

## Angular dependence of the upper critical field in Nb/CuMn multilayers

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We present data related to the temperature ( $T$ ) and angular ( $\Theta$ ) dependencies of the upper critical field ( $H_{c2}$ ) of Nb(superconductor)-CuMn(spin glass) multilayers as a function of CuMn layer thickness and Mn concentration. We observe two-dimensional (2D) behavior for large CuMn thickness. As the CuMn layer thickness is decreased, the  $H_{c2}(T)$  curves correspond to three-dimensional (3D) behavior, while the  $H_{c2}(\Theta)$  dependencies measured in the range  $[-1.5^\circ, +1.5^\circ]$  are more sensitive to the dimensionality of the system. In particular, the experimental data reveal a 3D $\rightarrow$ 2D crossover at very low angle when the Mn percentage is increased at fixed CuMn thickness. This behavior can be related to vortex dimensionality change in anisotropic superconductors. [S0163-1829(98)00905-9]

The coexistence of superconductivity and magnetism seems to play an important role in high-temperature superconductors (HTSC).<sup>1,2</sup> To better investigate the mutual influence of these two phenomena it is possible to realize artificial multilayered structures consisting of superconducting and magnetic layers ( $S$ - $M$ - $S$  structures). In such multilayers one can easily control and change the anisotropy, the dimensionality, and the nature of the coupling between the superconducting layers. Spin polarization in magnetic layers gives rise to interesting effects, such as the anomalous temperature behavior of the anisotropy<sup>3,4</sup> and the critical temperature oscillations versus the magnetic layer thickness.<sup>5-8</sup> At the same time, the  $S$ - $M$ - $S$  structures present a special interest from the viewpoint of model systems for a better understanding of vortex mechanisms in HTSC.<sup>9</sup> In fact, the superconducting order parameter goes very fast to zero in  $M$  layers,<sup>10</sup> and this gives the possibility to use  $M$ -layer thicknesses of a few angstroms, practically of the same order of magnitude of the lattice constants in HTSC.

It is well known that in artificial multilayers the dimensionality can be changed with the temperature due to the temperature dependence of the perpendicular coherence length  $\xi_\perp(T)$ . In fact, when the coherence length  $\xi_\perp(T)$  is larger than the nonsuperconducting layer thickness, the multilayer behaves like a three-dimensional (3D) system and the temperature dependence of the parallel critical magnetic field, assuming the well-known Ginzburg-Landau equation,<sup>11</sup> is linear,  $H_{c2\parallel} = \phi_0/2\pi\xi_\perp\xi_\parallel \propto (T_c - T)$ , while when  $\xi_\perp(T)$  is smaller than the nonsuperconducting layer thickness  $d_n$ , the system is bidimensional (2D) and  $H_{c2\parallel} = \phi_0/2\pi d_n\xi_\parallel \propto (T_c - T)^{1/2}$ . The existence of dimensional crossover was proved also by measurements of  $H_{c2}$  angular dependence for different nonsuperconducting layers thicknesses.<sup>12</sup> As it is known,

for 3D Josephson coupled superconductors the angular dependence  $H_{c2}(\Theta)$  obeys the Lawrence-Doniach (LD) equation:<sup>13</sup>

$$\left[ \frac{H_{c2}(\Theta)\sin\Theta}{H_{c2\perp}} \right]^2 + \left[ \frac{H_{c2}(\Theta)\cos\Theta}{H_{c2\parallel}} \right]^2 = 1, \quad (1)$$

where  $\Theta$  is the angle between the film surface and the magnetic-field direction and  $H_{c2\perp}$  is the perpendicular critical magnetic field. For a 2D-thin-film Tinkham has obtained the following expression:<sup>14</sup>

$$\left| \frac{H_{c2}(\Theta)\sin\Theta}{H_{c2\perp}} \right| + \left[ \frac{H_{c2}(\Theta)\cos\Theta}{H_{c2\parallel}} \right]^2 = 1. \quad (2)$$

The general feature related to Eq. (1) is that at  $\Theta=0$  (i.e., at parallel magnetic field) the first derivative is zero,  $dH_{c2}/d\Theta=0$ , and the curve is smooth and “bell” shaped. Vice versa, from Eq. (2) follows that  $H_{c2}(\Theta)$  has a cusp at  $\Theta=0$ .

The dimensional crossover has been observed also in HTSC, confirming the layered structure of these materials. Measurements of  $H_{c2}(\Theta)$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  films show a dimensional crossover from the isotropic 3D behavior at temperature close to  $T_c$  to the 2D thin-film behavior while lowering the temperature.<sup>15</sup>

Recently, angular-dependent dimensional crossover in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  thin films has been observed at fixed temperature.<sup>16</sup> In this case, the  $H_{c2}(\Theta)$  curve follows Eq. (1), but, at very low angle values, it presents a pronounced rise with a cusp at  $\Theta=0$ , indicating a 3D $\rightarrow$ 2D crossover induced by the field orientation relative to the film surface. This behavior has been explained in terms of a model for

TABLE I. Characteristics of six Nb/CuMn multilayer samples.  $d_{\text{Nb}}$  and  $d_{\text{CuMn}}$  are, respectively, the Nb and CuMn thicknesses, %Mn is the Mn concentration in the nonsuperconducting layers,  $\beta_{10}$  is the ratio between the 300 K,  $\rho_{300\text{ K}}$ , and 10 K,  $\rho_{10\text{ K}}$ , resistivities,  $M/m$  is the anisotropic Ginzburg-Landau (GL) mass ratio at  $T=4.2\text{ K}$ , and  $\xi_{\perp}$  is the perpendicular coherence length at  $T=4.2\text{ K}$ .

Sample	$d_{\text{Nb}}$ (Å)	$d_{\text{CuMn}}$ (Å)	%Mn	$T_c$ (K)	$\beta_{10}=\rho_{300\text{ K}}/\rho_{10\text{ K}}$	$\rho_{300\text{ K}}$ ( $\mu\Omega\text{ cm}$ )	$M/m$	$\xi_{\perp}$ (Å)
NC16	230	16	0	8.05	2.52	25	4.3	92
NCM4	230	5	6.6	7.61	2.40	25	2.7	90
NCM16	230	19	6.6	5.92	2.45	27	23.0	49
NCM4A	230	4	14.4	7.08	2.24	28	2.6	91
NCM16A	230	16	14.4	4.57	2.25	30	96.0	34
NCM50A	230	50	14.4	4.79	2.15	28	55.0	54

layered superconductors,<sup>17</sup> which describes the coupling between the layers using an electron interlayer interaction parameter  $g_3$ .

In this paper, we present the experimental data related to the temperature and angular dependencies of  $H_{c2}$  for Nb(superconductors)-CuMn(spin glass) multilayers. In these systems we have observed changes in the dimensional behavior not only due to the perpendicular coherence length temperature dependence or to the different nonsuperconducting layer thickness, but also due to the different Mn content in the CuMn alloy, without changing the temperature and the CuMn thickness. An angular-dependent 3D $\rightarrow$ 2D crossover in the  $H_{c2}(\Theta)$  curve has been measured. We discuss this behavior in terms of the  $g_3$  model, which seems particularly suitable for superconducting/spin-glass multilayers.

The samples with a constant Nb thickness (230 Å) and fixed number of bilayers (10) were grown on sapphire (100) substrates using magnetically enhanced dc triode sputtering with a rotating substrate holder alternately passing over the targets.<sup>18</sup> The bottom layer was always CuMn and the top layer was Nb. The magnetic phase composition was determined by Rutherford backscattering spectroscopy analysis. Samples were characterized by four-point dc resistive transition measurements  $R(T, H)$  with an external magnetic field (0–5 T) applied both perpendicular and parallel to the substrate surface. High-resolution  $H_{c2}$  angular measurements were performed at  $T=4.2\text{ K}$ . The angle  $\Theta$  between the film surface and the external magnetic-field orientation was changed using a stainless-steel worm-gear rotation mechanism placed in a liquid-helium bath. The angular resolution of the mechanism is equal to  $0.02^\circ$ . The  $H_{c2}$  value was obtained at half of the resistive transition  $R(T, H)$ . The bias current density was about  $1\text{ A/cm}^2$ . A superconducting solenoid with high uniformity of the field in the zone where the samples were situated was used to produce magnetic field up to 5 T. The characteristic features of the investigated samples are summarized in Table I. In the case of CuMn thickness  $d_{\text{CuMn}} \approx 16\text{ Å}$  a Nb/Cu multilayer with similar  $d_{\text{Cu}}$  value was also realized to compare the behavior between magnetic and nonmagnetic cases. In the inset of Fig. 1 are shown typical transition curves of a Nb/CuMn multilayer with  $d_{\text{CuMn}} = 6\text{ Å}$  and Mn concentration of 6.6%, taken in the presence of external magnetic-field applied parallel to the plane of the film. The curves are only shifted to lower temperatures by the application of increasing magnetic fields, without any change in the width of the resistive transitions, which are sharper than 0.1 K. Similar behavior is shown by the transi-

tion curves when the temperature is fixed and the external magnetic-field amplitude is varied. Our experimental errors are therefore comparable or lower than the dimensions of the data points in the figure.

For the samples with small values of the nonsuperconducting layer thickness  $d_n$  ( $d_n < 10\text{ Å}$ ) the temperature and the angular  $H_{c2}$  behaviors were related to the presence of surface superconductivity, independently from the Mn concentration values. As an example, in Fig. 1(a) are shown the temperature dependencies of the parallel and perpendicular critical magnetic fields for sample NCM4A. The solid lines represent the linear best fits. The  $H_{c2\parallel}(T)$  linear behavior

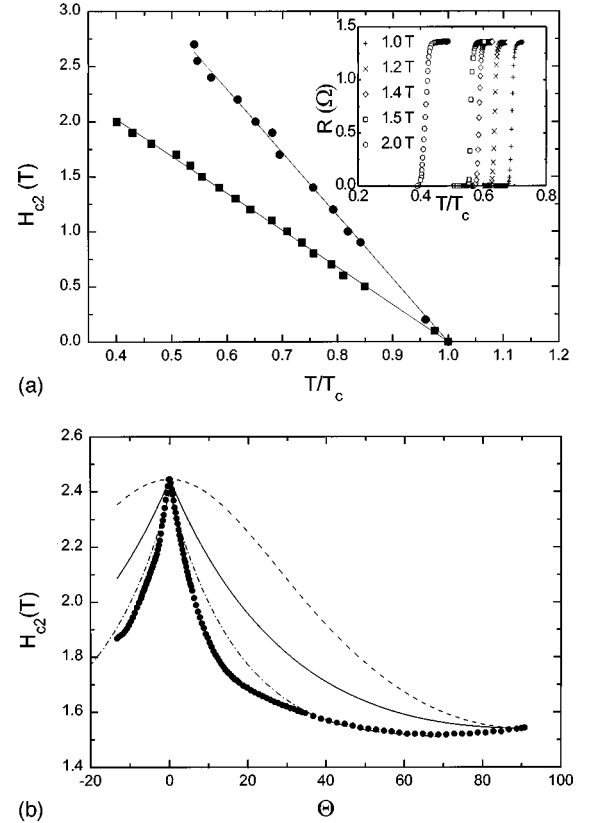


FIG. 1. (a) Perpendicular (squares) and parallel (circles) critical magnetic fields vs temperature for sample NCM4A. The solid lines represent the linear best fits. The inset shows typical resistive transitions for our samples in parallel magnetic field. (b) Angular dependence  $H_{c2}(\Theta)$  for sample NCM4A. The solid line is calculated from Eq. (2) (2D case). The dashed line is calculated from Eq. (1) (3D case). The dash-dotted line is calculated from Eq. (3).

reveals the 3D nature of the system and the ratio  $H_{c2\parallel}/H_{c2\perp} \approx 1.62$  strongly indicates the presence of surface superconductivity.

This suggestion is confirmed by the angular  $H_{c2}(\Theta)$  dependence for this sample, shown in Fig. 1(b). A small  $H_{c2}$  rise around  $\Theta = 90^\circ$ , also observed for Nb/Cu,<sup>19</sup> Nb/Nb<sub>x</sub>O<sub>y</sub>,<sup>20</sup> and Nb/NbZr (Ref. 21) multilayers, is present and can be described as an influence of the pinning centers perpendicular to the film surface (for example, grain boundaries<sup>20</sup>). The solid line in Fig. 1(b) corresponds to the 2D case [Eq. (2)] and the dashed line to the 3D case [Eq. (1)]. As was pointed out by Banerjee and Schuller,<sup>19</sup> when there is a surface superconductivity contribution, the experimental  $H_{c2}(\Theta)$  dependence is very similar to that expected from Eq. (2). In our case, for the NCM4A sample, the curve is qualitatively similar to that obtained from Eq. (2), but all the points fall below the theoretical curves. As was shown in Ref. 22, the angular dependence of the surface superconducting critical field  $H_{c3}$  is given by

$$\left[ \frac{H_{c3}(\Theta)}{H_{c2\parallel}} \cos \Theta \right]^2 [1 + \tan \Theta \times (1 - \sin \Theta)] + \left[ \frac{H_{c3}(\Theta)}{H_{c2\perp}} \sin \Theta \right] = 1. \quad (3)$$

The dashed-dotted line in Fig. 1(b), calculated from Eq. (3), gives a better description of the experimental points.

A similar behavior has been observed in the  $H_{c2}(\Theta)$  dependence of sample NC16, which has  $d_n = 16$  Å. Due to the Mn absence in the nonsuperconducting layers, its anisotropic Ginzburg-Landau (GL) mass ratio, defined in the usual way as the ratio  $M/m = (H_{c2\parallel}/H_{c2\perp})^2$ , is equal to 4.3, much smaller than the mass ratio observed, as an example, in sample NCM16A, which has a very similar  $d_n$  value, but a Mn percentage of 14.4% in the nonsuperconducting layers (see Table I).

In Fig. 2(a) we show the  $H_{c2}(T)$  dependencies for sample NCM50A, which has  $d_n = 50$  Å with a Mn percentage of 14.4%.<sup>23</sup> Close to  $T_c$  the  $H_{c2\parallel}(T)$  curve is linear while at lower temperatures ( $T < 4.59$  K) the behavior is no longer linear, starting to be square-root-like. As we pointed out before, this effect was observed in many superconducting multilayers and is related to a 3D→2D crossover.<sup>24</sup> We want to stress that the important parameter in order to observe the  $H_{c2\parallel}(T)$  dimensional crossover is the ratio  $\xi_{\perp}(T)/d_n$  and not the anisotropic GL mass ratio. For a Josephson coupled multilayer the 3D→2D crossover in  $H_{c2\parallel}$  should happen when  $\xi_{\perp}(T)/(d_n + d_s) \approx 0.7$ ,<sup>25</sup> where  $d_s$  is the superconducting layer thickness. In our samples this ratio, with the values of  $\xi_{\perp}$  obtained from the Ginzburg-Landau dependence of the critical fields,<sup>11</sup> was always lower than 0.4 (0.3 for NCM50A). At the same time for the crossover point we get  $\xi_{\perp}(T)/d_n > 1$ . As an example, for sample NCM50A we have  $\xi_{\perp}(T)/d_n \approx 1.5$ . We relate such a discrepancy with the case of Josephson coupled superconductors to the different coupling mechanism between layers. The  $H_{c2}(\Theta)$  measurements, Fig. 2(b), performed at  $T = 4.2$  K confirmed the 2D nature of the sample. The solid line in Fig. 2(b) corresponds

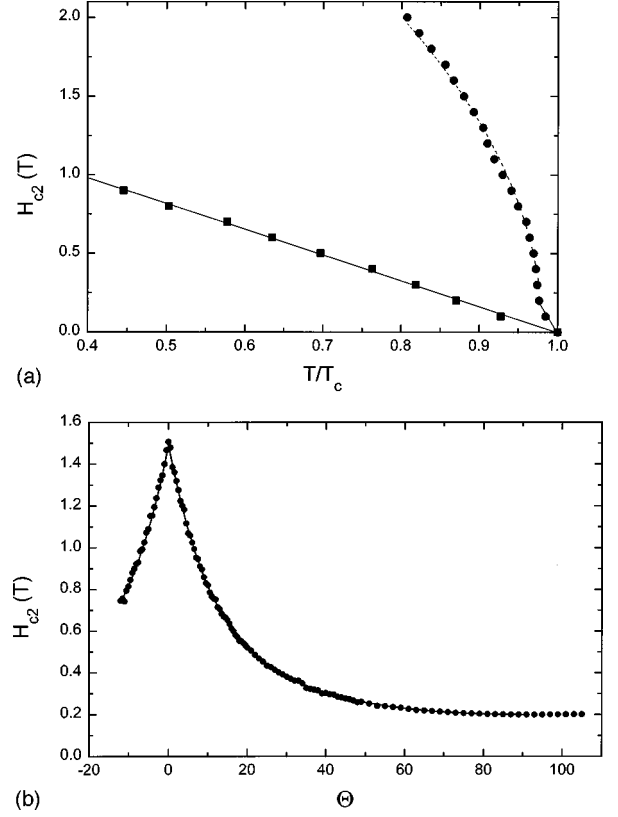


FIG. 2. (a) Perpendicular (squares) and parallel (circles) critical magnetic fields vs temperature for sample NCM50A. The solid lines correspond to the linear best fits. The dashed lines correspond to  $H_{c2}(T) \propto (1 - T/T_c)^{1/2}$ . (b) Angular dependence  $H_{c2}(\Theta)$  for sample NCM50A. The solid line is calculated from Eq. (2).

to Tinkham's formula without using any free fitting parameter. The very good agreement between theory and experiment is evident.

In the intermediate range of  $d_n$  values,  $10 \text{ Å} < d_n < 30 \text{ Å}$ , we have observed an angular-dependent dimensional crossover by changing the Mn percentage in the nonsuperconducting layer. In Fig. 3(a) the  $H_{c2}(T)$  dependencies are shown for sample NCM16 with 6.6% of Mn. Solid lines represent the linear best fits and the agreement with the data indicates the 3D behavior of the sample in the whole observed temperature range. For this sample we have performed high-resolution  $H_{c2}(\Theta)$  measurements in the range  $[-1.5^\circ, 1.5^\circ]$ . The result is drawn in Fig. 3(b). As it is seen, close to  $\Theta = 0$  the  $H_{c2}(\Theta)$  curve is smooth and bell shaped, again indicating a 3D behavior at 4.2 K. The LD fit, solid line in Fig. 3(b), obtained without free fitting parameters, well describes the data in the region close to  $\Theta = 0$ . At higher angles,  $|\Theta| > 1^\circ$ , the agreement between the LD fit and the experimental data is not very good. The anisotropic GL mass ratio of this sample is 23.0, much higher than the value 2.8 expected in the case of predominant surface superconductivity effects. The disagreement between the LD theory and the experimental data at  $|\Theta| > 1^\circ$  could probably be related to the different coupling mechanism in Nb/CuMn multilayers with respect to  $S$ - $I$ - $S$  or  $S$ - $N$ - $S$  systems (with  $I$  and  $N$ , respectively, denoting insulators and normal metals).

Models other than LD can be used to describe the interlayer interaction in  $S$ - $M$ - $S$  multilayers, which are not limited

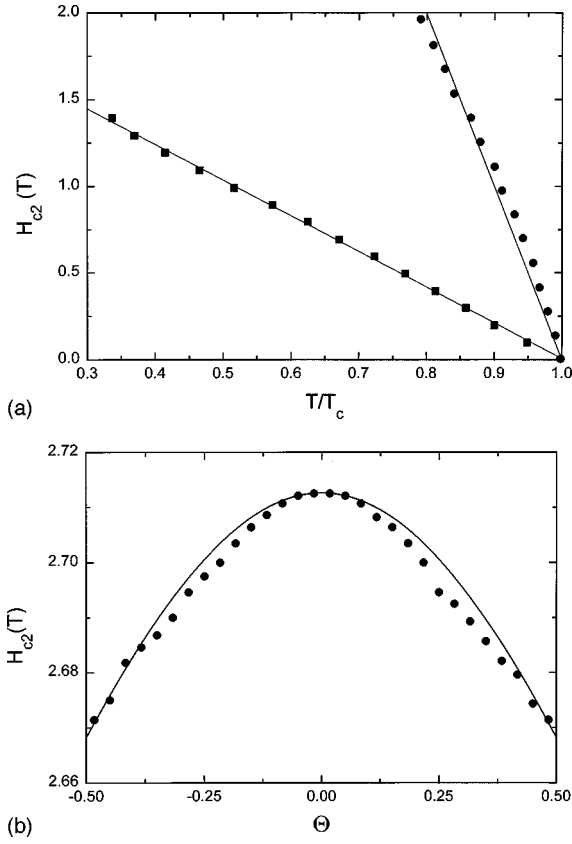


FIG. 3. (a) Perpendicular (squares) and parallel (circles) critical magnetic fields vs temperature for sample NCM16. Solid lines represent the linear best fits. (b) Angular measurements,  $H_{c2}(\Theta)$ , for sample NCM16. The solid line is calculated from Eq. (1).

to the case of the Josephson coupling.<sup>6,17,26</sup> In particular, Schneider and Schmidt<sup>17</sup> (SS) proposed a model that takes into account the coupling between the layers in terms of an interlayer interaction electron parameter  $g_3$ . This model is not limited to the weak-coupling case and moreover, depending upon the sign of  $g_3$ , it can also describe the case when the order parameter between adjacent layers change from positive to negative, the so-called  $\pi$  phase. The presence of such a  $\pi$  phase in  $S$ - $M$ - $S$  multilayers has recently been suggested to explain the critical temperature oscillations versus the magnetic layer thickness.<sup>5–8</sup>

By increasing the Mn content in CuMn layers, we can increase the anisotropy of the system, reaching the situation where at the same values of Nb and CuMn layer thicknesses the multilayer dimensionality might be changed (see Table I). In Fig. 4(a) we show the  $H_{c2}(T)$  curves for sample NCM16A, which has layer thicknesses very similar to those of sample NCM16, but a higher Mn concentration. The solid lines are the linear best fits. The 3D behavior of the sample in the measured temperature and magnetic-field range is evident. The anisotropic mass ratio of sample NCM16A is higher, 96.0, than that measured in sample NCM16, 23.0, confirming that we can change the anisotropy of this system by only changing the Mn percentage in the nonsuperconducting layers. For sample NCM16A the  $H_{c2}(\Theta)$  curve is bell shaped at  $|\Theta| > 0.2^\circ$ , but rises sharply at  $\Theta = 0$ , resulting in a cusp, Fig. 4(b). The rise in  $H_{c2}(\Theta)$  is well above the experimental error, which is of the order of the dimensions of the

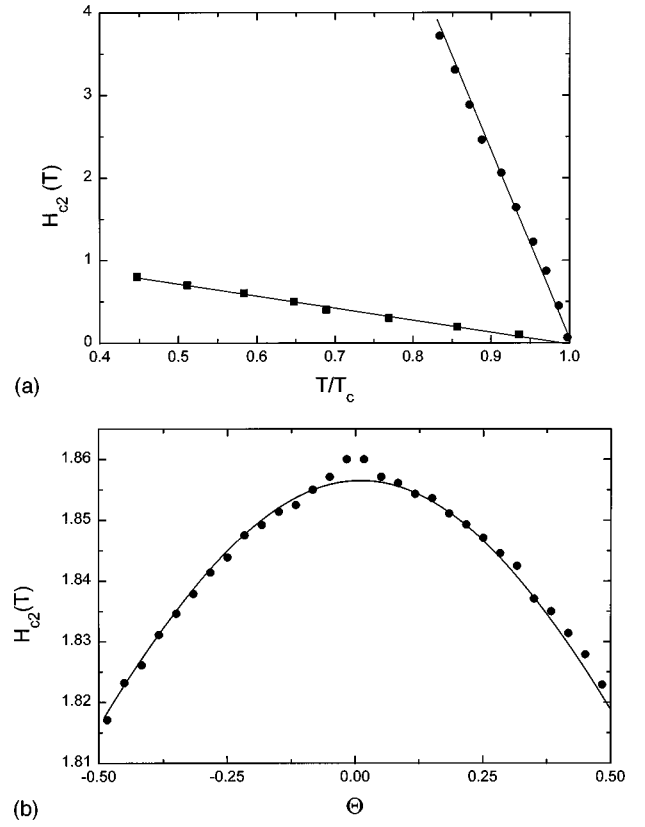


FIG. 4. (a) Perpendicular (squares) and parallel (circles) critical magnetic fields vs temperature for sample NCM16A. Solid lines represent the linear best fits. (b) Angular measurements,  $H_{c2}(\Theta)$ , for sample NCM16A. The solid line is calculated from Eq. (1).

data points in figure, and does not follow the 3D curve obtained by Eq. (1) without free fitting parameters. Therefore, the sample can be described as a 3D system at high angles, while at angles very close to zero it has a 2D behavior. This angular-dependent crossover is similar to that observed on  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  thin films<sup>16</sup> and can be interpreted in terms of the  $g_3$  model. According to Schneider and Schmidt,<sup>17</sup> when the system is in a temperature range close to the point where the  $H_{c2\parallel}(T)$  dependence changes from linear to square root, the  $H_{c2}(\Theta)$  dependence presents a cusplike behavior at  $\Theta = 0$  on the top of a bell-shaped curve (Fig. 4 in Ref. 17). Therefore, one can observe this angular-dependent crossover for temperatures at which the  $H_{c2\parallel}(T)$  curve still behaves linearly. In the case of our  $H_{c2}(\Theta)$  measurements in Fig. 4(b) the angular range, where the 2D behavior is observed, is very small  $[-0.2^\circ, 0.2^\circ]$ . The angular experimental error in the  $H_{c2\parallel}(T)$  measurements can be higher than  $\pm 0.2^\circ$ , and therefore one can argue that this could be the reason for the observed linear behavior. It is then interesting to point out that all our  $H_{c2}(\Theta)$  measurements have been performed at 4.2 K. At this temperature the ratio  $\xi_{\perp}/d_n$  is 2.8 for sample NCM16, while  $\xi_{\perp}/d_n = 2.1$  for sample NCM16A. As already observed, for sample NCM50A, which has the same Mn concentration of sample NCM16A, the 3D  $\rightarrow$  2D crossover in  $H_{c2\parallel}(T)$  takes place when  $\xi_{\perp}/d_n = 1.5$ . Therefore, sample NCM16A should behave linearly in  $H_{c2\parallel}(T)$  at 4.2 K, independently from the angular experimental error. Sample NCM16A, with a higher Mn concentration, has a smaller  $\xi_{\perp}/d_n$  ratio at 4.2 K than sample NCM16, being closer to the

point where the  $H_{c2\parallel}(T)$  curve changes behavior. According to the SS model, this is the reason why the angular dimensional crossover is observed in sample NCM16A and not in sample NCM16.

The physical nature of such an unusual  $H_{c2\parallel}(\Theta)$  dependence could be traced back to vortex dimensionality changes in anisotropic superconductors.<sup>21,27</sup> In fact, close to the 3D→2D crossover temperature for the  $H_{c2\parallel}(T)$  curve, the adjacent superconducting layers are only weakly interacting. When  $\Theta \gg |0.2^\circ|$  the vortex lines in the multilayers have probably a “staircase” form,<sup>28</sup> where 2D pancake Abrikosov vortices are connected through Josephson-like vortices.

Changing the orientation of the external magnetic-field relative to the layers, one can decouple them in the limit of perfect alignment ( $\Theta=0$ ) reaching the situation where the vortices with Josephson-like cores in the zones between adjacent superconducting layers<sup>29</sup> are no longer connected through 2D pancake vortices.

In conclusion, we have examined the dimensionality of the Nb/CuMn multilayers by means of temperature and high-

resolution angular measurements of the upper critical magnetic field. The dimensionality of our samples was varied by changing both the CuMn layer thickness and the Mn percentage. The 2D behavior for sample NCM50A was unambiguously confirmed both by temperature and angular measurements, while for smaller  $d_n$  values ( $d_n < 10 \text{ \AA}$ ), the effects of surface superconductivity were predominant. At intermediate  $d_n$  values,  $10 < d_n < 30 \text{ \AA}$ , the  $H_{c2\parallel}$  dependencies are linear, indicating 3D behavior, while the angular measurements performed with high-resolution (down to  $0.02^\circ$ ) revealed an angular 3D→2D crossover similar to that observed in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  films<sup>16</sup> as the Mn concentration is increased. This result confirms the special interest in studying *S-M-S* artificial multilayers as model systems to better understand many properties of the HTSC compounds.

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