Nonequilibrium Magnetization near the Reorientation Phase Transition of Fe/Ag(100) Films

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The static and dynamic magnetic properties of ultrathin Fe/Ag(100) films are investigated in the vicinity of the reorientation transition where the magnetic easy axis changes from out-of-plane to in-plane orientation. In a certain temperature range below the reorientation transition temperature T_r a time dependent decay of the remanent out-of-plane magnetization is observed. The origin of this relaxation behavior is discussed, and implications for a thermodynamic description of the reorientation phase transition are outlined.

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In recent years, ultrathin ferromagnetic films have attracted a tremendous amount of attention. One of the most fascinating topics in this field is the alteration of magnetic properties by the very existence of surfaces or interfaces. In particular, magnetic anisotropies in ultrathin films are strongly modified compared to bulk values, due to the broken symmetry at the interfaces [1]. This is of fundamental interest because only anisotropic systems can exhibit a long-range ferromagnetic order in two dimensions [2]. Furthermore, the ability to alter anisotropy values is of technological importance for magnetic recording applications [3].

Ultrathin Fe/Ag(100) films have been found to exhibit a strongly enhanced magnetocrystalline anisotropy with an easy axis perpendicular to the surface plane [4]. At low film thickness and temperature, this perpendicular anisotropy is sufficient to overcome the demagnetizing field and align the magnetization perpendicular to the surface plane. Furthermore, a reversible reorientation transition of the magnetization from out of plane to in plane has been observed as a function of temperature [5]. In the vicinity of this reorientation phase transition (RPT) the remanent magnetization, in plane as well as out of plane, is vanishing, which has been explained by the existence of domains in the out-of-plane phase [5–8]. But even though this RPT phenomenon has attracted a substantial theoretical interest [9–16] only very few experimental studies are available [5–8,17]. In particular, it is not clear whether the magnetic properties in the vicinity of the RPT are determined by thermal equilibrium or whether relaxation phenomena occur. This is of fundamental importance because all theoretical descriptions are based on a thermodynamic approach [9–16]. Relaxation effects near the Curie temperature have already been observed in Fe/Ag(100) films by Volkening *et al.* using Mössbauer spectroscopy [18].

In this paper, we investigate the static and *dynamic* magnetic properties of Fe/Ag(100) films in the vicinity of the RPT. For this purpose, we have performed conventional hysteresis loop measurements as well as time dependent measurements of the out-of-plane magnetization using the magneto-optical Kerr effect (MOKE). The entire experiment, film preparation and magnetic characterization, was performed under ultrahigh vacuum conditions. The Fe films were grown by evaporation from an *e* beam evaporator onto a clean and well annealed Ag(100) substrate, held at room temperature. The growth rate was chosen to be approximately 1 monolayer per min and was controlled *in situ* by a quartz monitor. Subsequently, the films were heated to 440 K for 30 min, which improved sharpness of the LEED spots significantly, as previously reported by Qiu *et al.* [8]. Using two sets of coreless coils we were able to determine the in-plane and out-of-plane magnetization components as a function of field, temperature, as well as time. In agreement with previous reports, we have found a strong thickness dependence of the reorientation transition temperature T_r [8]. Therefore, we focused our study on film thicknesses $d \approx 4$ monolayers to have the reorientation transition well within the experimentally accessible temperature range.

Figure 1 shows the temperature dependent magnetization for a 4.3 monolayer film. The out-of-plane magnetization in remanence (thin solid line) decreases strongly in a relatively narrow temperature range around $T^* = 120$ K. For lower temperatures, the remanent magnetization is equal to the saturation magnetization, which is measured here in an applied field of 90 Oe (thick solid line). In the temperature range between 120 and 210 K, the remanent out-of-plane magnetization is very small or even zero, whereas a small field of 90 Oe is still sufficient to fully saturate the film out of plane. Furthermore, no in-plane magnetization (solid squares, open circles) is observed in this temperature range. Above 210 K, the in-plane magnetization increases strongly and stays constant for $T >$ 240 K, whereas the induced out-of-plane magnetization decreases rapidly in this temperature region. This behavior can be consistently explained by a temperature dependent reorientation transition from an out-of-plane magnetized state to an in-plane state at $T_r \approx 220$ K. As shown by the schematics in Fig. 1, the magnetization is lying in the film plane above T_r. For temperatures below T_r, the easy axis of magnetization is perpendicular to the film plane. But, because of the magnetostatic energy associated with a

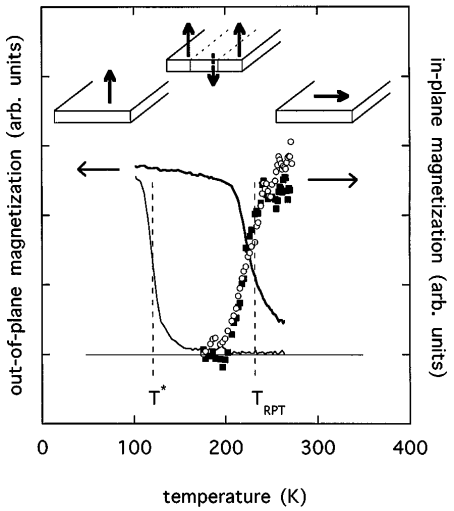

FIG. 1. Temperature dependent magnetization for a 4.3 monolayer Fe/Ag(100) film: out-of-plane magnetization (in remanence: thin solid line; in an applied field $H = 90$ Oe $\perp$ surface: thick solid line), in-plane magnetization (in remanence: ■; in an applied field $H = 50$ Oe $||$ surface: ○). The corresponding magnetization states in remanence are illustrated.

perpendicular magnetization, the system forms domains of alternating magnetization direction to minimize its energy [19]. The existence of domains, experimentally verified by Allenspach and Bischof for Fe/Cu(100) films [7], results in the absence of a remanent out-of-plane magnetization, i.e., the observed remanence gap. In agreement with the existence of a domain structure, a small field of several 10 Oe is sufficient to restore the full out-of-plane magnetization, as predicted by Kashuba and Pokrovsky [12]. Such small values of the saturation field are caused by the extremely small energy gain of multidomain structures in ultrathin films [19]. But even though the domain state is the lowest energy state for all values of the anisotropy constant $K > K_c$ [20], the remanent magnetization increases with decreasing temperature and is even equal to the saturation magnetization at low temperatures. Thus, at low temperatures they system is again in a single domain state, which as also been observed previously [5–8]. This M_r increase had been attributed to the exponential domain size increase with anisotropy; i.e., above a certain threshold value a finite sample should exhibit a single domain state [12]. But the only available experimental domain study on this temperature dependent reorientation phenomenon does not show any significant change in domain size with temperature [7]. Thus, it is an open question what determines the temperature T^* at which the single domain state in the out-of-plane magnetization occurs.

In Figure 2(a), the time dependence of the out-of-plane remanent magnetization is shown for a 4 monolayer film at various temperatures. In these experiments, a field of 90 Oe was applied perpendicular to the surface up to the time $t = 0$ (on the scale shown in Fig. 2) and the time dependent measurements were subsequently performed in

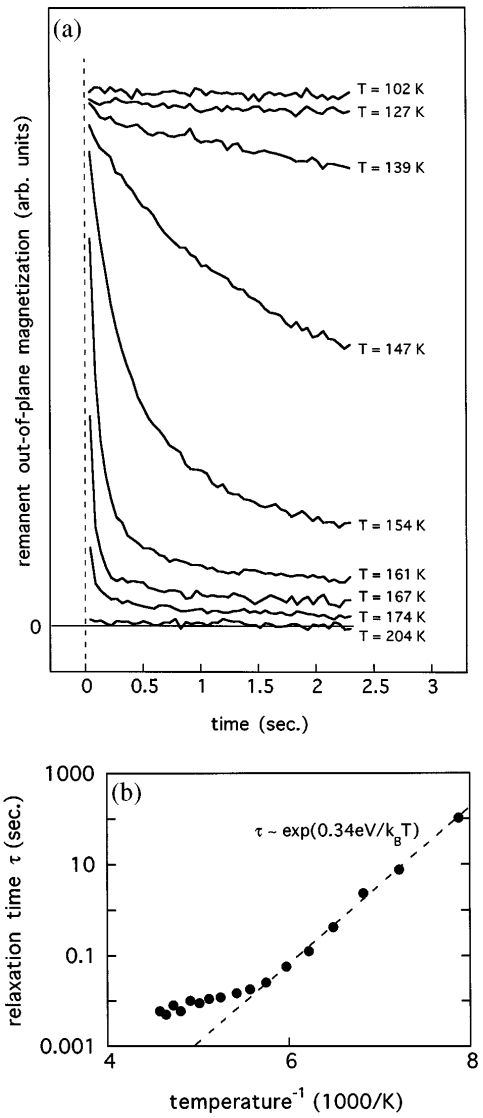

FIG. 2. (a) Remanent out-of-plane magnetization vs time measured for a 4 monolayer Fe/Ag(100) film at various temperatures; (b) relaxation time vs inverse temperature determined from the data shown in (a).

zero field. For all temperatures shown here such a field was sufficient to saturate the magnetization, i.e., produce a single domain state with out-of-plane magnetization. Thus, for $t = 0$, the film is in a single domain state independent of the temperature. The time dependence of this single domain state, however, is very different for different temperatures. For low temperatures, up to $T = 120$ K, no change, i.e., no reduction of the remanent magnetization, can be observed as a function of time. But for temperatures in the region of T^* ($\approx$160 K), where an $M_r(T)$ decrease is observed for this sample, the remanent magnetization also shows a pronounced time dependence at constant temperature. With increasing temperature, the decay of the magnetized single domain state becomes faster and, for $T > 200$ K, the decay becomes so fast that the remanent magnetization is equal to zero, even for the shortest

duration time ($t = 40$ ms) in this study. So, these time resolved measurements clearly demonstrate that the remanent out-of-plane magnetization has a pronounced time dependence; i.e., the out-of-plane magnetized single domain state has a certain lifetime, which is strongly temperature dependent. For a determination of this lifetime, we have analyzed our relaxation data [Fig. 2(a)] using an exponential decay function [21]. In Fig. 2(b), these relaxation times are plotted as a function of the inverse temperature T^{-1}. It is obvious from Fig. 2(b) that the relaxation time shows an exponential increase with increasing T^{-1}, i.e., exhibits a typical activation barrier behavior. Thus, the uniform single domain state is not unstable as discussed by Kashuba and Pokrovsky [12], but is at least metastable for temperatures $T \leq T^*$, and an activation energy is necessary to form a nucleus of reversed magnetization. From Fig. 2(b) the activation energy is determined to be $E_a = 0.34 \pm 0.02$ eV, but this value varies considerably for different samples. The saturation of the relaxation time values at $\tau \approx 10$ ms, observed for high temperature data in Fig. 2(b), is caused by the response time of our experimental setup.

In general, our time resolved measurements show that the single domain state represents a local energy minimum, as schematically shown in Fig. 3. By applying a field this state is populated because it has the lowest energy in a sufficiently large field $H > H_{cr}$, which is smaller than 90 Oe here. But even after reducing the external field to zero, the film remains in this single domain state for some time, because of its metastable nature and because the system has to overcome the activation energy to form domains first. Thus, the increase of the remanent magnetization at T^* and the existence of a single domain state below T^* is due to the dramatic increase of the lifetime of this metastable state and not caused by an equilibrium domain size comparable to the sampling size. Such an increased domain size might cause an out-of-plane $M_r(T)$ behavior as shown in Fig. 1, but does not explain the time dependence of $M_r(t)$ at a fixed temperature. Consistent with this explanation is our observation that these films can be put into a stable (lifetime $\tau > 100$ s) demagnetized state, independent of the temperature, even for temperatures $T \ll T^*$. One would not expect this to happen if the increase of M_r at T^* is caused by a domain size effect in thermal equilibrium. Thus, the occurrence of a remanent out-of-plane magnetization marks the temperature range where nonequilibrium magnetic properties become important; i.e., the thermodynamic relaxation time becomes large compared to the observation time. This can also be interpreted as a significant reduction of fluctuations for $T < T^*$ caused by the effective anisotropy. In addition, our observation of a metastable single domain state is fully consistent with spin-wave calculations for the 2D anisotropic Heisenberg model [11]. Erickson and Mills reported a softening of spin waves for anisotropy values close to the reorientation transition but for anisotropy values just slightly larger than K_c the spin-wave gap opens up again and the uniform state is at least metastable [11].

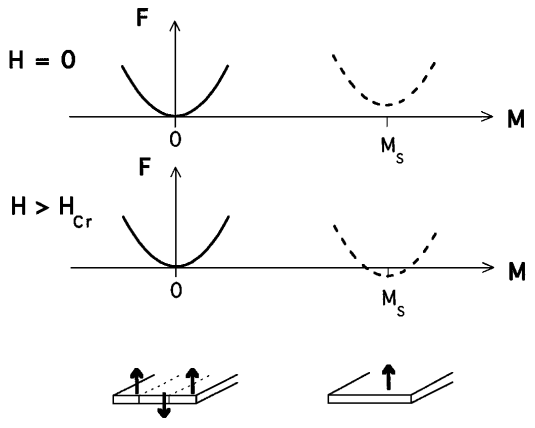

FIG. 3. Schematic of the free energy dependence on the macroscopic magnetization M for a film with out-of-plane magnetization orientation. In zero field ($H = 0$), the demagnetized multidomain state has the lowest energy, but in a sufficiently large field ($H > H_{cr}$) the single domain state is energetically favorable.

The schematic in Fig. 3 also indicates that the stability of the metastable single domain state may be enhanced by a small field parallel to the magnetization direction. By applying such a field the nucleus of reversed magnetization has an activation energy larger than in the case of a field free environment due to the additional Zeeman energy contribution. Thus, the metastable single domain state should have an increased lifetime. The effect of such a field dependent lifetime enhancement can be seen in Fig. 4, where several hysteresis curves are shown for temperatures close to T^* ($T^* \approx 270$ K for the 3.8 monolayer film shown here). For $T \approx 270$ K, the hysteresis effect in the center of the $M(H)$ loops disappears, which corresponds to a short lifetime of the single domain state at $H = 0$. However, for field values $H \approx 10$ Oe, a hysteresis effect can still be observed up to $T \approx 310$ K, which indicates that the relaxation time as well as the necessary activation energy for a decay of the single domain state is substantially increased by an applied field.

In summary, we have investigated the static and dynamic magnetic properties of Fe/Ag(100) films in the vicinity of the reorientation phase transition. For temperatures below the reorientation transition it is shown that the occurrence of a remanent out-of-plane magnetization is a relaxation time effect caused by the metastable nature of the single domain state. The temperature as well as the field dependence of this relaxation time behavior has been investigated. Our measurements also show that nonequilibrium phenomena do not play a role in the immediate vicinity of T_r, at least not on time scales investigated here, i.e., $t > 10$ ms. Thus, a thermodynamic description should be appropriate for $T \approx T_r$.

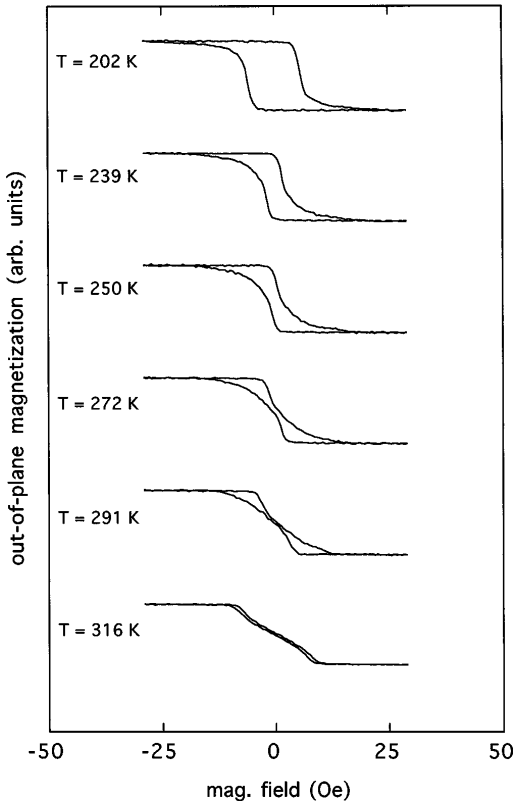

FIG. 4. Hysteresis loops measured for a 3.8 monolayer Fe/Ag(100) film with out-of-plane orientation of the applied field; temperatures as indicated.

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[20] The anisotropy constant K is defined by using $F = -K[\cos(\Theta)]^2$ as an expression for the angular dependent free energy F with Θ being the angle between the magnetization and the surface normal. K_c is the critical anisotropy constant for which the reorientation transition occurs.
[21] A detailed analysis of the curves in Fig. 2(a) shows the remanent magnetization does not exhibit an ideal exponential decay. Especially for small residual magnetization the decay is much slower than an exponential decay given by only one relaxation time τ.