Experimental Observation of Disorder-Driven Hysteresis-Loop Criticality

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We have studied the effect of magnetic disorder on the magnetization reversal process in thin Co/CoO films. The antiferromagnetic CoO layer allows a reversible tuning of the magnetic disorder by simple temperature variation. For temperatures above a critical temperature T_c, we observe a discontinuous magnetization reversal, whereas smooth magnetization loops occur for $T < T_c$. Our measurements establish the existence of a disorder-driven critical point in the nonequilibrium phase diagram. In addition, we observe scaling behavior in the vicinity of the critical point and determine the critical exponents $\beta = 0.022 \pm 0.006$ and $\beta\delta = 0.30 \pm 0.03$ for this two-dimensional system.

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The phenomenon of hysteresis in the magnetization reversal process is a fundamental aspect in magnetism. It is also technologically significant, because magnetic storage applications are based upon this very property, which makes the study of hysteresis phenomena an eminent topic in fundamental as well as applied science [1]. The occurrence of hysteresis during magnetization reversal is easily understood as the system undergoes a first order phase transition at which the system has a multitude of metastable states [1]. The system can be driven out of its equilibrium ground state and a magnetization hysteresis is observed upon cycling the applied field. A detailed quantitative understanding of this phenomenon, however, is still missing, primarily due to the fact that all length scales from atomic to even macroscopic dimensions are almost equally important. This fundamental challenge has limited progress in the past to the development of phenomenological models only.

The recent use of computer simulations as well as the application of renormalization group methods to nonequilibrium phenomena has opened up a new chapter in this field and substantial progress has been made [2–6]. In particular, these novel theoretical approaches have been successfully applied to the description of Barkhausen jumps, i.e., magnetization avalanches, and excellent agreement between theoretical predictions and experimental results has been reported [7–17]. However, there is substantial disagreement in the literature on a more fundamental level, namely, about the existence of criticality. Based on the various models, some argue that the Barkhausen jump distribution exhibits self-organized criticality (SOC) [7,11–13], whereas others claim the absence of criticality or conventional critical behavior [2–6,9,18]. In particular, Sethna and co-workers argue for the existence of an ordinary critical point upon fine-tuning of the disorder in the random-field Ising model (RFIM) [2–6]. Their studies suggest that the hysteresis loop itself can become critical and undergo a nonequilibrium second order phase transition from a $M(H)$ loop exhibiting a discontinuity, i.e., a macroscopic Barkhausen jump, to a curve with a smoothly varying $M(H)$ dependence upon continuous variation of the magnetic disorder. The Barkhausen jump distribution exhibits scaling up to the length scale of the system size only near the critical point. This excludes the possibility of SOC, where the system is critical independent of external parameters. So far, however, the predicted critical point has not been observed experimentally [13]. Thus, a crucial link between theory and experiment is missing, which would provide a sound experimental base for the validity of the recently developed hysteresis models.

In this paper, we report the experimental observation of disorder-driven hysteresis-loop criticality for an exchange coupled Co/CoO-bilayer structure. This particular material system, in which a ferromagnetic Co film is in contact with an antiferromagnetic CoO overlayer, was chosen for two reasons. First, the thin film geometry with in-plane orientation of the magnetization suppresses the influence of dipolar effects. This is important, because true criticality should be observable only in the limit of vanishing demagnetizing effects [13]. Second, the exchange coupled antiferromagnetic (AFM) CoO layer allows the reversible tuning of the effective magnetic disorder by varying the temperature in the vicinity of the CoO Néel temperature T_N. This tunability enables a detailed study of the critical region, in contrast to an earlier experimental attempt on epitaxial Gd films [19]. Even though a 20 nm thick film constitutes a system with almost pure three-dimensional (3D) thermodynamic behavior [20], our films are two dimensional (2D) with respect to the nonequilibrium phenomena investigated here, which can be understood in the following way. A lower limit for the elemental unit, which can independently switch in a magnetic field without alteration of the magnetic state in adjacent material, is given by the domain wall width, which is approximately 50 nm for our Co films [21]. Such elemental units are the building blocks, the "atoms" of the reversal mechanism and as such zero-dimensional entities. So, a 20 nm film has the thickness of only one elemental building block and extends approximately 10^5 building blocks in each of the two lateral dimensions. The films investigated here are therefore 2D entities.

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The Co/CoO samples were prepared by dc-magnetron sputtering. A 20 nm thick Co film was first deposited onto Si substrates at room temperature. After deposition, the pure Co films were oxidized for 10 min in a plasma with oxygen pressure of 8 mTorr. The samples were then covered with a 5 nm Au layer. Low-angle x-ray diffraction measurements reveal that this procedure produces a 3 nm thick CoO film on top of the Co film. The AFM order in these CoO films has been verified by the observation of the exchange-bias effect at low temperature after cooling the sample in a magnetic field [22]. Magnetization measurements were performed using an extraction magnetometer allowing for quasistatic measurements with acquisition times of 5 sec per point during which the magnetic field is held constant. Measurements with longer acquisition times show virtually identical results.

Figure 1 shows a set of hysteresis loops at four different temperatures. For temperatures from 300 to 200 K, we observe a rectangular hysteresis loop, in which almost the entire sample magnetization switches sharply in one jump, a result that is commonly observed for well-ordered thin films with in-plane orientation of the magnetization. Upon lowering the temperature, this behavior changes dramatically in a small temperature interval around $T \approx 190$ K. At this temperature as well as for all lower temperatures, we find a substantial widening of the magnetization reversal region. Thus, the system undergoes a transition from an infinite avalanche to a gradual magnetization reversal behavior as predicted by the theory of disorder-driven criticality [4]. Besides this dramatic change in the sharpness of the magnetization reversal we also observe an exchange-bias shift of the hysteresis loops, but only at significantly lower temperatures ($T < 80$ K).

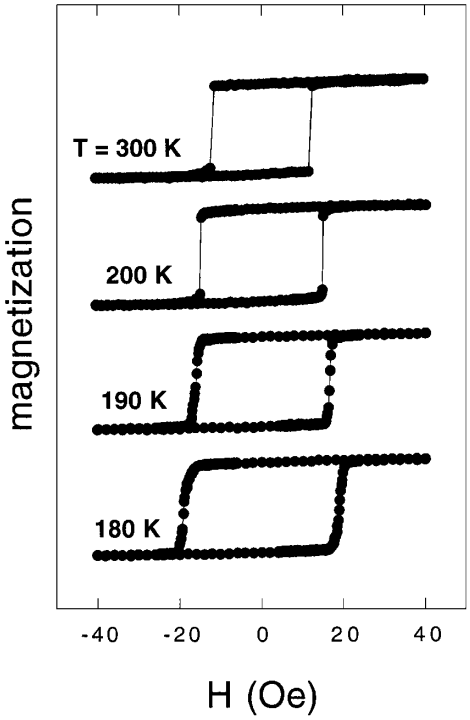

FIG. 1. $M(H)$ loops measured on a Co/CoO-bilayer structure for the temperatures indicated. The thin lines are guides to the eye.

To establish the connection between our experimental observation of criticality and the theoretical studies on disorder-driven transitions, Fig. 2 illustrates schematically the temperature dependent ordering in the Co/CoO-bilayer system. In essence, it is the interface roughness in combination with the interface exchange coupling that facilitates an effective magnetic disorder in the FM/AFM-bilayer system, which can be reversibly tuned by simple temperature variation. The property we can directly influence by varying the sample temperature is the degree of AFM order in the CoO layer. Above the Néel temperature of the CoO film [Fig. 2(a)], which has a bulk value of $T_N = 291$ K, only the ferromagnetic Co [T_C(bulk) = 1388 K] is magnetically ordered. Thus, the bilayer behaves like a single thin film by showing rectangular hysteresis loops for $T > T_N$. For temperatures near the Néel temperature, the Co film starts to induce AFM order in the CoO film due to the exchange coupling between the layers. At this point, interface imperfections cause spin frustration in the interface and introduce magnetic disorder into the system, which causes variations of the local correlation between FM and AFM order [Fig. 2(b)]. Near T_N, the AFM order in the CoO is not stable enough to induce an exchange bias, but already influences the reversal

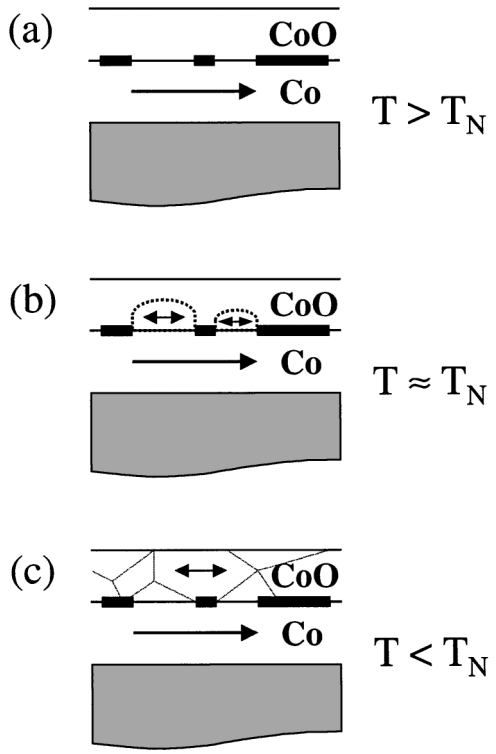

FIG. 2. Schematic of the magnetic state in a Co/CoO-bilayer for (a) $T > T_N$, (b) $T \approx T_N$, and (c) $T < T_N$. The thick black lines in the Co/CoO interface indicate areas with large interfacial roughness, where the correlation between FM order in the Co and AFM order in the CoO is diminished.

mechanism by effectively increasing the activation volume and thereby increasing H_C [23]. At lower temperatures, the effective interfacial coupling between the now stable AFM state and the FM state is also laterally varying due to the interface roughness [Fig. 2(c)]. Even though both regimes [Figs. 2(b) and 2(c)] play a role in our samples, our measurements suggest that the disorder according to Fig. 2(b) is dominant in the temperature range of interest here, because we observe exchange-bias shifted hysteresis loops only for $T < 80$ K. It should also be mentioned that the microscopic mechanisms involved in the exchange-bias effect are rather controversial and presently a field of intense research efforts [22–24].

To verify the above explanation for the observed hysteresis loop widening, Fig. 3 shows the relative hysteresis loop transition width w vs temperature for the Co/CoO bilayer (open circles) as well as for a pure Co film (closed circles). Both samples were deposited simultaneously under identical conditions. For high temperatures $T > 200$ K, both samples show identical behavior with a transition width of $w \approx 0$. However, below $T \approx 200$ K, there is a substantial increase of w for the Co/CoO-bilayer sample, corresponding to the gradual magnetization reversal seen in Fig. 1, whereas the pure Co film continues to show a discontinuous transition. Thus, the dramatic change in the reversal behavior for the Co/CoO sample cannot be a simple thermal effect caused by the reduced probability to overcome the energy barrier of an intermediate metastable state during the reversal process. Such a "freezing" would be identical in both samples. These reference measurements verify that the AFM/FM-interface coupling induced disorder is the origin of the hysteresis loop criticality observed in Fig. 1.

The existence of the critical point is visualized in Fig. 4, in which magnetization data near H_C are displayed in a color-coded image as a function of magnetic field and temperature. The full saturation magnetization is displayed as blue (positive) and red (negative) with green corresponding to $M = 0$. We see that for $T \geq 195$ K, there is a discontinuous transition from negative to positive magnetization values, whereas for lower temperatures, we find an intermediate field region, in which the sample exhibits small magnetization values (green). This "green" region of gradual magnetization reversal widens upon cooling, which corresponds to an increase in magnetic disorder.

In accordance with the observed existence of a critical point at $T \approx 195$ K, we have performed a scaling analysis of all data sets for $T < 195$ K using an approach similar to the one described in [6] for the analysis of computational results. We do not have a direct experimental measure of the magnetic disorder and can therefore not quantify it. However, the magnetic disorder itself is not a critical quantity in the temperature range of interest here, which allows us to assume a linear relationship between disorder and temperature in the immediate vicinity of the critical point. Thus, we can replace the disorder as tuning parameter with the temperature in the scaling analysis. Under those provisions, the scaling relation can be written as

$$M(\tau, h) \sim \tau^{\beta} M\left(\frac{h}{\tau^{\beta\delta}}\right) \tag{1}$$

with τ as the reduced temperature $(T_c - T)/T_c$ according to the critical temperature T_c, and $h = (H - H_C)/H_C$, where H_C is the coercive field at each individual temperature [25]. As scaling function M we used an asymmetric $\tan^{-1}$ function

$$M(x) = M_S \tan^{-1}\left[\frac{x}{(w + ax^b)^2}\right], \tag{2}$$

with M_S as the saturation magnetization, w as the relative transition width ($w \approx \delta H/H_c$), and a and b as asymmetry parameters. This function describes the individual $M(H)$ curves extremely well and proved to be a much better choice than the scaling function for the mean-field case of the RFIM, which is exact only in six dimensions [3]. Figure 5 shows the results of our scaling analysis. Clear evidence for scaling behavior

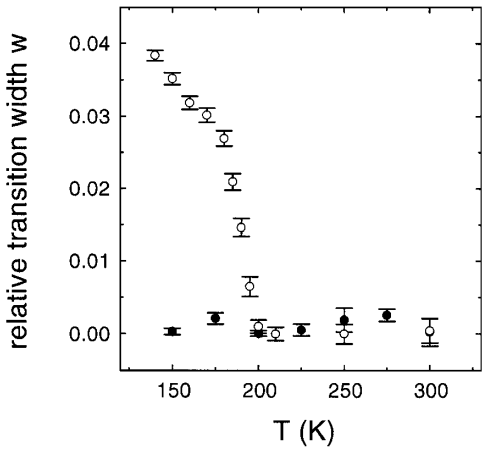

FIG. 3. Relative width of the magnetization reversal region w vs T for the Co/CoO bilayer (open circles) and a Co-film reference sample (closed circles). The values are determined from least-squares fits according to Eq. (2).

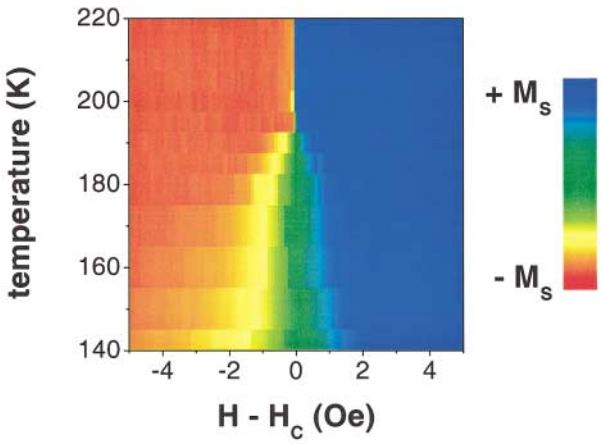

FIG. 4 (color). Magnetization $M(H, T)$ map near the critical point for the increasing field branch of the hysteresis loop. The color-coding scheme is indicated.

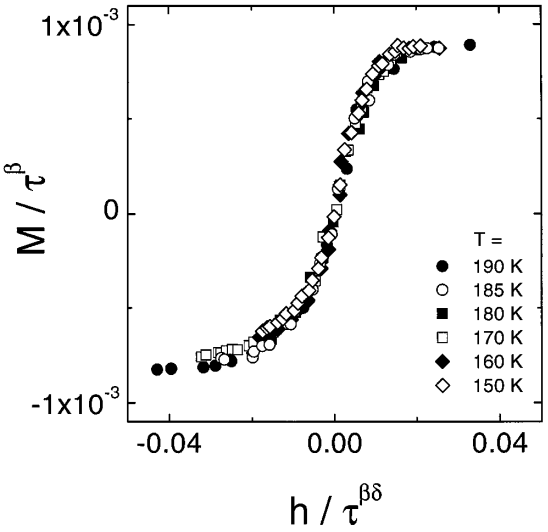

FIG. 5. Scaling analysis plot M/τ^{β} vs $h/\tau^{\beta\delta}$ for data taken in the temperature range $150 \leq T \leq 190$ K using the scaling analysis parameter $T_c = 191.4 \pm 0.5$ K, $\beta = 0.022 \pm 0.006$, and $\beta\delta = 0.30 \pm 0.03$.

in the vicinity of the critical point is the fact that all data in the range $150 \leq T \leq 190$ K can be collapsed onto one curve. From the numerical analysis, the critical temperature is found to be $T_c = 191.4$ K $\pm$ 0.5 K and the critical exponents are determined as $\beta = 0.022 \pm 0.006$ and $\beta\delta = 0.30 \pm 0.03$ for this 2D magnetic system. Unfortunately, no reliable estimates for these exponents are available in the literature for the 2D case. β is similar to the value calculated for three dimensions, but $\beta\delta$ is significantly smaller than the 3D numerical value of 1.81 ± 0.32. Besides the different dimensionality, which could be responsible for this discrepancy, one also has to realize that our measurements were not done at zero or even low temperatures in contrast to all theoretical calculations. In addition, the experimental disorder is not known in detail and the possibility of local disorder correlations cannot be excluded. On the other hand, even our polycrystalline samples should fall into the Ising symmetry class, because the remanent magnetization and the applied field break the rotational symmetry [26]. Our estimate for $\delta = 13.6 \pm 5.6$ is actually remarkably close to the value $\delta = 15$ for the thermodynamic 2D Ising model.

In summary, we have studied the effect of temperature dependent interface frustration on the hysteresis-loop criticality for exchange coupled Co/CoO-bilayer structures. Our experiments firmly establish the existence of a conventional critical point, which is associated with the level of magnetic disorder that can be reversibly tuned via temperature variation. In addition, we report scaling behavior in the vicinity of the critical point, which allows the determination of critical exponents for this 2D magnetic system. The comparison of the Co/CoO-bilayer study with the behavior of single Co films highlights the ability to reversibly tune the magnetic disorder in such bilayer samples making them a powerful tool for experimental hysteresis research.

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