices exhibit a structural and magnetic transition with increasing Fe layer thickness, thus increasing the Curie temperature. An enhancement in the superconducting transition temperature is found in proximity coupled Nb/[Fe/Cu] layers as the Fe thickness increases. These results indicate a weakening of the magnetic proximity effect for the material with higher Curie temperature, and suggest a dominant role of spin-flip scattering in the pair breaking processes which give rise to the proximity effect. [S0163-1829(99)03221-X]

The interaction between superconductivity and magnetism has long been a field of interest in research where a rich variety of phenomena can be found. From the first observation of reentrant superconductivity in $ErRh_4B_4$,[1] many different phenomena have been described ranging from the superconductivity suppression by the strong pair breaking effects of magnetic impurities[2] to the coexistence of giant magnetoresistance and superconductivity in $YBa_2Cu_3O_7/La_{0.67}Ba_{0.33}MnO_3$ superlattices.[3]

This superconductivity-magnetism interaction has been studied in many different geometrical configurations such as magnetic atoms in a lattice,[1] multilayers and superlattices,[4] magnetic dots on a superconductor,[5] etc. In particular, superconducting/ferromagnet (SC/F) multilayers[4,7–12] are useful model systems for the controlled study of the interplay between these two phenomena and as tests of various theoretical predictions. The superconducting transition temperature (T_C) is the most basic parameter to be affected by the (SC/F) multilayers structure. Experimental studies of the changes in T_C with ferromagnetic layer thickness (t_F) have revealed different qualitative behaviors in different systems: a fast decrease in Fe/V (Refs. 6 and 7) and Nb/Gd,[11] and steps or oscillations in Nb/Gd,[10] Nb/CuMn,[10] NbN/GdN,[11] and Nb/Fe.[12]

The dependence of T_C with t_F is a consequence of pair breaking and is understood with different theoretical approaches.[2,13–16] Monotonic dependences are straightforward consequences of proximity effect theories[2,16] whereas periodic switching from the traditional ''0-phase'' to ''π-phase'' coupling between superconducting layers through the magnetic material could give rise to oscillatory behavior.[13] In this latter model, the characteristic decay constant of the order parameter within the ferromagnetic layers is complex, which opens the possibility of π-phase difference between two neighboring superconducting layers.

However, a clear interpretation of experiments in (SC/F) multilayers can be difficult because the oscillations in T_C are predicted and observed in a range of t_F where the layer magnetic properties are also changing. For example, in Nb/Gd, the Gd layer Curie temperature becomes zero around $t(\mathrm{Gd}) = 10$ Å, close to the observed anomalies in the T_C vs $t(\mathrm{Gd})$.[9] In Nb/Fe, the nonmonotonous dependence was attributed to a loss of ferromagnetic order for small Fe layer thicknesses.[12] To clarify the role of these different mechanisms it is interesting to investigate the behavior of a superconductor/ferromagnet system in which the effects of π coupling can be ruled out, and the changes in magnetic order can be tuned in a controlled way through a well defined magnetic transition.

In this work, we have studied the superconducting properties of superconductor/ferromagnet bilayers, where the superconductor is a Nb film and the ferromagnet is an Fe/Cu multilayer. In these bilayers the effect of π coupling is ruled out by the geometry since there is a single superconducting layer in the structure. Changing the Fe layer thickness $t(\mathrm{Fe})$ in the multilayer, a structural and magnetic transition can be induced in the Fe layers from fcc γ-Fe to bcc α-Fe.[17] The fcc γ-Fe phase is stable only for thin films ($\leqslant 10$ Å), favored by the strain due to the fcc Cu layers whereas for large $t(\mathrm{Fe})$, the Fe layers grow in the usual ferromagnetic bcc phase which has a much higher Curie temperature (T_{Curie}). This transition in the magnetic layer has a clear influence on the superconducting properties of the Nb film, giving rise to a nonmonotonic T_C dependence. This indicates that the material with higher T_{Curie}, i.e., with stronger magnetic order, produces a weaker ferromagnetic proximity effect, suggesting that spin-flip scattering is the dominant pair breaking mechanism.

Nb/[Fe/Cu] multilayers were prepared on Si(100) substrates at room temperature. The samples have been grown

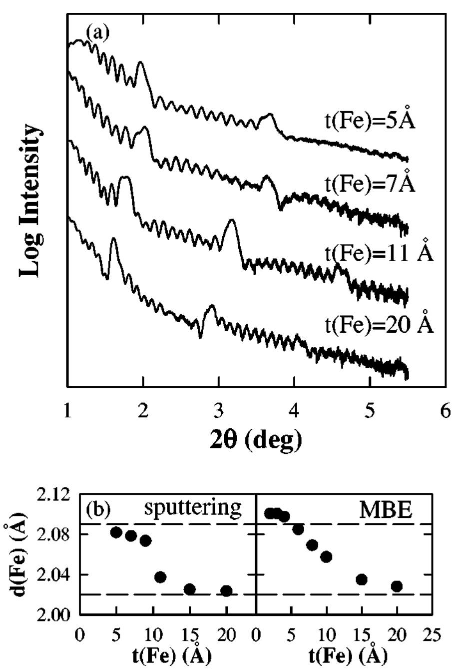

FIG. 1. (a) Low angle x-ray-diffraction scans of several $[\mathrm{Fe}(t)/\mathrm{Cu}(42)]_8/\mathrm{Nb}(200)$ multilayers grown by sputtering. The Fe layer thickness is varied from $t=5$ Å to $t=20$ Å. (b) Lattice parameter of the Fe layer as a function of the Fe layer thickness for two series of samples grown by sputtering and MBE. These values are calculated from the high angle diffraction data as indicated in the text. Dashed lines correspond to the constant values $d=2.09$ Å and $d=2.02$ Å.

either by dc sputtering or molecular beam epitaxy (MBE) to compare the results from these two different growth techniques. Each sample begins with an $[\mathrm{Fe}(t)/\mathrm{Cu}(42\ \text{Å})]_8$ multilayer. The Fe thickness $t(\mathrm{Fe})$ varies in the range $t(\mathrm{Fe})=0$–25 Å. Then a 200 Å Nb film is deposited on top of the last Fe layer. This thickness is of the order of the superconducting coherence length ξ_0, estimated from dH_{c2}/dT at T_C as 125 and 95 Å for the sputtered and MBE samples, respectively. Finally the sample is covered with a capping layer to prevent oxidation (40 Å of Cu for the sputtering samples and 40 Å of Ag for the MBE ones). Similar series of samples were also grown with thicker Nb layers up to 500 Å. Each series of multilayers used for the study of the T_C dependence on $t(\mathrm{Fe})$ was prepared in the same run in order to avoid uncertainties due to the possible scatter in T_C between samples grown in different runs. It is important to characterize the structural and magnetic transition from γ Fe to α Fe in these multilayers as the particular thickness at which it occurs depend on the growth conditions and $t(\mathrm{Cu})$.[17–21] This has been done by low and high angle x-ray diffraction using a rotating anode Rigaku diffractomer with Cu $K(\alpha)$ radiation and superconducting quantum interference device magnetometry up to 10 kOe in the 10 – 300 K temperature range. The superconducting transitions were obtained from four lead transport measurements (R vs T curves) in a helium cryostat and dc susceptibility with a 10-Oe field perpendicular to the sample plane.

Figure 1(a) presents the low angle θ–2θ x-ray scans for a series of $\mathrm{Nb}(200\ \text{Å})/[\mathrm{Fe}(t)/\mathrm{Cu}(42\ \text{Å})]_8$ multilayers grown by sputtering. This graph indicates the high quality of the multilayer structure: clear superlattice peaks arise from the Fe/Cu multilayer up to the third order and finite-size effect oscillations, corresponding to the total thickness of the sample, appear up to $2\theta=5°$. The superlattice parameter $\Lambda=t(\mathrm{Fe})+t(\mathrm{Cu})$ and the total thickness of the sample obtained from the low angle diffraction data are in good agreement with nominal values derived from deposition rates.

Figure 1(b) is a plot of the perpendicular lattice parameter $d(\mathrm{Fe})$ of the Fe layers as a function of Fe layer thickness for two series of samples grown by sputtering and MBE. This lattice parameter $d(\mathrm{Fe})$ extracted from the high angle peak position of the superlattice, arises from the weighted average of the constituents lattice parameters.[17,22] For the Cu layers, which are always much thicker than the Fe ones, the bulk value $d_{111}(\mathrm{Cu})=2.087$ Å has been used since they can be assumed to be unstrained. In both types of samples, the Fe lattice parameter is around 2.09 Å for small $t(\mathrm{Fe})$ and saturates close to the value for bulk bcc α-Fe $d_{110}(\mathrm{Fe})=2.02$ Å for large Fe layer thickness. This change in $d(\mathrm{Fe})$ is consistant with a structural transition from γ-Fe to α-Fe.[17] The lattice parameter for thin Fe layers $d(\mathrm{Fe})=2.09$ Å is an intermediate value between $d_{111}(\gamma\text{-Fe})=2.071$ Å, extrapolated at 295 K for bulk antiferromagnetic γ-Fe,[23] and $d_{111}(\gamma\text{-Fe})=2.102$ Å, corresponding to the theoretical prediction for ferromagnetic γ-Fe.[24] It is worth to note that the lattice parameter of the Nb layer is independent of the Fe layer thickness, $d(\mathrm{Nb})=2.332\pm0.007$ Å, indicating that there is not any significant change on the strain field on the Nb at this structural transition.

It is necessary to characterize the magnetic properties of these Fe/Cu superlattices, since the magnetic order in γ-Fe is strongly dependent on the unit cell volume, and it can exist in a nonmagnetic, antiferromagnetic, or ferromagnetic state.[24,25]

The low temperature in plane hysteresis loops are typical of a ferromagnet, with a $\sim$100 Oe coercive field and $\sim$2 kOe saturation field (see inset of Fig. 2). In Fig. 2, the temperature dependence of the saturation magnetization M_S at 4 kOe, normalized by the low-temperature value $M_S(10\ \mathrm{K})$, has been plotted for a series of representative $\mathrm{Nb}(250\ \text{Å})/[\mathrm{Fe}(t)/\mathrm{Cu}(42\ \text{Å})]_8$ sputtered multilayers. For the samples with thicker Fe layers, the decrease in $M_S(T)$ is small, as expected for α-Fe layers with $T_{\mathrm{Curie}}=1043$ K, whereas for $t(\mathrm{Fe})\leqslant7$ Å, M_S is strongly temperature dependent so that $M_S(300\ \mathrm{K})$ is close to zero. For these latter samples, T_{Curie} can be estimated from the M_S^2 vs T plot[26] in order to eliminate the high-temperature tail of the curve due to the finite magnetic field used in the measurement. The obtained values are around 210 K, in good agreement with reported values for ferromagnetic γ-Fe.[18] The sample with $t(\mathrm{Fe})=9$ Å presents an intermediate behavior suggesting that the structural and magnetic transition is taking place in the range $t(\mathrm{Fe})=8$–10 Å.

The drastic reduction of the Fe/Cu superlattices T_{Curie} at the magnetic transition has a clear influence on the Nb layer properties. In Fig. 3, the superconducting transition temperature T_C of several sputtered $\mathrm{Nb}(200\ \text{Å})/[\mathrm{Fe}(t)/\mathrm{Cu}(42\ \text{Å})]_8$ multilayers has been plotted as a function of the Fe layer thickness. The inset shows a typical $\chi_{dc}=M/H$ vs T curve for one of these samples that has been used to obtain T_C

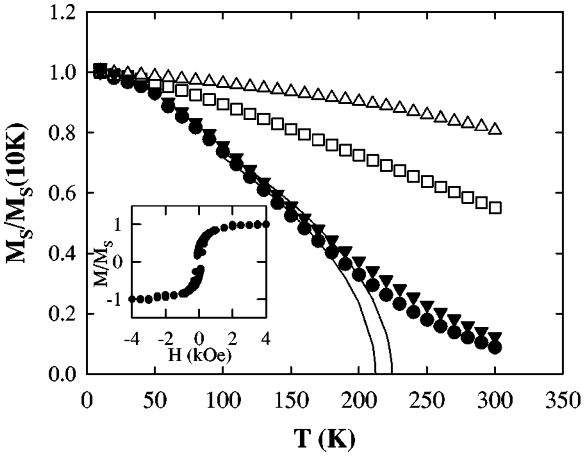

FIG. 2. Temperature dependence of the saturation magnetization, measured with $H=4$ kOe parallel to the sample plane, for several $[\mathrm{Fe}(t)/\mathrm{Cu}(42)]_8/\mathrm{Nb}(250)$ multilayers grown by sputtering. Filled symbols correspond to multilayers with fcc γ-Fe (circles, $t=5$ Å; triangles, $t=7$ Å) and hollow symbols to multilayers with bcc α-Fe (squares, $t=9$ Å; triangles, $t=11$ Å). Solid lines are the fits used to extrapolate T_{Curie}. Inset shows the hysteresis loop of an $[\mathrm{Fe}(7)/\mathrm{Cu}(42)]_{10}$ superlattice measured at 10 K.

from the onset of the diamagnetic signal. In all the samples the T_C is depressed in comparison with a single 200-Å Nb film grown in the same conditions, $T_C(\mathrm{film})=6.04$ K, $\Delta T_C \simeq 0.1$ K,[27] due to the proximity effect with the magnetic layer. The most interesting feature in this graph is a clear increase of more than 1 K in T_C observed in the range $7.5 \leqslant t(\mathrm{Fe}) \leqslant 9$ Å, close to the magnetic transition. As the Fe layer thickness increases beyond this point, T_C saturates at about 5 K.

The same qualitative enhancement in T_C at the magnetic transition is also observed in the MBE grown multilayers, as shown in Fig. 4. These Nb (200 Å)$[\mathrm{Fe}(t)/\mathrm{Cu}(42\text{ Å})]_8$ samples present sharp superconducting transitions, with a 90%–10% width of the order of 0.1 K, much smaller than

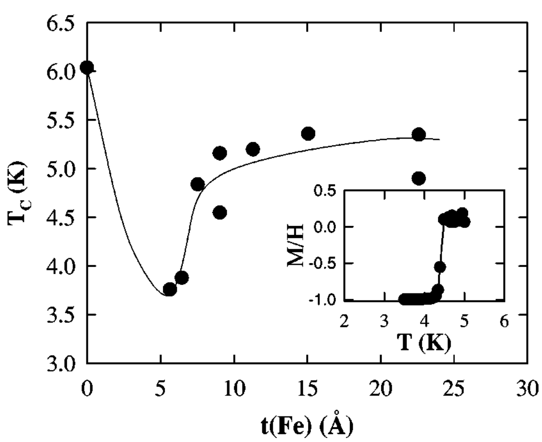

FIG. 3. Superconducting transition temperature of sputtered $[\mathrm{Fe}(t)/\mathrm{Cu}(40)]_8/\mathrm{Nb}(200)$ multilayers as a function of the Fe layer thickness, obtained from the onset of the diamagnetic signal. Solid line is a guide to the eye. Inset shows a typical dc susceptibility $\chi_{dc}=M/H$ vs temperature curve of one of these multilayers.

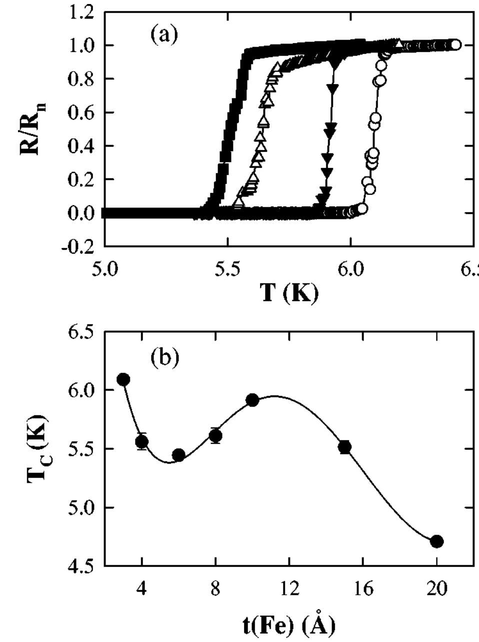

FIG. 4. Superconducting transition temperature of MBE grown Nb(200 Å)/$[\mathrm{Fe}(t)/\mathrm{Cu}(42\text{ Å})]_8$ multilayers as a function of Fe layer thickness. Symbols indicate the midpoint of the resistivity transition and error bars correspond to 10–90% transition width. Solid line is a guide to the eye.

the changes in T_C induced by the proximity effect of the magnetic layers (see Fig. 4). The superconducting transition has a minimum for $t(\mathrm{Fe})=6$ Å, and a ~ 0.5 K increase around $t(\mathrm{Fe})=10$ Å when the Fe layers change from fcc to bcc. Finally, as the bcc α-Fe layers become thicker, the usual decrease in T_C is found.

Nb/[Fe/Cu] multilayers with thicker Nb layers (300 and 500 Å) exhibit only a monotonous decrease of T_C. Therefore this increase in T_C vs $t(\mathrm{Fe})$ is only found if the Nb layer thickness is comparable to the superconducting coherence length ξ_0.

The experimental behavior of Nb/[Fe/Cu] multilayers shown in Figs. 3 and 4 exhibit a nonmonotonic dependence as a function of $t(\mathrm{Fe})$ with a clear enhancement in the Nb superconducting transition temperature when the T_{Curie} of the magnetic layers change from 210 K (fcc γ-Fe) to 1043 K (bcc α-Fe).

The usual approach to the ferromagnetic proximity effect of metallic superconducting/ferromagnet multilayers has been to consider the polarization of conduction electrons due to the exchange field in the ferromagnet as the main pair breaking mechanism,[13–15] which has been used to fit the behavior in some systems.[9–11] In this model, the proximity effect is characterized by a parameter ϵ, inversely related to the exchange energy J_{ex} in the magnetic material, so that superconductivity is suppresed strongly for higher values of J_{ex}. Therefore this theory would predict a reduction in the T_C of the Nb/[Fe/Cu] multilayers at the magnetic transition. The exchange energy is proportional to T_{Curie} and it increases by almost a factor of 5 at the transition, so that pair breaking by the exchange field should be enhanced. This is opposite to the observed experimental behavior described above and therefore this model cannot explain the results.

Another pair breaking mechanism that can play an important role in the proximity effect of magnetic materials is by spin-flip scattering processes between the Cooper pairs and the magnetic atoms. This effect has been proposed to be particularly relevant in the case of layered superconductors, in which the magnetic atoms are located out of the plane where transport takes place.[28] These processes are characterized by total spin conservation in the scattering event, so that the spin of the magnetic atom must flip when the Cooper pair is broken. Therefore as the spin-flip scattering is hindered by correlations between the magnetic atoms it becomes less effective for the material with larger exchange energy, i.e., higher T_{Curie}. Then, within this framework, even though there is not a fit to a rigorous model, the anomalous increase in T_C with $t(\mathrm{Fe})$ may be qualitatively explained as due to a pair breaking mechanism by spin-flip processes.

In summary, we have studied the influence of magnetic order in the superconducting properties of Nb(200 Å)/[Fe(t)/Cu(42 Å)]$_8$ multilayers grown by sputtering and MBE. In both type of samples an increase in the superconducting transition temperature is found when the T_{Curie} of the [Fe/Cu] multilayers change drastically from 210–1043 K. This behavior implies a more efficient magnetic proximity effect for the material with the lower Curie temperature, i.e., the pair breaking effect of the magnetic atoms is weakened by the stronger magnetic order. These results suggest that spin-flip scattering processes play a dominant role in the suppression of superconductivity by the magnetic material.

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