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Superconducting-critical-temperature oscillations in Nb/CuMn multilayers

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We have observed a nonmonotonic behavior of the superconducting critical temperature T_c versus the CuMn layer thickness in multilayers of Nb/CuMn with fixed Nb layer thickness ($\sim$250 Å). The results have been interpreted assuming the presence of the π phase in the case of layered superconducting systems with weak magnetic coupling, where the phase ϕ of the superconducting order parameter can vary in the range $0<\phi<\pi$ between adjacent superconducting layers. [S0163-1829(96)05721-9]

The coexistence of superconductivity and magnetism has been largely studied in the last thirty years and lately, because of the role that it seems to play in many properties of the high-temperature superconducting compounds,[1] this issue has gained even more interest.

Recently,[2] nontrivial superconducting ground state has been observed in Nb/Gd multilayers, where the local exchange field in the ferromagnetic layers is assumed[3,4] to produce a phase shift $\Delta\phi$ in the superconducting order parameter in adjacent Nb layers. Depending upon the magnetic layer thickness, a state with $\Delta\phi=\pi$, between neighboring superconducting layers, can have a higher superconducting critical temperature T_c than the ordinary state with $\Delta\phi=0$. As a result, T_c shows a nonmonotonic behavior when the Gd layer thickness is varied.

In recent years,[5] many experiments on high-T_c superconducting junctions have been interpreted in terms of weak links made from superconductors with d-wave pairing,[6] in the presence of the so-called π junctions. The existence of such π junctions was originally proposed by Bulaevskii, Kuzii, and Sobyanin[7] in the case of junctions with magnetic impurities in the barrier and was related to spin flip assisted coherent tunneling.

Although the π phase analyzed by Buzdin and Kupriyanov,[3] and by Radovic *et al.*[4] neglects all the dynamical spin processes, it is interesting to study its presence in other multilayered superconducting and/or magnetic systems to better clarify the nature of the superconducting ground state in these conventional low-T_c materials with reduced dimensionality which can be used as simple models to compare different theories related to high-T_c superconductors.

The superconducting and/or spin-glass multilayers are an interesting class of artificially layered materials and, due to the reduced pair-breaking effect in the magnetic layers when compared to the ferromagnetic case, allow to perform investigations of the simultaneous presence of superconductivity and magnetism in a much wider range of relative thicknesses.

Among several superconducting–spin-glass systems we have focused on Nb/CuMn multilayers because Nb is the single element with the highest critical temperature, $T_c=9.2$ K, CuMn is a well-known metallic spin-glass[8] and in literature there is a large number of works about CuMn-based and Nb/Cu multilayers.[9,10] In the obtained samples we have observed T_c oscillations with the CuMn layer thickness at a fixed Nb layer thickness. These results give evidence of the presence of the π phase on our multilayers.

The samples have been deposited by using a magnetically enhanced dc triode sputtering system with a rotating substrate holder alternately passing over the targets. Typically, the deposition rates, controlled by quartz crystal monitors, were $\sim$5 Å/s on the CuMn and $\sim$12 Å/s on the Nb target. Two different Mn concentrations have been studied, 0.7 and 1.3 %. As was pointed out in Ref. 2, in order to make a careful study of the dependence of T_c versus the magnetic layer thickness, it is essential that the samples are deposited under identical conditions. With this purpose, we have developed a technique allowing the realization of a complete series of multilayers in only one deposition run. By using a suitable shutter on the CuMn target we let a 2 inch (100) Si substrate pass over the cathode in controlled steps to get different thicknesses of CuMn along a diameter of the wafer. After this phase, the substrate is brought to pass over the Nb target with a continuous velocity to obtain the next superconducting layer. At the end of the deposition process, the Si substrate is cut into strips about 3 mm wide and 1 cm long, perpendicularly to the obtained wedge. The designed Nb/CuMn multilayer series have constant Nb thickness d_{Nb}($\sim$250 Å) with CuMn thicknesses d_{CuMn} increasing arithmetically (d_{CuMn}, $2d_{\mathrm{CuMn}}$, $3d_{\mathrm{CuMn}}$,...). By changing the stopping times between successive steps on the CuMn target we could suitably adjust the initial value of d_{CuMn}. Both the Nb and the CuMn actual layer thicknesses were checked, after the deposition, by low angle x-ray measurements and electron dispersive spectroscopy (EDS) analyses. On certain samples Rutherford back scattering (RBS) analyses were also performed to measure both the thicknesses and the Mn concentrations. A zone ($\sim$5 mm wide) of the substrate in each deposition, was exposed only to the Nb target, in order to get information about the electrical and superconducting properties of the Nb layers present in the samples of the series. The thickness of the Nb film in this zone was about 2500 Å.

In this article we present data of two Nb/CuMn series, one (R22295) with 0.7% of Mn and d_{CuMn} thicknesses in the range 21–184 Å and the other (R16295) with 1.3% of Mn and d_{CuMn} values in the range 8–132 Å. A series of Nb/Cu multilayers with similar d_{Cu} values was also realized to compare the behavior between the magnetic and nonmagnetic case. In Fig. 1 are shown the superconducting transition

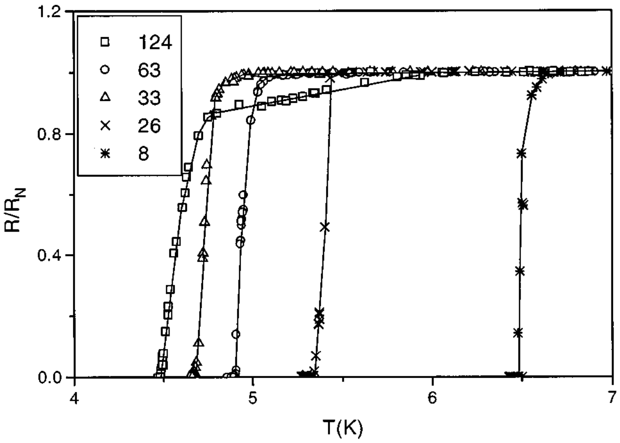

FIG. 1. Superconducting transition curves for some representative samples of the series R16295. The numbers in the legend are the CuMn layer thicknesses in Å. The curves are normalized to the normal state resistance.

curves for some of the samples of the series R16295, measured using a standard four probe technique. Some of the curves are broadened as a probable result of damage when the samples were cut.[2] Similar behaviors were observed in the transition curves of the Nb/Cu series. The ratios $\rho_N(300\ \mathrm{K})/\rho_N(10\ \mathrm{K})$, with ρ_N the normal state resistivity, were around 1.4 for the series R16295 and 1.6 for the series R22295.

In Fig. 2 we plot the values of the critical temperature T_{c0} versus the CuMn layering (T_{c0} is the temperature at which the electrical resistance R of the sample is less than $10^{-4}\ \Omega$) for the samples of the series R22295, Fig. 2(a), and of the series R16295, Fig. 2(b). The definition of T_{c0} and its use in Fig. 2 are related to the coincidence, observed in Ref. 2, between the T_c values determined by susceptibility measurements and the temperature where $R(T)=0.1R_N$. An oscillatory behavior of T_{c0} vs d_{CuMn} is present outside the experimental error. A similar nonmonotonic behavior was also present in the $T_{c,\mathrm{onset}}$ vs d_{CuMn} and in the $T_c(R/2)$ vs d_{CuMn} curves, where $T_{c,\mathrm{onset}}$ is defined as the temperature at which $R(T)=0.9R_N$ and $T_c(R/2)$ as the point at which $R(T)=0.5R_N$. On the other hand, a monotonic behavior of the T_{c0} versus d_{Cu} was observed in the Nb/Cu series, where the experimental data could be well fitted by the de Gennes-Werthamer theory.[11]

We have tried to explain the experimental data for the Nb/CuMn series extending the Radovic *et al.* theory to the case of superconducting–spin-glass multilayers. We point out that all our samples were not grown symmetrically, the first layer being always CuMn and the last Nb. Considering that all samples were made of 10 bilayers of Nb and CuMn, their asymmetry should not invalidate the applicability of the theory to our data. Moreover, we would like to address that previous measurements[12] on similar Nb/CuMn multilayers with higher Mn concentrations (7 and 14 %) have shown that we were in the presence of strongly coupled ($\xi_\perp > d_{\mathrm{CuMn}}$), where $\xi_\perp$ is the superconducting coherence length perpendicular to the Nb layers, bidimensional ($d_{\mathrm{Nb}} < \xi_{\mathrm{Nb,bulk}}$) superconducting systems. The reduction of the Mn concentration, increasing $\xi_\perp$, should enhance the coupling between Nb layers so that, in our case, the observed superconducting properties are related to the overall sample and not only to the topmost Nb layer. On the other hand, if only the top Nb layer were superconducting, the critical temperature of the samples in a series should be rather independent from the CuMn layer thicknesses.

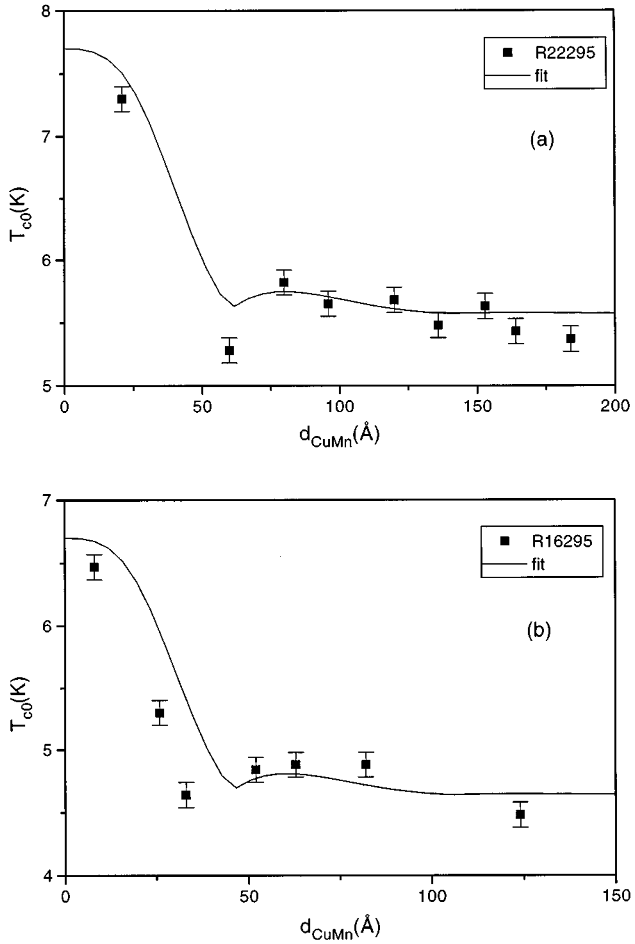

FIG. 2. T_{c0} vs d_{CuMn} curves for the series R22295 (a) and R16295 (b). The solid lines are the best fit curves obtained by using Eq. (3).

Following Ref. 13, we calculated T_c in the limit of small values of the parameter γ, given as

$$\gamma=\sigma_N\xi_S/\sigma_S\xi_N, \tag{1}$$

where $\sigma_{N,S}$ are the normal state conductivities of the magnetic, N, and superconducting, S, layers and $\xi_{N,S}$ are defined by

$$\xi_{N,S}=(D_{N,S}/2\pi T_c^*)^{1/2} \tag{2}$$

with $D_{N,S}$ the diffusion coefficient in the magnetic, N, and superconducting, S, layers and T_c^* the critical temperature of the bulk superconductor. As pointed out in Ref. 13, in the case of Nb/rare earth multilayers the values of γ are in the range 0.25–0.35 and the equations obtained in the limit $\gamma \ll 1$ well describe the experimental data. We have assumed this limit to be also valid for our samples. Assuming a parallel shunt resistor model,[14] we have generally measured, $\sigma_N/\sigma_S \sim 1$, so that we have $\xi_S < \xi_N$ if $\gamma < 1$. In the considered limit,[13] the critical temperature T_c is a function of T_c^*, ξ_S, ξ_N, γ and a, where a is a parameter inversely proportional to the exchange energy I. We obtained T_c^* directly by measuring the critical temperature of the single Nb film deposited in each series. These values were always in good agreement with the universal curve of the T_c vs ρ_N behavior.[15] The relation $\xi_S \sim \gamma \xi_N$ reduces the actual free fit parameters to a, γ, and ξ_N. The solid lines in Figs. 2(a) and 2(b) are the best fit curves obtained using the relation[13]

$$\frac{d_N}{2}\,\frac{d_S}{2}\,\frac{(T_c^* - T_c)}{\gamma \xi_N \xi_S T_c^*} = 3F(\phi, k_N d_N), \tag{3}$$

where $d_{N,S}$ are the magnetic, N, and superconducting, S, layer thicknesses, $k_N = (1+i)(I/D_N)^{1/2}$ and F is a function defined in Ref. 13 depending upon the phase ϕ of the superconducting order parameter. The values for T_c are obtained by minimizing F with respect to ϕ. For both series we have obtained $\gamma = 0.4$ and for the series R16295 we have obtained $a = 0.7$ and $\xi_N = 27$ Å ($\xi_S = 11$ Å), while for the series R22295 we have had $a = 0.85$ and $\xi_N = 30$ Å ($\xi_S = 12$ Å). The obtained ξ_S values are of the same order of the ξ_S values calculated from the relation

$$\xi_S = \sqrt{\xi_{\mathrm{BCS}} l / 3.4} \tag{4}$$

with $\xi_{\mathrm{BCS}} = 0.18 \hbar v_F / k_B T_c^*$. Our high resistivities values ($\rho \sim 10^{-7}$ Ω m) give rise to short mean free paths l and then to low ξ_S values ($\xi_S \sim 50$ Å). As shown in Fig. 2, the agreement with the experimental data is good both in the part with small d_{CuMn} values, where a fast decrease in T_{c0} is observed, and in the part with large d_{CuMn} values, where the T_{c0} oscillatory behavior starts to take place. In agreement with the Radovic *et al.* theory the first T_{c0} minimum shifts to lower d_{CuMn} values when increasing the Mn concentration (i.e., the magnetic influence). Moreover the T_{c0}/T_c^* minimum values are in the range 0.7–0.8, as it is expected in the case of large d_{Nb}/ξ_S values. From the a values, $a = (2\pi T_c^*/I)^{1/2}$, we obtain the exchange energy I for the two series. We have $I \simeq 5.8$ meV for the series with 0.7% of Mn and $I \simeq 7.4$ meV for the series with 1.3% of Mn. These values scale in the right way with the Mn concentration and are, as expected, about three orders of magnitude lower than the values measured in the case of superconducting–ferromagnetic multilayers. In fact, in Nb/Gd/Nb trilayers[16] a value of $I \simeq 0.17$ eV was found while $I \simeq 3.0$ eV was estimated in V/Fe multilayers.[17]

In conclusion, we have observed an oscillatory behavior of T_{c0} versus the CuMn layer thicknesses in superconducting (Nb)/ spin-glass (CuMn) multilayers. This effect is well explained in terms of the Radovic *et al.* theory, assuming the presence of the π phase in layered systems with weak magnetic coupling between the superconducting layers.

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[1] A. J. Millis and H. Monien, Phys. Rev. Lett. **70**, 2810 (1993); S. Katano, T. Matsumoto, A. Matsushita, T. Hatano, and S. Funahashi, Phys. Rev. B **41**, 2009 (1990).

[2] J. S. Jang, D. Davidovic, D. H. Reich, and C. L. Chien, Phys. Rev. Lett. **74**, 314 (1995).

[3] A. I. Buzdin and M. Y. Kupriyanov, JETP Lett. **52**, 487 (1990).

[4] Z. Radovic, M. Ledvij, L. Dobrosavljevic-Grujic, A. I. Buzdin, and J. R. Clem, Phys. Rev. B **44**, 759 (1991).

[5] W. Braunisch, N. Knauf, V. Kataev, S. Neuhausen, A. Grütz, A. Koch, B. Roden, D. Khomskii, and D. Wohlleben, Phys. Rev. Lett. **68**, 1908 (1992); C. Heinzel, T. Theilig, and P. Ziemann, Phys. Rev. B **48**, 3445 (1993); D. A. Wollman, D. J. Van Harlingen, W. C. Lee, D. M. Ginsberg, and A. J. Legget, Phys. Rev. Lett. **71**, 2134 (1993); P. Chaudari and S. Lin, *ibid.* **72**, 1084 (1994); A. G. Sun, D. A. Gajewski, M. B. Maple, and R. C. Dynes, *ibid.* **72**, 2267 (1994).

[6] M. Segrist and T. M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992).

[7] L. N. Bulaevskii, V. V. Kuzii, and A. A. Sobyanin, JETP Lett. **25**, 290 (1977).

[8] L. Hoines, R. Stubi, R. Loloee, J. A. Cowen, and J. Bass, Phys. Rev. Lett. **66**, 1224 (1991), and references therein.

[9] G. G. Kenning, D. Chu, B. Alavi, J. M. Hammann, and R. Orbach, J. Appl. Phys. **69**, 5240 (1991); C. Attanasio, L. Maritato, S. L. Prischepa, M. Salvato, B. N. Engel, and C. M. Falco, *ibid.* **77**, 2081 (1995).

[10] I. Banerjee, Q. S. Yang, C. M. Falco, and I. K. Schuller, Solid State Commun. **41**, 805 (1982); C. S. L. Chun, G. G. Zheng, J. L. Vicent, and I. K. Schuller, Phys. Rev. B **29**, 4915 (1984).

[11] P. G. deGennes and E. Guyon, Phys. Lett. **3**, 168 (1963); N. R. Werthamer, Phys. Rev. **132**, 2440 (1963).

[12] C. Attanasio, C. Coccorese, L. Maritato, S. L. Prischepa, M. Salvato, B. N. Engel, and C. M. Falco, Phys. Rev. B **53**, 1087 (1996).

[13] A. I. Buzdin, B. Bujicic, and M. Y. Kupriyanov, Sov. Phys. JETP **74**, 124 (1992).

[14] M. Gurvitch, Phys. Rev. B **34**, 540 (1986).

[15] C. Camerlingo, P. Scardi, C. Tosello, and R. Vaglio, Phys. Rev. B **31**, 3121 (1985).

[16] C. Strunk, C. Sürgers, U. Paschen, and H. v. Löhneysen, Phys. Rev. B **49**, 4053 (1994).

[17] P. Koorevaar, Y. Suzuki, R. Coehoorn, and J. Aarts, Phys. Rev. B **49**, 441 (1994).