

**Disorder-induced critical phenomena in magnetically glassy Cu-Al-Mn alloys**Jordi Marcos,<sup>1</sup> Eduard Vives,<sup>1,\*</sup> Lluís Mañosa,<sup>1</sup> Mehmet Acet,<sup>2</sup> Eyüp Duman,<sup>2</sup> Michel Morin,<sup>3</sup>  
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Measurements of magnetic hysteresis loops in Cu-Al-Mn alloys of different Mn content at low temperatures are presented. The loops are smooth and continuous above a certain temperature, but exhibit a magnetization discontinuity below that temperature. Scaling analysis suggest that this system displays a disorder-induced phase transition line. Measurements allow one to determine the critical exponents  $\beta=0.03\pm 0.01$  and  $\beta\delta=0.4\pm 0.1$ , which coincide with those reported recently in a different system, thus supporting the existence of universality for disorder-induced critical points.

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**I. INTRODUCTION**

Beyond equilibrium, the response of a physical system to an external applied force shows hysteresis. Close to the rate independent limit, when thermal fluctuations do not control the kinetics of the system, the hysteresis path is reproducible from cycle to cycle. This athermal behavior is observed, for instance, in the magnetization reversal through a first-order transition in many magnetic systems at low enough temperatures. Modeling the shape of the hysteresis loops is a difficult problem that has attracted a great deal of interest for many years. Theoretical studies of the zero-temperature (absence of thermal fluctuations) hysteresis in a number of lattice models with different kinds of quenched disorder (random fields,<sup>1</sup> random bonds,<sup>2</sup> random anisotropies,<sup>3</sup> etc.) have shown that by increasing the strength of disorder, the hysteresis loops change from displaying a discontinuous jump in the magnetization reversal (ferromagnetic character) to behaving smoothly with the applied field. This change has been interpreted as a disorder-induced nonequilibrium phase transition. For this phase transition, critical exponents corresponding to different models have been computed from numerical simulations and renormalization-group methods. Moreover, universal scaling has been predicted. For instance, for short-range 3D models<sup>4</sup>  $\beta=0.035\pm 0.028$  and  $\beta\delta=1.81\pm 0.32$ , whereas for mean-field models<sup>5</sup>  $\beta=1/2$  and  $\beta\delta=3/2$ .

In contrast to the considerable amount of theoretical research, only very few experimental works have been devoted to corroborate the actual existence of disorder-induced phase transitions in the hysteresis loop. Obradó *et al.*<sup>6</sup> showed that the hysteresis loops in the magnetically glassy phase of Cu-Al-Mn change with increasing disorder (related to the alloy composition) as predicted by the models. However, no critical exponents were estimated in that work. Later, Berger *et al.*<sup>7</sup> studied the effect of magnetic disorder on the reversal process in Co/CoO bilayers. In this case, the antiferromagnetic CoO layer allows a reversible tuning of disorder by temperature variation. They prove the existence of hysteresis-loop criticality and determine critical exponents.

Nevertheless, the obtained exponents ( $\beta=0.022\pm 0.006$  and  $\beta\delta=0.30\pm 0.03$ ) show important discrepancies with the theoretical predictions. Therefore more effort is needed in the experimental analysis of other candidates in order to confirm the existence of disorder induced phase transitions as predicted by theory.

In this paper, we present magnetic hysteresis loops for different compositions and temperatures below the freezing temperature in a Cu-Al-Mn alloy. A critical transition line associated with the onset of a discontinuity in the hysteresis loop in the temperature-field-composition space has been determined, and the critical exponents have been obtained from the scaling of the magnetization curves. Our exponents show a remarkable agreement with the previous experimental estimates,<sup>7</sup> thus supporting the idea of universality in such disorder induced transitions. Results also indicate that  $T=0$  theoretical models may lack an essential physical ingredient in order to account for finite temperature measurements.

**II. EXPERIMENTAL RESULTS**

A Cu<sub>66.6</sub>Al<sub>20.4</sub>Mn<sub>13.0</sub> single crystal has been grown by the Bridgman method. The crystallographic structure is  $L2_1$  ( $Fm\bar{3}m$ ) which can be viewed as four interpenetrated fcc sublattices.<sup>6</sup> From the ingot, a parallelepiped specimen ( $a\approx 3.4$  mm,  $b\approx 2.4$  mm, and  $c\approx 2.2$  mm) has been cut using a low-speed diamond saw, with  $a$  and  $b$  oriented along the [001] and [110] directions, respectively. The sample was annealed for 10 min at 1080 K, quenched in a mixture of ice and water, and aged at room temperature for several days.

The magnetic properties of Cu-Al-Mn arise from localized magnetic moments at Mn-atoms. These moments are coupled through an oscillating effective Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction.<sup>8</sup> Due to nonstoichiometry, Mn atoms are not uniformly distributed on one of the  $L2_1$  sublattice sites.<sup>9</sup> This atomic disorder leads to the coexistence of ferromagnetic and antiferromagnetic interactions, which is at the origin of the glassy behavior observed at low temperatures.<sup>10</sup>

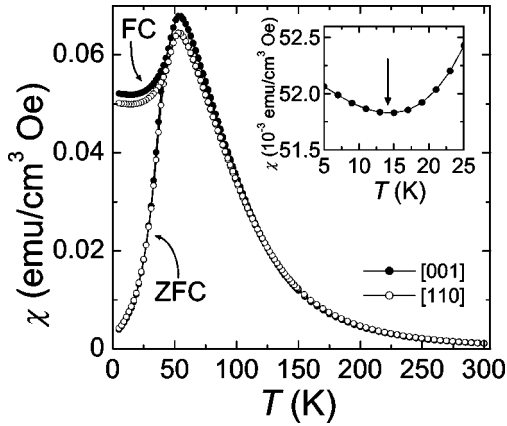


FIG. 1. ZFC and FC low-field susceptibility of a  $\text{Cu}_{66.6}\text{Al}_{20.4}\text{Mn}_{13.0}$  single crystal with  $H=100$  Oe along the [001] (●) and [110] (○) crystallographic directions. The inset reveals the existence of a minimum at  $T_c \approx 15$  K in the FC curve.

The magnetization as a function of temperature and magnetic field has been measured with a superconducting quantum interference device magnetometer. Results presented here have been corrected for demagnetizing field effects. Figure 1 shows the behavior of the zero-field-cooled (ZFC) and field cooled (FC) susceptibilities as a function of temperature. Measurements have been done by applying a field of 100 Oe. The different symbols correspond to the field applied in the [001] and [110] crystallographic directions. No significant anisotropy is observed. The ZFC and FC curves split at a temperature of 43 K below the position of the  $\chi$  peak at  $\sim 55$  K. The splitting confirms the onset of irreversibility and the glassy behavior at low temperatures. Moreover, the inset shows an enlarged view of the FC susceptibility in this glassy region. The existence of a minimum in this curve indicates that, along this metastable path, there is a tendency for the system to develop a field induced ferromagnetic component at low temperatures. No indication of such a tendency can be observed in the ZFC curve.

Figure 2 shows a series of examples of the central part of hysteresis cycles at selected temperatures in the range from 5 K up to 110 K. Each loop has been obtained by cooling from 150 K in the absence of an external field down to the selected temperature and slowly varying the field from  $0 \rightarrow 50$  kOe  $\rightarrow -50$  kOe  $\rightarrow 50$  kOe. This allows one to obtain the virgin magnetization curve and the full hysteresis loop.

Above 40 kOe (and below  $-40$  kOe) the loops show already a reversible behavior. Nevertheless, saturation of the magnetization is still not reached at 50 kOe, as can be seen in Fig. 3. Such a reversible behavior is a signature of the existence of antiferromagnetic interactions. To a high degree of accuracy, the loops are symmetric under inversion of the magnetization and field axis. Moreover, we have checked that loops are reproducible and that no significant relaxation effects occur for field sweeping rates in the range 0.25–30 Oe/s. Note first that above  $T \approx 43$  K no hysteresis effects are observed, consistently with the behavior of  $\chi(T)$  in Fig. 1. Second, the loops in the range from 15 to 45 K are typical of a glassy system while at  $T < 15$  K the loops display a jump associated with the field induced ferromagnetic component.

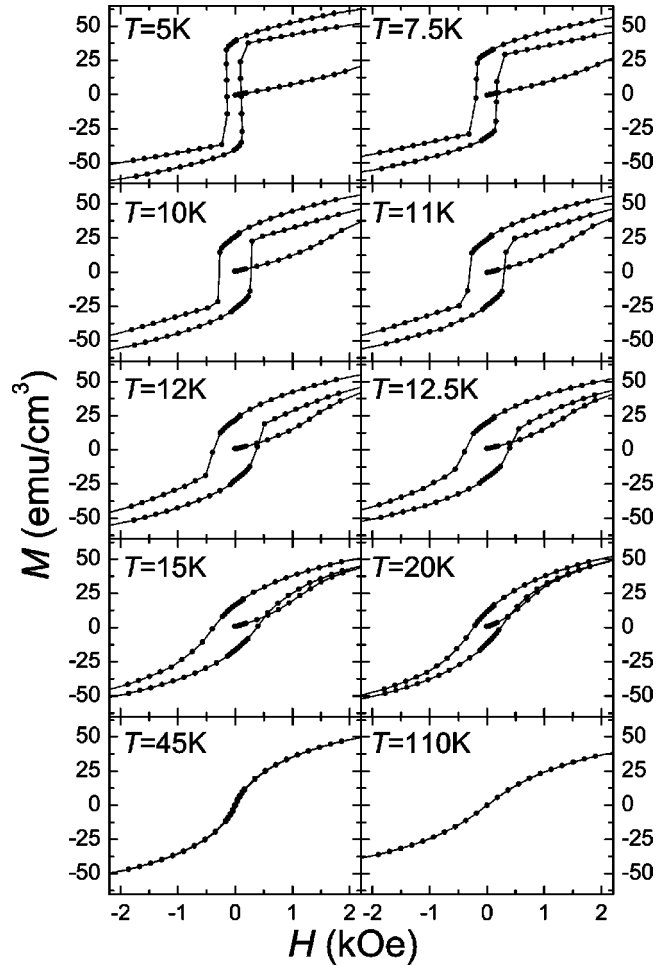


FIG. 2. Central part of the hysteresis cycles at different temperatures of a  $\text{Cu}_{66.6}\text{Al}_{20.4}\text{Mn}_{13.0}$  single crystal, obtained after cooling in the absence of an external field and slowly varying the field from  $0 \rightarrow 50$  kOe  $\rightarrow -50$  kOe  $\rightarrow 50$  kOe.

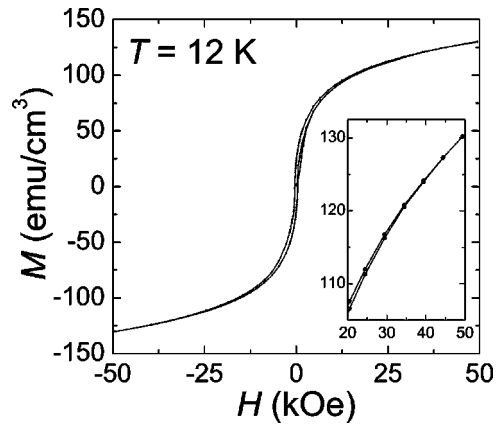


FIG. 3. Hysteresis cycle of a  $\text{Cu}_{66.6}\text{Al}_{20.4}\text{Mn}_{13.0}$  single crystal obtained at  $T=12$  K after a zero-field cooling, illustrating that magnetization does not saturate for fields up to 50 kOe but shows a reversible behavior for  $|H| \geq 40$  kOe. The inset shows a magnified view of the region of high applied fields ( $> 20$  kOe) with the same units.

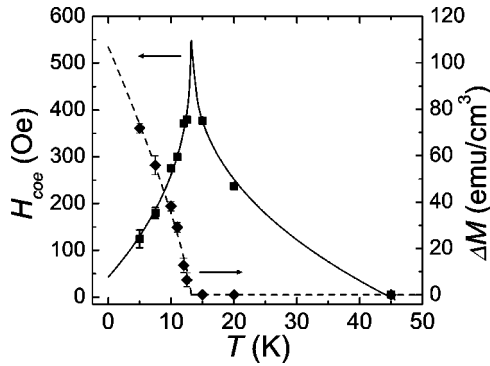


FIG. 4. Behavior of the coercive field  $H_{coe}$  and the magnetization jump  $\Delta M$  as a function of temperature of a  $\text{Cu}_{66.6}\text{Al}_{20.4}\text{Mn}_{13.0}$  single crystal. The continuous line is a fit of the integral of Eq. (3), and the discontinuous line is a guide to the eyes.

This becomes more pronounced as the temperature is lowered. On the other hand, the virgin curves do not show any signature of a ferromagnetic component, in agreement with the behavior of the ZFC susceptibility in Fig. 1.

The characterization of the onset of the ferromagnetic component is presented in Fig. 4, where we show the behavior of the estimated coercive field  $H_{coe}(T)$  and the magnetization jump  $\Delta M(T)$  as a function of temperature.<sup>11</sup> The coercive field exhibits a peak at  $T_c \approx 13$  K which, within the errors, coincides with the temperature where  $\Delta M$  vanishes and is close to the position of the minimum in the FC susceptibility. These results corroborate the existence of a continuous phase transition which occurs within the glassy phase at nonzero applied field.

### III. SCALING ANALYSIS

In order to understand the role played by the configurational disorder and/or thermal fluctuations in this transition it

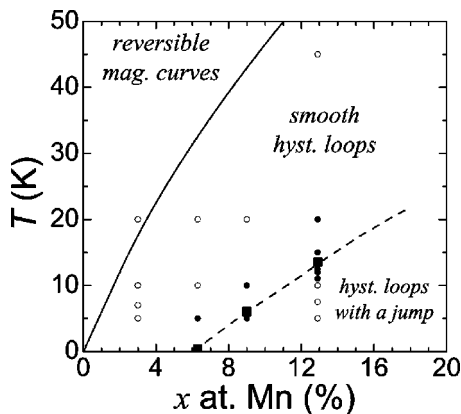


FIG. 5.  $x$ - $T$  phase diagram for the Cu-Al-Mn system at low temperatures. The measured compositions and temperatures are indicated by dots (● data used in the scaling analysis; ○ data not used). The continuous line, which separates loops with and without hysteresis, is taken from Ref. 10. The dashed line, which separates loops with and without a jump, has been estimated in the present work. The solid squares (■) correspond to the values of  $T_c(x)$  determined from the scaling analysis.

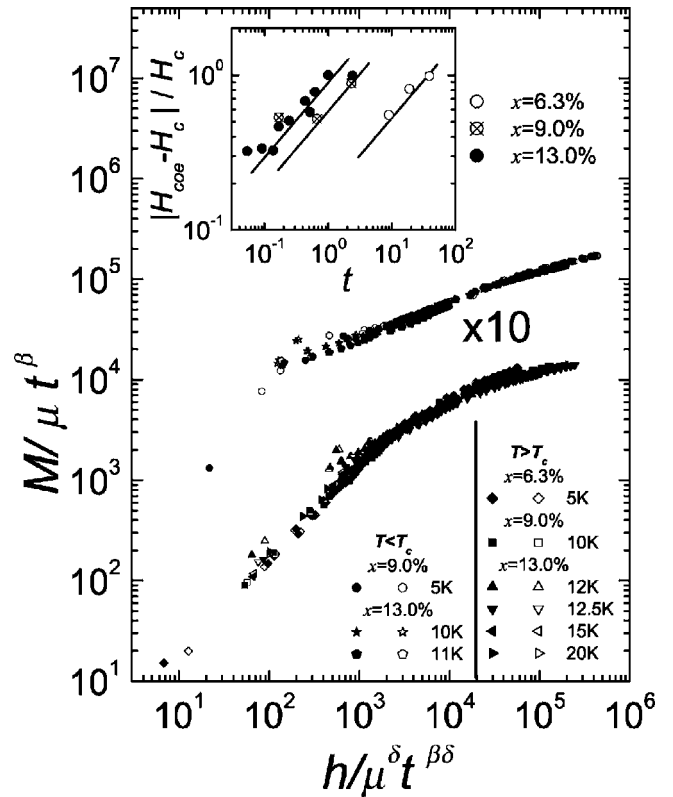


FIG. 6. Scaling of the hysteresis loops corresponding to different temperatures and compositions as indicated by the legend. The two sets of data correspond to the  $T > T_c(x)$  (lower) and  $T < T_c(x)$  (upper and multiplied by a factor 10). Solid and open symbols stand for the  $\mathcal{M}_+$  and  $\mathcal{M}_-$  branches, respectively. The inset shows in log-log scale the behavior of  $H_{coe}(x, T)$  when approaching  $T_c(x)$  for different values of  $x$ . The lines have a slope  $\beta\delta = 0.4$ .

is important to study results corresponding to other compositions for which the different stoichiometry give rise to different degrees of disorder. Besides the measurements presented above corresponding to  $x = 13.0\%$ , we have used data for polycrystalline samples with  $x = 3.0\%$ ,  $x = 6.3\%$ , and  $x = 9.0\%$ , published previously.<sup>6</sup> The behavior of the hysteresis loops for these samples is compiled in Fig. 5. In this figure, the continuous line is an estimation of the freezing temperature obtained from the susceptibility maxima (from Fig. 1 and from Ref. 10). To a good approximation, this temperature determines the spin freezing temperature in the low concentration region, and thus it separates the reversible magnetization curves from those that exhibit hysteresis. However, for concentrations larger than 8%, as can be seen in Fig. 1, the position of the peak overestimates the temperature where irreversibility occurs. Besides, the dashed line located at lower temperatures separates the smooth hysteresis loops from those that display a discontinuous jump.

To characterize the critical behavior we have performed a scaling analysis of the  $M(H)$  curves. The direct fitting of the  $\beta$  exponent from the  $\Delta M$  vs  $T$  data (in Fig. 4) does not provide reliable estimates of  $\beta$  since the values of  $\Delta M$  are strongly influenced by the numerical procedure used in the extrapolations.<sup>11</sup> For a fixed Mn content, the distance to the critical point is measured through the reduced temperature

$t = [T - T_c(x)]/T_c(x)$  and reduced field  $h = [H - H_{coe}(x, T)]/H_{coe}(x, T)$ . The standard scaling hypothesis<sup>1,7</sup> for the positive and negative hysteresis loop branches is  $M_{\pm} = t^{\beta} \mathcal{M}_{\pm}(h/t^{\beta\delta})$ . In order to scale data for different compositions, we generalize the preceding expression to include the fact that the magnetic moment  $\mu(x)$  depends on the Mn content. Renormalization of the magnetization by a factor  $\mu(x)$  yields the following scaling hypothesis:

$$M_{\pm} = \mu(x) t^{\beta} \mathcal{M}_{\pm}[h/\mu(x)^{\delta} t^{\beta\delta}]. \quad (1)$$

This hypothesis is tested in Fig. 6, where we have represented in a log-log scale  $M/\mu t^{\beta}$  versus  $h/\mu^{\delta} t^{\beta\delta}$ . Data correspond to scaling of hysteresis loops at different temperatures and compositions, as indicated by the solid dots in Fig. 5.

A very good scaling of the curves is obtained. The best data collapse corresponds to  $\beta = 0.03 \pm 0.01$  and  $\beta\delta = 0.4 \pm 0.1$ . The upper curve in Fig. 6 (which has been shifted by a factor of 10 in the vertical scale for clarity) corresponds to  $T < T_c(x)$  data and the lower curve to  $T > T_c(x)$  data. Solid (open) symbols correspond to  $\mathcal{M}_+$  ( $\mathcal{M}_-$ ) branches. The values of  $\mu(x)$  for each of the studied composition are estimated using the derivative with respect to  $H$  of Eq. (1):

$$\mu(x) \propto \left[ \left( \frac{\partial \mathcal{M}_{\pm}}{\partial H} \right)_{H_{coe}} H_{coe} \right]^{1/(1-\delta)} t^{-\beta}. \quad (2)$$

The obtained values, except for a multiplicative factor, are  $\mu(x = 6.3\%) = 0.566$ ,  $\mu(x = 9.0\%) = 0.575$ , and  $\mu(x = 12.9\%) = 0.625$ . The numerical estimations of  $T_c(x)$ , obtained from the improvement of the collapses in Fig. 6, are  $T_c(x = 6.3\%) = 0.5$  K,  $T_c(x = 9.0\%) = 6.0$  K, and  $T_c(x = 13.0\%) = 13.2$  K. These values are shown in Fig. 5 as solid squares. It is important to note that the collapse of the data corresponding to  $x = 13.0\%$ , considered separately from the other compositions (without the need of fitting  $\mu$ ), render values of  $\beta$  and  $\beta\delta$  within the same error bars.

An independent method to check the consistency of the scaling is to analyze the critical behavior of the  $H_{coe}(x, T)$  surface. According to the Clausius-Clapeyron equation, assuming the standard scaling behavior for the entropy and the magnetization, and using Griffiths equality,<sup>12</sup> one obtains

$$\left( \frac{\partial H_{coe}(x, T)}{\partial T} \right)_x \sim t^{\beta\delta - 1}. \quad (3)$$

Integration yields  $[H_{coe}(x, T) - H_c(x)]/H_c(x) \sim t^{\beta\delta}$ , where  $H_c(x) = H_{coe}(x, T_c(x))$ . The consistency of the data with this predicted power-law behavior is checked in the inset of Fig. 6. The straight lines have a slope of  $\beta\delta = 0.4$ . For the  $x = 13\%$  case, for which the amount of data is larger, we can perform a linear fit of such a behavior, rendering  $H_c(x = 13.0\%) = 560 \pm 15$  Oe. The fit is shown as a continuous line in Fig. 4.

Figure 7 shows the complete  $(H, T, x)$  metastable phase diagram. The continuous line across the  $(x, T)$  plane is the estimation of the freezing temperatures already shown in Fig. 5. The thick line is the line of critical points, at the edge of the first-order transition surface (shaded), where the magne-

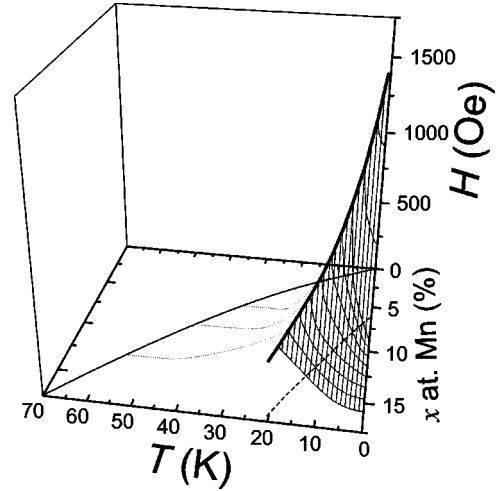


FIG. 7. Full  $(H, T, x)$  phase diagram. The thick continuous line indicates the second-order disorder induced phase transition whose projection in the  $(x, T)$  plane is shown by the dashed line. The continuous line across the  $(x, T)$  plane is an estimation of the freezing transition, where metastable hysteresis phenomena appears. The thin dotted lines indicate the behavior of  $H_{coe}$  for different values of  $x$  for  $T > T_c(x)$ . The grid indicates the first-order transition surface where discontinuity of the magnetization occurs.

tization exhibits a discontinuity. The projection of this critical line on the  $H = 0$  plane, which corresponds to the dashed line of Fig. 5, is also shown here as a dashed line.

#### IV. DISCUSSION AND CONCLUSIONS

In the present work we have reported measurements of hysteresis loops in Cu-Al-Mn alloys. The results have been interpreted in terms of a disorder-induced phase transition, and the critical exponents characterising the transition have been estimated through a scaling analysis.

The obtained values of the exponents  $\beta$  and  $\beta\delta$  are coincident, within the errors, to those reported recently in the study of Co/CoO bilayers.<sup>7</sup> In that case, the authors argue that their system exhibits a disorder-induced phase transition. The amount of disorder is controlled through the antiferromagnetic order of the CoO layer, which changes when the temperature crosses the Néel point. In our case we can guess a similar origin, since it is known that ferromagnetism and antiferromagnetism coexist in nonstoichiometric Heusler alloys.<sup>6,13-15</sup> This hypothesis is also consistent with the behavior of the hysteresis loops that become closed at relatively small fields but do not saturate up to high fields. We suggest that even at the low temperatures studied in the present work, the antiferromagnetically interacting moments act as random fields on the frozen ferromagnetic moments. Temperature changes are able to alter this effective disorder causing the disorder-induced phase transition.

It should be noted that the evolution with temperature of the hysteresis loops observed in our system is opposite to those reported for Co/CoO bilayers. While in that case the loops change from behaving continuously for  $T < T_N$  (the Néel temperature of the antiferromagnetic CoO layer) to displaying a jump when the temperature is increased above  $T_N$ ,

in our system the transition from continuous to discontinuous hysteresis loops is induced by lowering temperature. This can be understood assuming that the antiferromagnetic components of our system (essentially misplaced Mn atoms) exhibit a reentrant behavior in the presence of a high enough magnetic field,<sup>16</sup> as suggested by the FC curve in the inset of Fig. 1. This figure indicates that the temperature at which the reentrant transition of the antiferromagnetic components takes place is close to  $T_c$ . Therefore, when  $T$  is lowered below  $T_c$ , the antiferromagnetically interacting Mn atoms become aligned with the magnetic field and a significant fraction of the magnetic disorder is removed from the system.

We also want to note that the zero-temperature model for Cu-Al-Mn,<sup>6</sup> which considers the random position of Mn atoms on the four interpenetrated sublattices in the fcc structures and the coupling through an RKKY interaction, also predicts that such a system will display a disorder induced phase transition at  $T=0$  and  $x=7.5\%$ . This is very close to the estimated value of  $x$  for which  $T_c \rightarrow 0$ . As occurs with previous experimental estimations for Co/CoO bilayers, the low value of  $\beta$  is in agreement with the predicted theoretical

exponents for short-ranged zero-temperature 3D models with metastable dynamics. However, the value of  $\beta\delta$  falls out of the error bars of the numerical estimations. It is worth noting that Berger *et al.* claimed that their system, in contrast to ours, behaved as a 2D system. This is, however, in contradiction with the fact that for the 2D case, no disorder-induced phase transition is theoretically expected,<sup>17</sup> although this point needs further clarification.

In conclusion, we have proven the existence of criticality in the magnetic hysteresis loops in Cu-Al-Mn. The non-equilibrium critical line has been determined in the  $(H, T, x)$  diagram. Both temperature and composition are the variables that allow us to tune the critical amount of disorder. The coexistence of ferromagnetic and antiferromagnetic components in the system is at the origin of the observed behavior.

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