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Critical-temperature-oscillations dependence on Mn concentration in superconducting Nb/CuMn multilayers

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We have measured, at different Mn concentrations, the superconducting critical temperature T_c of Nb/CuMn multilayers in the cases both where CuMn layer thickness was varied with fixed Nb layer thickness and, vice versa, keeping constant CuMn and varying Nb layer thicknesses. The T_c values exhibit an oscillating behavior strongly dependent upon the Mn concentration. Several models have been discussed to interpret our measurements. The analysis of the experimental data seems to suggest the presence of a π phase difference of the superconducting order parameter across neighboring Nb layers driven by the magnetic nature of the CuMn alloy. [S0163-1829(98)02822-7]

INTRODUCTION

The influence of magnetism on superconductivity gives rise to interesting phenomena that can be easily investigated on artificially layered structures where superconducting (S) layers alternate with magnetic (M) layers. Recently, the observation of oscillations of the superconducting transition temperature T_c versus the thickness of the magnetic layers has been reported on Nb/Gd multilayers (superconducting/ferromagnetic multilayers[1]) and on Nb/Gd/Nb trilayers.[2] The T_c oscillations have been explained in terms of a time irreversible pair-breaking mechanism and the theoretical model used[3] deals with the spatial variation of the anomalous Green function F, describing the condensate of pairs, in the presence of an exchange field I. A nontrivial superconducting ground state is found: The phase difference φ of the superconducting order parameter between two neighboring S layers will no longer be only 0, but can take a value between 0 and π, depending on the M layer thickness. An intrinsic phase difference $\varphi=\pi$ was also proposed for junctions with magnetic impurities in the tunnel barrier, the so-called π junctions,[4] and for weak links of superconductors with d-wave pairing.[5]

A nonmonotonic T_c behavior was also observed by Mühge *et al.* in Fe/Nb/Fe trilayer samples with varying d_{Fe} at fixed d_{Nb}.[6] They attribute this behavior to the existence of magnetically dead Fe layers at the interfaces and to the change of the effective electron-electron attractive interaction in these layers at the onset of ferromagnetism for $d_{\mathrm{Fe}} \geqslant 7$ Å.

In a previous work, we have observed oscillations of T_c versus the thickness of the nonsuperconducting layers in Nb/CuMn multilayers,[7] where the nonsuperconducting material CuMn is a well known metallic spin glass.[8,9] The superconducting/spin-glass systems allow us to investigate the coexistence of superconductivity and magnetism in a wider range of S and M thicknesses because of the weaker pair-breaking effect of a spin glass when compared to a ferromagnet. Moreover, in the case of Nb/CuMn multilayers the strength of the exchange field I can be easily varied by changing the Mn percentage in the magnetic layers.

In Ref. 7 we explained the observed T_c oscillations by the presence of a time irreversible pair-breaking mechanism also in the case of superconducting/spin-glass multilayers. In particular, we considered two series of Nb/CuMn multilayers with two different Mn concentrations (0.7% and 1.3%), both with a fixed Nb layer thickness (d_{Nb}) of 250 Å and with varying CuMn layer thickness (d_{CuMn}). All the experimental data were well explained in terms of the Radovic *et al.* theory[3] developed in the case of specular electronic scattering at the Nb/CuMn interfaces.

Nevertheless, to better check the applicability of this theory to the superconducting/spin-glass case and to have an accurate comparison with previous data in Refs. 1, 2, and 6, we have decided to perform measurements on different series of Nb/CuMn, with different Mn concentrations and different fixed Nb layers varying d_{CuMn} on one side and with varying Nb layer thickness keeping constant d_{CuMn} on the other side. In the series with varying d_{CuMn} at different Mn concentrations we have observed oscillations in the $T_c(d_{\mathrm{CuMn}})$ curves, with the presence of several relative maxima and minima. We discuss different possible interpretations of this effect. Even though the agreement with the experimental data is worse at higher Mn concentrations, the model that predicts the presence of a phase difference in the superconducting order parameter between adjacent superconducting layers explains some of the observed features, with the behavior of the fitting parameters in the different series in good agreement with that expected on the basis of physical considerations. For the sake of generality, the theoretical analyses in this work have been performed using the more general case where a finite transparency electronic coefficient of the S/M interfaces has been assumed.

We point out that the model in Ref. 3 applies to symmetric structures, while our multilayers always begin with a CuMn layer and terminate with a Nb layer. However this asymmetry should not invalidate the applicability of the theory to our data because all our samples are made of ten bilayers and, moreover, the observed superconducting properties are related to the overall sample (we are considering strongly coupled bidimensional superconducting systems[10]).

MEASUREMENTS AND DISCUSSION

The samples were grown on Si(100) substrates by a dual-source magnetically enhanced dc triode sputtering system,

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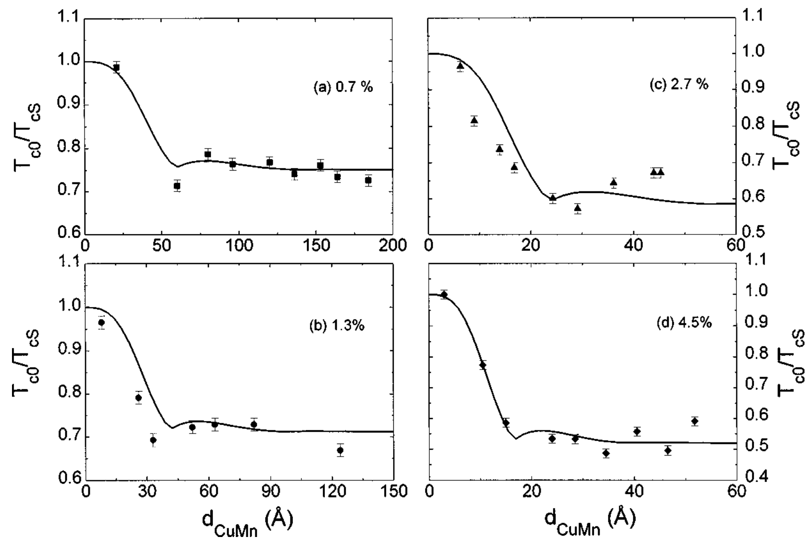

FIG. 1. Reduced transition temperatures T_{c0}/T_{cS} vs d_{CuMn} curves for the series (a) M22295 (%Mn=0.7, d_{Nb}=250 Å), (b) M16295 (%Mn=1.3, d_{Nb}=250 Å), (c) M4696 (%Mn=2.7, d_{Nb}=260 Å), and (d) M191095 (%Mn=4.5, d_{Nb}=250 Å). The solid lines are the best fit curves obtained with Eq. (1).

with a movable substrate holder. The deposition conditions were similar to those of the multilayers described previously and the fabrication technique is the same with a complete series obtained in only one deposition run.[7] We point out that in the series with varying d_{CuMn} there is always a single Nb sample from which we deduce the electrical and superconducting properties of the Nb layers present in the samples of the series.

Energy dispersive spectroscopy and low-angle x-ray (LXR) analyses have been performed to determine the thicknesses and the samples quality. In particular, LXR analyses have shown in our samples a typical interfacial roughness of ~5 Å.

The superconducting transitions of all the samples were measured resistively using a standard dc four-probe technique. The ratios $\rho_N(300\ \mathrm{K})/\rho_N(10\ \mathrm{K})$, with ρ_N the normal state resistivity, were in the range 1.5–2.0 for all the series, with a spread of values in the same series of less than 5%, confirming the high uniformity of the transport properties in samples obtained in the same deposition run.

In Fig. 1 we show the reduced transition temperatures T_{c0}/T_{cS} versus d_{CuMn} (T_{c0} is the temperature at which the electrical resistance R of the sample becomes less than $10^{-4}\ \Omega$ and T_{cS} is the single Nb sample transition temperature) for the samples of the four series with Mn concentrations 0.7%, 1.3%, 2.7%, and 4.5% and $d_{Nb}\simeq 250$ Å. Depending on the Nb layer thickness, the T_{cS} values were generally in the range 6.5–8.0 K. An oscillatory behavior of T_{c0}/T_{cS} vs d_{CuMn} is present in the measured series outside the experimental error, with the presence of several minima and maxima.

Such an oscillating behavior of the critical temperature T_c vs the thickness of the magnetic layers d_M is similar to that observed in Nb/Gd multilayers and Nb/Gd/Nb trilayers, which has been interpreted in terms of the Radovic *et al.* theory. A nonmonotonic behavior in the $T_c(d_M)$ curves has also been reported by Mühge *et al.*[6] in the case of Fe/Nb/Fe trilayers. The authors have related the minimum in T_c at $d_M \sim 7$ Å to the onset of ferromagnetism in the Fe layers. A similar explanation could be tried in the case of our Nb/CuMn multilayers observing that, due to the finite-size effect,[9] the spin-glass freezing temperature T_g of our CuMn layers is influenced by the thickness value d_{CuMn}. At low d_{CuMn} values, one has $T_c \geqslant T_g$ with the CuMn layers being in a paramagnetic state at the superconducting transition. In this regime T_c should monotonically decrease with d_{CuMn}.[11,12] With increasing d_{CuMn} values, T_c drops below T_g and the CuMn layers are in the spin-glass state at the superconducting transition. An increase or saturation of the $T_c(d_{CuMn})$ curve could be expected in this case, with a minimum followed by a maximum with a monotonic decrease of T_c at higher d_{CuMn} values.

In our samples, at low d_{CuMn} values, the $T_c(d_{CuMn})$ curves present a downward curvature (series with 0.7%, 1.3%, and 4.5% of Mn). This behavior is well described by the Radovic *et al.* theory, while a paramagnetic pair-breaking mechanism[11,12] generally gives an upward curvature in this limit. Moreover, the Werthamer-Hauser theory[12] cannot explain the strong decrease of the critical temperature for small d_{CuMn}.[11] On the other hand, direct measurements of T_g are very difficult to perform on our multilayers, expecially in the low d_{CuMn} value region, due to the small amount of magnetic material in the samples.

Critical magnetic field H_{c2} measurements in all our samples[13] performed in both the perpendicular and parallel directions with respect to the plane of the films, have never shown any sign that could be related to some magnetic transition when the sample is in the superconducting state. Only the well known transition from a three-dimensional (linear) to a bidimensional (square-root-like) behavior of the H_{c2} versus temperature curves has been observed, when expected, in the parallel case.[14,13]

The Radovic *et al.* theory[3] foresees the presence of multiple oscillations in the T_c vs d_M curve and was successfully used to explain the data with 0.7% and 1.3% of Mn.[7] Therefore, we have tried to explain also our present experimental data, taken at different Mn concentrations, using this model.

TABLE I. Fabrication and fitting parameter values.

Parameter \ Series	M22295	M16295	M23596	M4696	M25996	M201095	M191095	M251095	M2596
d_{Nb} (Å)	250	250	360	260	165	350	250	150	300
%Mn	0.7	1.3	2.7	2.7	2.7	4.5	4.5	4.5	7
ξ_M (Å)	50	38	19	19	19	14	14	14	11
$\eta\xi_S^2$ (Å^2)	130	122	100	90	230	70	56	60	38
χ_0^2	0.79	4.19	12.59	9.51	0.29	1.17	2.17	12.88	2.15

The solid lines in Fig. 1 are the best fit curves obtained using the relation

$$\frac{T_c}{T_{cS}}=1-12G_0(x)\,\frac{\eta\xi_S^2}{d_M d_S}, \qquad (1)$$

where $d_{M,S}$ are the magnetic (M) and superconducting (S) layer thicknesses, η is a phenomenological parameter related to the S/M interface transparency for the Cooper pairs, $\xi_S = \sqrt{D_S/2\pi T_{cS}}$ is the coherence length in S (D_S is the diffusion coefficient in S), and finally $G_0(x)$ is the minimum of $G_0(x,\varphi)$ with respect to $\varphi\in[0,\pi]$, where $x=d_M/\xi_M$ and $\xi_M=\sqrt{4D_M/I}$ (D_M is the diffusion coefficient in M and I the exchange energy). The expression of the function $G_0(x,\varphi)$ is

$$G_0(x,\varphi)=2\left[\sin^2\frac{\varphi}{2}\,S(x)+\cos^2\frac{\varphi}{2}\,T(x)\right],$$

with

$$S(x)=x\,\frac{\sin 2x+\sinh 2x}{\cosh 2x-\cos 2x}, \quad T(x)=x\,\frac{-\sin 2x+\sinh 2x}{\cos 2x+\cosh 2x}.$$

Expression (1) is a generalization of the equation deduced in Ref. 15 and used in our previous report.[7] It has been obtained by using the boundary conditions for the Green function F at S/M interfaces

$$F_S=F_M, \quad \frac{dF_S}{dx}=\eta\,\frac{dF_M}{dx},$$

where $\eta=\sigma_M/\sigma_S$ only in the specular scattering case and $\sigma_{M,S}$ are the normal state conductivities of M and S layers. The parameter in Ref. 15, $\gamma=\sigma_M\xi_S/\sigma_S\xi_N$, is substituted now by $\gamma=\eta\xi_S/\xi_N$ (here $\xi_N=\sqrt{D_M/2\pi T_{cS}}=\xi_M\sqrt{I/8\pi T_{cS}}$). In the limit of small γ, which is our case, as we will see *a posteriori*, T_c is given by expression (1). Depending on which is larger between $S(x)$ and $T(x)$, the 0 phase $[S(x)>T(x),\ \varphi=0]$ or the π phase $[S(x)<T(x),\ \varphi=\pi]$ will be present in the sample and this switch from one phase to another gives rise to the oscillatory T_c versus d_M behavior.

Expression (1) has two fitting parameters ξ_M and $\eta\xi_S^2$, while T_{cS} is determined by measuring the R versus T curve of the single Nb sample of the series. The fabrication and fitting parameter values of all the series are collected in Table I.

In Fig. 2 we show the T_{c0}/T_{cS} vs d_{CuMn} curves of three different series with the same Mn concentration (4.5%) and with different fixed Nb layer thicknesses (d_{Nb}=150, 250, and 350 Å). We observe that the thinner the d_{Nb} the stronger the T_c suppression and the more rapid its drop with increasing d_{CuMn} in the range of few angstroms. In particular, in the series with d_{Nb}=150 Å, no oscillations of T_c were observed down to the limit of our lowest obtainable temperature of 1.5 K. The solid lines in Fig. 2 are the best fit curves obtained by the discussed model, which describes well also the case where the T_c oscillation was not observed. Similar results have been obtained in the series with 2.7% of Mn and in the series with 7% of Mn.

In Table I the values of

$$\chi_0^2=\frac{1}{\nu}\sum_{i=1}^{N}\frac{\left[\left(\frac{T_c}{T_S}\right)_{\rm expt}-\left(\frac{T_c}{T_S}\right)_{\rm th}\right]^2}{\left(\frac{\Delta T_c}{T_S}\right)^2}$$

are reported, where $\Delta T_c/T_S$ is the experimental error on the measured $(T_c/T_S)_{\rm expt}$ values, the $(T_c/T_S)_{\rm th}$ values are obtained by Eq. (1), and ν is the number of degrees of freedom. The agreement between the experimental data and the theoretical curves seems to be satisfactory in many series. The model used is developed for the case of ferromagnetic materials, while CuMn is a well known metallic spin glass. Moreover, a change in the CuMn magnetic behavior is expected with increasing Mn percentage.[16]

The series with the same Mn concentration are characterized by the same ξ_M value. We have obtained ξ_M=19 Å for the three series with 2.7% of Mn and ξ_M=14 Å for the three

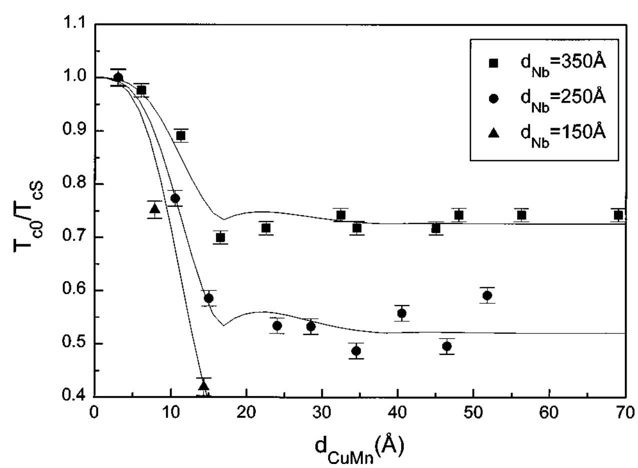

FIG. 2. T_{c0}/T_{cS} vs d_{CuMn} curves for the series M201095 (d_{Nb} =350 Å), M191095 (d_{Nb}=250 Å), and M251095 (d_{Nb}=150 Å), all with 4.5% of Mn. The solid lines are obtained with Eq. (1).

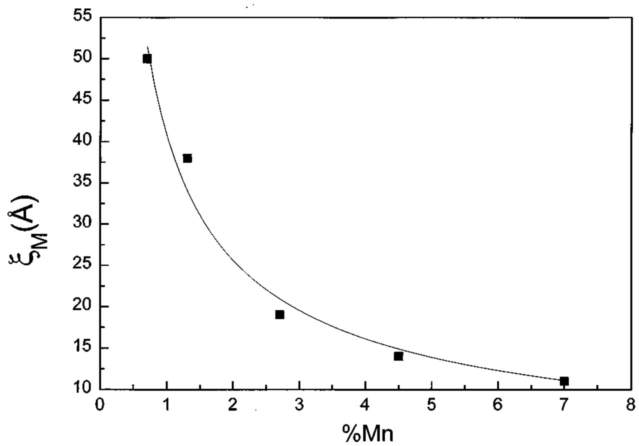

FIG. 3. Fit parameter ξ_M vs Mn concentration behavior. The solid line is a power-law fit with the dependence $\xi_M \propto (\%\mathrm{Mn})^{-0.67}$.

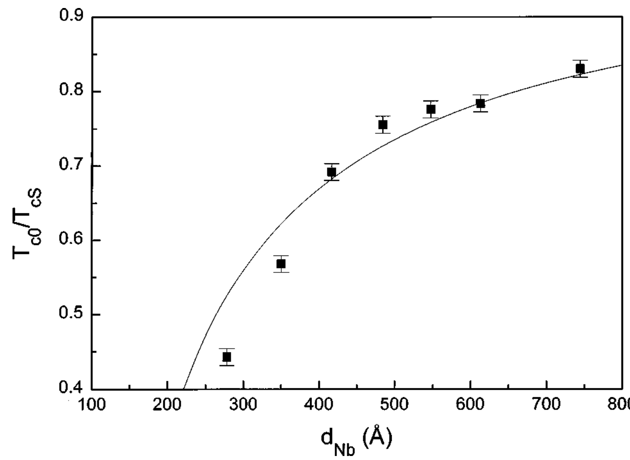

FIG. 4. T_{c0}/T_{cS} vs d_{Nb} curve for the series M211096 (d_{CuMn}=30 Å). The solid line is the best fit curve obtained with Eq. (1).

series with 4.5% of Mn. To do a comparison, a value of $\xi_M = 13$ Å was found in Nb/Gd (Ref. 1) and of $\xi_M = 6$ Å in V/Fe.[17] It is difficult to estimate the error affecting the quantity ξ_M. On the other hand, variations of a few angstroms in ξ_M give fitting curves in strong disagreement with our data. By this procedure, we have been able to estimate the relative error on all the ξ_M values to be $\pm 7\%$. In addition to the absolute value of ξ_M, which depends also upon the diffusion coefficient, it is interesting to observe the ξ_M behavior with Mn concentration plotted in Fig. 3: ξ_M decreases with increasing Mn concentration. From the definition given above, ξ_M is proportional to $I^{-1/2}$. Knowing the I dependence on the Mn concentration in the CuMn, we can compare directly the observed ξ_M dependence on the Mn percentage with that expected from the definition of ξ_M given in the Radovic *et al.* model.

The experimental behavior of the freezing transition temperature T_g versus magnetic impurity concentration (see, for example, Ref. 8) can give information about the qualitative I dependence on the spin-glass composition. The nearly linear behavior of T_g vs the Mn percentage, at least for Mn concentrations lower than 8%, can be translated into a qualitatively linear behavior of I vs the Mn concentration.

Therefore, by fixing the Mn percentage, we fix the exchange energy and this explains our result that ξ_M is the same for all the series with the same Mn concentration. To be more specific, we have tried to fit the data of ξ_M versus the Mn concentration with the dependence $\xi_M \propto (\%\mathrm{Mn})^b$ (solid line in Fig. 3), obtaining $b = -0.67 \pm 0.07$, a value close to the -0.5 exponent expected in the case of the linear behavior of I vs the Mn percentage.

The exchange energy I (and then also the parameter ξ_M) depends on the magnetic material thickness because of the finite-size effect.[9] Therefore, the I value changes from one sample to another in the same series, while in the Radovic *et al.* model I is assumed to be constant. This implies that the value of ξ_M we have used in a series is a kind of average parameter.

As for the second fitting parameter $\eta \xi_S^2$, by measuring the temperature dependence of the critical magnetic field perpendicular to the film plane, $H_{c2\perp}(T)$, we can extract the ξ_S value, using the relation $\xi_S = 2\xi_\parallel(0)/\pi$, with $\xi_\parallel(0)$ the zero-temperature Ginzburg-Landau coherence length in the direction parallel to the film plane.[18] This measurement performed on the Nb sample of the series M4696 with 2.7% of Mn has given $\xi_S \simeq 40$ Å, consistent with the high resistivity values ($\rho \simeq 10^{-7}\ \Omega$ m). The same measurements done on all the samples of this series have given ξ_S values in the range 40–70 Å. Therefore, for this series, we can deduce from the value of the fitting parameter $\eta \xi_S^2 = 90$ Å^2 a value of $\eta \simeq 0.06$. On the other hand, the resistivity measured values of single Nb and CuMn films give rise to a ratio σ_M/σ_S of the order of 1. Then, for this series we have obtained $\eta \neq \sigma_M/\sigma_S$, which implies a nonspecular scattering at S/M interfaces and justifies the initial assumption of a finite transparency electronic coefficient at the S/M interfaces.

In all the measured samples the ξ_S were in the range 40–180 Å, giving values of η of the same order of magnitude as that obtained for the series M4696. These values are generally higher than those found in superconducting/ferromagnetic multilayers ($\eta = 0.013$ in V/Fe).[17]

Once we know the values of η, ξ_S, and ξ_M, we can check *a posteriori* that we are effectively in the small γ regime. For example, in the series M4696 it is $\eta \xi_S/\xi_M \simeq 0.12$ and then it will be $\gamma = \eta \xi_S/\xi_N < 0.12$, with $\xi_M < \xi_N$ (ξ_M is the characteristic penetration length of Cooper pairs in magnetic layers and ξ_N is the corresponding length in a normal metal).

Finally, we discuss the measurements performed on the series with fixed d_{CuMn} and varying d_{Nb}. Also these data are consistent with the Radovic *et al.* scenario. In Fig. 4 we plot T_{c0} versus d_{Nb} for the series with varying d_{Nb}, fixed $d_{\mathrm{CuMn}} = 30$ Å, and a Mn concentration of 2.7%. T_{c0} decreases strongly with decreasing Nb layer thickness. The solid line is again obtained with Eq. (1). In the case of varying d_{CuMn} and fixed d_{Nb}, with varying $x = d_M/\xi_M$, we went from the $\varphi = 0$ to the $\varphi = \pi$ state. Now, d_M is fixed as well as the phase φ for all the samples. We want to remark that from the best fit curves we have again obtained $\xi_M = 19$ Å as for all the series with 2.7% of Mn with varying d_{CuMn}. The value of the second fitting parameter is $\eta \xi_S^2 = 114$ Å^2. Assuming $\eta \simeq 0.06$ also for this series, we obtain $\xi_S \simeq 42$ Å, of the same order as for the M4696 series.

CONCLUSIONS

We have observed an oscillating behavior of T_{c0} versus d_{CuMn} in superconducting (Nb)/spin-glass (CuMn) multilayers with different Mn concentrations and different d_{Nb} values. This behavior, together with that of T_{c0} versus d_{Nb}, could be explained by means of a time irreversible pair-breaking mechanism, assuming a finite electronic transparency coefficient at the S/M interfaces. In fact, from the best fit curves of the T_c vs d_{CuMn} data, we have obtained the same value of the ξ_M fitting parameter for all the series with the same Mn concentration and with different d_{Nb} fixed values. Moreover, the ξ_M versus Mn concentration behavior was in reasonable agreement with that expected on the basis of simple physical assumptions.

In the series where d_{Nb} was varied keeping d_{CuMn} fixed, the T_{c0} versus d_{Nb} behavior was again fitted by the same ξ_M value obtained in the case of the series with the same Mn concentration but fixed d_{Nb}. These observations seem to indicate as plausible the extension of the Radovic *et al.* theory to the case of our Nb/CuMn multilayers. A final confirmation of the validity of this theory in the case of superconductor/spin-glass multilayers could be given by experiments sensitive to the phase of the superconducting order parameter, similar to those probing the pairing symmetry in high-T_c superconductors.

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