Disorder-Driven Hysteresis-Loop Criticality in Co/CoO-Films

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Abstract:

The effect of magnetic disorder on the magnetization reversal process in thin Co/CoO-films has been investigated. 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 non-equilibrium phase diagram. In addition, we observe scaling behavior in the vicinity of the critical point and determine the critical exponents to $\beta = 0.022 \pm 0.006$ and $\beta\delta = 0.30 \pm 0.03$.

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The phenomenon of hysteresis in the magnetization reversal process is a fundamental aspect in magnetism. Its occurrence is easily understood as the magnetic system undergoes a first order phase transition at which it has a multitude of metastable states [1]. Therefore, the system can easily be driven out of its equilibrium groundstate 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 the 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 (RG) methods to non-equilibrium 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 very good 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 coworkers 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 non-equilibrium 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. This fundamental difference in the magnetization reversal behavior can be seen in figure 1. The upper curve displays an $M(H)$ -loop with an almost fully synchronized magnetization switch in the entire sample, which occurs for disorder parameter values R smaller than the critical disorder R_c . In contrast, the $M(H)$ -dependence for $R_c > R$ shows a very smooth and gradual magnetization reversal, which indicates a very limited spatial correlation of the reversal process. Details of the calculated results have been published elsewhere [2-6]. Within the RFIM, 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.

The discrepancies between the various theoretical models at this very fundamental level, namely the existence of criticality, make the necessity for experimental studies obvious. Only a comparison between experimental data and the different theoretical models

will allow an evaluation of their relevance for real material systems. So far, however, experimental work in this field has been rather limited due to the fact, that it is very difficult to continuously and independently tune the magnetic disorder level of materials. Only recently we have been able to observe the predicted critical point experimentally [19, 20]. Thus, a crucial link between theory and experiment has been established, which will 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 only be observable in the limit of vanishing demagnetizing fields [13]. Secondly, 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 Neel temperature T_N . This tunability enables a detailed study of the critical region, in contrast to earlier experimental studies on the annealing temperature dependence of hysteresis loop properties for epitaxial Gd-films [20]. Even though a 20 nm thick film constitutes a system with almost pure three dimensional (3D) thermodynamic behavior [21], our films are two dimensional (2D) with respect to the non-equilibrium phenomena investigated here. This can be understood in the following way. A lower limit for the elemental unit, that 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 [22]. 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 is therefore a 2D entity.

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 minutes 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 [23]. Magnetization measurements were performed using an extraction magnetometer (PPMS) allowing for quasi-static measurements with acquisition times of 5 seconds per point during which the magnetic field is held constant. Measurements with longer acquisition times show virtually identical results.

Figure 2a shows a set of hysteresis loops at four different temperatures. For temperatures from 300 K 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).

To illustrate the significance of the Co/CoO-interface for the unusual temperature dependence of the magnetization reversal process, Fig. 2b shows the relative hysteresis loop transition width $w \sim (dM/dH)^{-1}$ 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 condition. 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. 2a, 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 is essential for the hysteresis loop criticality observed in Fig. 2a.

In essence, it is the interface roughness in combination with the interface exchange coupling that is responsible for the observed hysteresis loop change because it facilitates an effective magnetic disorder in the FM/AFM-bilayer system, which can be reversibly tuned by simple temperature variation [19]. The property we can directly influence by varying the sample temperature is the degree of AFM order in the CoO-layer. Above the Neel temperature of the CoO-film, which has a bulk value of $T_N = 291$ K, only the ferromagnetic Co [$T_C(\text{bulk}) = 1388$ 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 Neel temperature, the Co-film starts to induce AFM order in 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. This local variation of the magnetic

properties then results in the fundamentally altered magnetization reversal mechanism shown in Fig. 2a. Further details about the here utilized disorder-tuning mechanism via interface frustration have been published elsewhere [19].

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, therefore, cannot 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

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

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. As scaling function M we used an asymmetric $\tan^{-1}$ -function

$$M(x) = M_s \cdot \tan^{-1} \left[\frac{x}{(w + a \cdot x^b)^2} \right], \quad (2)$$

with M_s as the saturation magnetization, w as the relative transition width, 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 6 dimensions [3]. Figure 3 shows the results of our scaling analysis. Clear evidence for scaling behavior in the vicinity of the critical point is the fact that all data in the range $150 \text{ K} \leq T \leq 190 \text{ K}$ can be collapsed onto one curve. From the numerical analysis, the critical temperature is found to be $T_c = 191.4 \text{ K} \pm 0.5 \text{ K}$ 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 very well 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 [24]. Our estimate for $\delta = 13.6 \pm 5.6$ is actually remarkably close to the value $\delta = 15$ for the thermodynamic 2D Ising model.

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Figure Captions:

- Fig. 1 Hysteresis loops obtained for the RFIM with an assumed gaussian distribution of width R for the random fields. The critical disorder level has been determined to $R_c = 2.16$ for the 3D case [6]. The upper curve has been calculated for $R = 1.7 < R_c$ and the lower one for $R = 4 > R_c$.
- Fig. 2 Temperature dependent magnetization reversal characteristics for a Co/CoO-bilayer sample: (a) $M(H)$ -loops measured for the temperatures indicated. The thin lines are guides to the eye; (b) 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 equation (2).
- Fig. 3 Scaling analysis plot M/τ^β vs. $h/\tau^{\beta\delta}$ for data taken in the temperature range $150 \text{ K} \leq T \leq 190 \text{ K}$ using the scaling analysis parameter $T_c = 191.4 \text{ K}$, $\beta = 0.022$, and $\beta\delta = 0.30$.

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Figure 1

Figure 2a

Figure 2b

Figure 3