# Magnetic reorientation transition of Gd(0001)/W(110) films

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The temperature-dependent magnetic properties of Gd(0001)/W(110) films were investigated using the magneto-optical Kerr effect. Measurements of hysteresis loops, remanent magnetization, and susceptibility were performed as a function of temperature and film thickness ($d = 10-130$ nm). Well annealed films of sufficient thickness ($d \geq 40$ nm) show an anomalous temperature dependence of the coercive field $H_c(T)$ and the remanent magnetization $M_r(T)$. In addition, a second peak in the in-plane as well as out-of-plane susceptibility was observed for these films. All experimental results for thick films can consistently be explained by a reorientation transition between a low-temperature phase with uniform in-plane magnetization and a high-temperature phase with a nonvanishing out-of-plane magnetization component. The thickness dependence of the transition temperature $T_r$, as well as the absence of a remanent out-of-plane magnetization implies that the reorientation transition is associated with the formation of a domain structure.

## I. INTRODUCTION

Rare-earth elements and their magnetic properties were extensively studied in the 60's and 70's. Particularly, neutron scattering played an essential role in unraveling the complicated magnetic structures in the rare earth (for reviews see, e.g., Refs. 1–3). It was found in these studies that gadolinium is the only rare-earth element with only one simple ferromagnetic phase below a Curie temperature $T_C$ of 293 K. Since Gd has a half-filled $4f$ shell with a spherical charge distribution magnetic anisotropies are small compared to the other are rare-earth elements. However, Gd shows a quite complicated temperature dependence of the magnetocrystalline anisotropy which also results in a pronounced temperature dependence of the easy axis of magnetization. Between 235 K and $T_C$ the easy axis is the $c$ axis (Gd crystallizes in the hcp structure). Below 235 K the easy axis continuously turns away from the $c$ axis up to a maximum angle of about 65° around 170 K and then turns back towards the $c$ axis to level off at an angle of 30° at low temperatures.[4] In addition, gadolinium exhibits an unusual temperature dependence of the magnetocrystalline anisotropy in the vicinity of the Curie temperature $T_C$.[5] Due to the nonideal hcp-lattice structure ($c/a = 1.59$ for Gd instead of $c/a = 1.633$ for a perfect hcp structure), the dipole-dipole interaction is anisotropic and favors a magnetization orientation along the $c$ axis.[6] Close to the Curie temperature, this dipolar anisotropy contribution becomes very important and causes an anisotropic behavior of Gd, even in the paramagnetic phase.[5]

In recent years, there has been a strong interest in the magnetism of thin Gd films, mostly due to the unusual surface magnetic properties of this material. In contrast to the ferromagnetic $3d$ metals, Gd surfaces were shown to have a ferromagnetic ordering temperature larger than the bulk Curie temperature.[7] The only other example known of this so-called surface-enhanced magnetic ordering is Tb.[8] Since rare-earth surfaces are very difficult to clean most of the recent surface studies were performed on epitaxial Gd films, especially films grown on W(110). The magnetic surface ordering of Gd(0001) has been a source of confusion. Weller *et al.*,[9] on the basis of spin-polarized low-energy electron diffraction (LEED) and photoemission studies, concluded that the Gd(0001) surface layer is aligned antiferromagnetically to the bulk. Indeed, first-principles electronic-structure calculations found the antiferromagnetic coupling energetically favorable.[10] However, recent spin-polarized experiments unambiguously show that well-prepared Gd(0001) surfaces exhibit predominantly ferromagnetic surface coupling.[11–14] This is additionally supported by recently improved *ab initio* calculations, in which the ferromagnetically coupled state exhibits the lowest energy and also results in a surface relaxation, which is in excellent agreement with observed experimental data.[15] Furthermore, it was found that the surface Curie temperature enhancement strongly depends on the surface conditions, although a systematic experimental study is not yet available. Experimental observations of a perpendicular surface magnetization component were also reported recently.[12,16]

Besides the strong interest in these special surface properties, Gd(0001)/W(110) films have been used as a model system for studies on thermodynamic properties and especially critical behavior of thin films. Due to its relatively low Curie temperature, one can investigate thin-film properties in the vicinity of $T_C$ over wide thickness range without being limited by substrate-film intermixing or other nonreversible alterations of the film structure. Thus, Farle *et al.* have been able to study the thickness dependence of $T_C$ in a range between $d = 5$ and 100 monolayers.[17] They reported a strong decrease of $T_C$ with decreasing thickness in accordance with theoretical predictions for thin films.[17–19]

It was generally believed that the magnetization in

 

these films is kept in the film plane by the demagnetizing field even though the magnetocrystalline anisotropy favors a magnetization component perpendicular to the film plane. Brillouin scattering on 30 nm films showed in-plane magnetization except at very low temperatures[20] and ferromagnetic resonance experiments implied a uniform in-plane magnetization state for films as thick as 130 nm.[21] Anomalies of the coercive field and the remanent magnetization just below $T_C$ were recently found,[22,23] but were attributed to surface effects. Furthermore, thick Gd films on Nb(110) showed anomalies in the remanent magnetization and the coercive field.[24] In a previous report, we showed that the anomalous magnetic properties of Gd films exhibit a strong thickness and annealing temperature dependence and also indicate an out-of-plane magnetization component for thick and well-annealed films.[25] Furthermore, it was shown that the anomalous magnetic behavior of thick films is not due to special surface properties. Recently, we demonstrated that thick Gd(0001)/W(110) films undergo a magnetic reorientation transition at a certain thickness-dependent temperature $T_r < T_C$.[26] Using material constants for bulk gadolinium, we also compared our experimental results to a micromagnetic model for the reorientation transition and found excellent agreement.[26] Details of this comparison will be presented in a subsequent paper.[27]

In this paper, we present in detail the results of our investigation of Gd(0001)/W(110) films. We performed *in situ* Kerr effect measurements of the hysteresis loop behavior, the remanent magnetization and the magnetic susceptibility, with in-plane as well as out-of-plane orientation of the applied magnetic field. All these quantities were studied as a function of the film thickness ($d = 10-130$ nm), temperature ($T_s = 160-300$ K) and the crystallographic order, which we were able to modify by using different annealing temperatures ($T_{an} = 570-870$ K). Section II of this paper gives a description of the experimental setup used in the present study, including film preparation and magneto-optical detection technique. In Sec. III, we present our experimental results for the various magnetic quantities and analyze their anomalous properties with respect to the magnetization state. The main results are discussed in Sec. IV and related to previous investigations.

## II. EXPERIMENTAL

Our experimental study has been entirely performed within a UHV chamber (base pressure $< 10^{-10}$ mbar) including substrate and film preparation as well as magnetic characterization. The UHV chamber was equipped with a cylindrical mirror analyzer for Auger electron spectroscopy (AES) and a commercial reverse-view LEED System. A tungsten crucible was used as a Gd evaporation source and a quartz crystal monitor enabled us to perform thickness measurements during the evaporation. As substrate, we used a mechanically polished W(110) single crystal. The substrate could be dosed with hydrogen and oxygen via two leak values. Furthermore, the UHV chamber contained an arrangement of coreless coils, which allowed us to apply magnetic fields up to 200 Oe parallel, as well as perpendicular to the film plane.

Gd films were prepared in the UHV by evaporation from a tungsten crucible onto a clean and well-annealed W(110) substrate. During deposition the substrate temperature was held at $T_s \approx 350$ K and the film thickness was monitored by a quartz microbalance. The deposition rate was chosen to be approximately 4 nm/min. After deposition, the samples were annealed by thermal treatment, i.e., held at a certain annealing temperature $T_{an}$ for 3 min. Subsequently, the film cleanness was checked via AES and the crystallographic order was examined by LEED. This procedure of low-temperature growth plus thermal annealing has been demonstrated to produce smooth and crystallographic well defined films.[28] Furthermore, it was found by other groups that there is a thickness-dependent upper limit for the annealing temperature above which the Gd films start to form islands.[28] Our annealing results are in excellent agreement with previously published data. All our films for thickness $d \geq 20$ nm have been annealed to $T_{an} = 870$ K or lower temperatures to perform studies on the effect of annealing itself.

After preparation, the Gd films were investigated by the magneto-optical Kerr effect. In this study, we measured the transverse and polar Kerr effect to determine the in-plane as well as out-of-plane magnetization component. Furthermore, we utilized our experimental setup to measure the magneto-optical response of the samples to an externally applied ac field, i.e., a quantity proportional to the magnetic susceptibility.[29] The setup used in this study is very similar to the one described by Bader[30] with the exception that we utilized the transverse instead of the longitudinal Kerr effect for in-plane magnetization detection. For measurements of the polar Kerr effect, we have applied a magnetic field perpendicular to the surface. The incoming light was chosen to be *s* polarized and the angle of incidence was set to approximately 3° to achieve a large sensitivity. The transverse Kerr effect was determined by applying a magnetic field in the film plane, perpendicular to the plane of incidence. In this case a large sensitivity was achieved by setting the angle of incidence to 45° and choosing an initial light beam polarization between pure *s* and *p* polarization.

Hysteresis loops were measured by monitoring the light intensity at the photodiode as a function of the external magnetic field. The experimental parameter were set to measure one cycle in approximately 1 s and average over ten cycles, i.e., the total acquisition time for hysteresis loops was about 10 s. For the determination of the remanent magnetization the following procedure was used. A sufficiently strong magnetic field of alternating sign, capable of reversing the magnetization, was applied to the sample. Subsequent to each field pulse, measurements of the light polarization as well as sample temperature were performed in zero field. By this procedure, the asymmetry of the light polarization between a positive and a negative field pulse was determined, i.e., a quantity which is proportional to the remanent magnetization. The total acquisition time for each data point was also set to approximately 10 s. Susceptibility measurements were performed by applying an oscillating magnetic field with

$\nu = 350$ Hz and an amplitude of 1 Oe. The magneto-optical response to this ac field results in an ac voltage at the photodiode, which was detected by a lock-in amplifier. To determine the susceptibility, this observed ac voltage was normalized to the total transmitted light intensity, which was simultaneously measured. The total acquisition time for each data point was set to 10–30 s.

After cooling the sample to $T \approx 160$ K, measurements were taken during the slow warmup. An entire warmup from $T_s \approx 160$ K to $T_s \approx 300$ K took about 2 h. Although the temperature increase was much faster at low temperatures, all measurements were performed for a temperature reading of $T_n \pm 1$ K with $T_n$ as the nominal temperature. In the case of hysteresis loop or remanent magnetization measurements, no magnetic field was applied during the preceding cooling procedure. This was not necessary, because a strong magnetic field was applied during the measurement itself. Prior to susceptibility measurements, a magnetic field $H = 100$ Oe was applied parallel to the film plane during the cool down. This was done to ensure that the sample is in a defined magnetic state during the susceptibility measurement, where no large fields are applied. A detailed discussion, how the applied field during the cooling procedure influences the measured susceptibility is given in Sec. IV, in connection with the discussion of our experimental results.

## III. RESULTS

### A. Hysteresis loop measurements

Figure 1 shows a set of hysteresis loops that were measured for (a) a 25 nm thick and (b) a 130 nm thick Gd film at various sample temperature $T_s$. Both films were prepared in the same way (annealing temperature $T_{an} = 870$ K) and the magnetic field was applied in the film plane during these measurements. The data for the 25 nm thick film, shown in Fig. 1(a), exhibit square hysteresis loop behavior which can be easily explained by a single domain state with in-plane magnetization. After

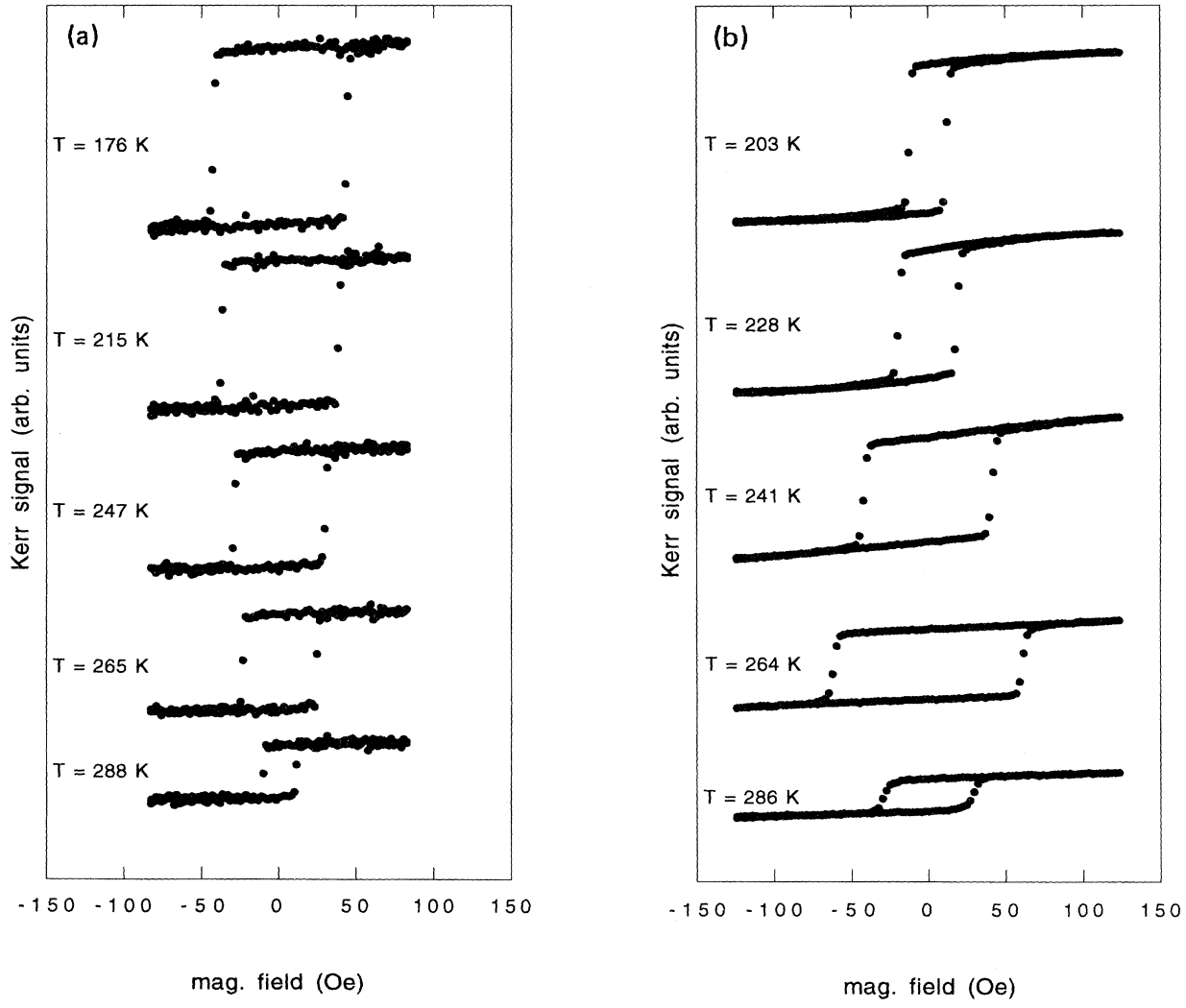


FIG. 1. Temperature-dependent hysteresis loops, measured for (a) a 25 nm thick, (b) a 130 nm thick Gd/W(110) film.

being fully magnetized by the maximum applied field strength, the sample stays in this state on reduction of the field strength and persists in this state even when the field direction is reversed, up to a certain maximum field. At this certain field strength, which is equivalent to the coercive field in this case, a stable nucleus of reversed magnetization is thermally excited and rapidly expands, leading to a fully reversed magnetization state.[31] With increasing temperature, the hysteresis loops in Fig. 1(a) show a monotonic decrease of the coercive field $H_c$. This can be explained by the fact that states with higher activation energy can be more easily excited at higher temperatures and therefore a smaller field is already sufficient to form a stable nucleus and initiate the magnetization reversal process.[31] Thus, the 25 nm thick Gd film behaves like the prototype of a single-domain particle with the external field applied along its easy axis. The magnetization behavior of the 130 nm thick Gd film, shown in Fig. 1(b) is substantially different. The most obvious difference between Figs. 1(a) and 1(b) is the fact that the coercive field is increasing with temperature for the thick film, at least for temperatures significantly lower than the Curie temperature $T_C$. This behavior is in obvious contrast to the thin-film case and the simple model for the magnetization reversal discussed there. In addition, the hysteresis loops in Fig. 1(b) exhibit a nonvanishing slope even far away from the magnetization reversal points which is most obvious for the hysteresis loops taken for $T_s = 228$ K and $T_s = 241$ K. Thus, there are some essential differences in the magnetic properties of thin and thick Gd films.

To study this difference in more detail, we investigated the temperature dependence of the coercive field $H_c$ for Gd films of varying thickness. The results for four different thicknesses are shown in Fig. 2. All films were prepared under the same conditions. In the case of the 25 nm thick film, as well as all thinner films we investigated, the coercive field decreases monotonically with increasing temperature. This behavior can be understood by assuming only one type of magnetization reversal process, as outlined before. For films with thickness $d \geq 40$ nm, the behavior is very different. All thick films are characterized by a low coercive field $H_c$ at low temperatures and also show a slight decrease of $H_c$ with temperature in this region. But upon further increase of the sample temperature $T_s$, the coercive field exhibits a strong increase, starting from a particular temperature $T_0$, which depends on the film thickness. For the 50 nm thick film, this increase of $H_c$ is observed at about $T_0 \approx 270$ K, resulting in a relatively sharp $H_c(T)$ peak just below the Curie temperature. At $T = T_C$, the long-range magnetic order is vanishing and therefore also $H_c$ has to approach zero, i.e., $H_c(T)$ decreases again in the immediate vicinity of $T_C$. For larger thickness $d$, the onset of this anomalous $H_c(T)$ increase is shifted towards lower temperatures resulting in a very broad $H_c(T)$ maximum for $d = 130$ nm (see Fig. 2). This anomalous sharp increase of $H_c$ indicates that the activation energy for the magnetization reversal process or the reversal process is altered at the onset temperature $T_0$. This can also be interpreted as a first experimental hint that even the magnetization state is altered at $T_0$.

In order to investigate the physical origin of this anomalous behavior, we have performed annealing studies for various film thicknesses. Figure 3 shows the temperature dependence of $H_c$ for a 50 nm thick film after subsequent annealing steps. Data for other film thicknesses have been reported previously.[25] As one can see from Fig. 3, the 50 nm film exhibits a monotonically decreasing coercive field for increasing temperature, i.e., no unusual behavior for annealing temperatures up to $T_{an} = 570$ K. Upon further increase of the annealing temperature, the coercive field is reduced for temperatures below $T_s \approx 270$ K. This magnetic behavior at low temperatures can be attributed to the annealing of defects. The domain-wall mobility is enhanced with improving crystal quality and results in a reduction of the coercive field.[32] In contrast to this simple behavior, a maximum of the coercive field

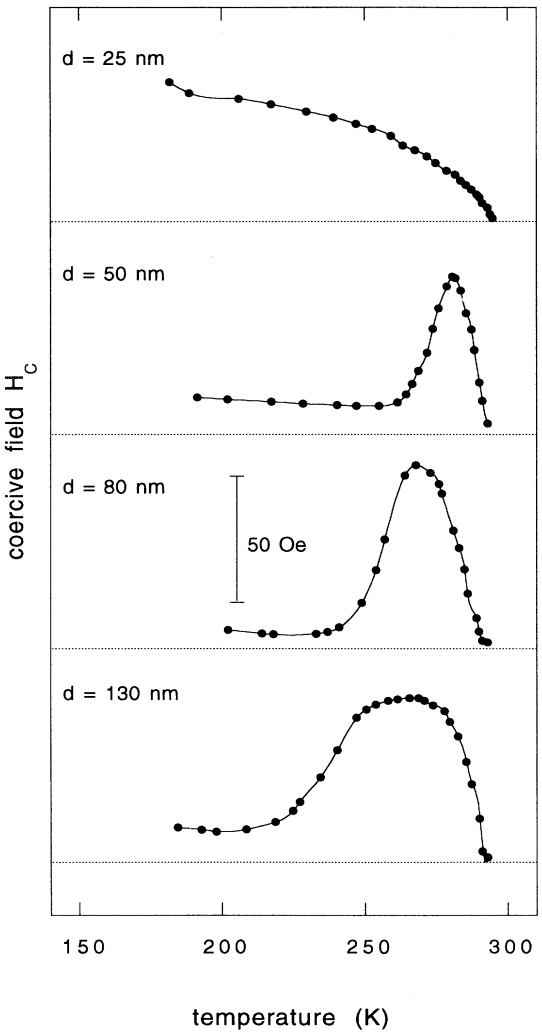


FIG. 2. Coercive field as a function of temperature for Gd/W(110) films (thickness $d$ as indicated, dotted lines correspond to $H_c = 0$ for each set of data).

starts to evolve just below the Curie temperature after annealing to $T_{an}=620$ K. Upon further increasing $T_{an}$, this $H_c(T)$ maximum becomes more and more pronounced and the coercive field at $T\approx 280$ K actually increases with increasing $T_{an}$. Similar effects of $H_c(T)$ have been found for all films with $d \geq 40$ nm. For thinner films with $d \leq 30$ nm, the annealing procedure only results in a reduction of $H_c$ in the entire temperature range.[25] So, Fig. 3 as well as all the other annealing studies show, that improvement of the crystalline structure is correlated with the occurrence of the observed anomalous $H_c(T)$ maximum, at least in films of sufficient thickness ($d \geq 40$ nm). To interpret this correlation, one has to keep in mind that the magnetocrystalline anisotropy is the magnetic property which is fundamentally related to the crystallographic order. Thus, the performed annealing studies suggest that the unusual $H_c(T)$ behavior observed for thick Gd films is related to the magnetocrystalline anisotropy in these films. To illustrate this correlation, Fig. 4 shows the observed $H_c(T)$ data for a well-annealed 130 nm thick film compared to the temperature dependent effective anisotropy constant $K_{eff}(T)$ for bulk Gd.[33] Due to the fact that the microscopic properties of well-annealed and sufficiently thick films should be nearly bulklike, one can expect to find certain similarities for $H_c(T)$ and $K_{eff}(T)$. This is exactly what can be seen in Fig. 4. $K_{eff}(T)$ exhibits a minimum at $T\approx 210$ K and a broad peak for temperatures just below the Curie temperature, very similar to $H_c(T)$. Thus, our conclusion that the anomalous $H_c(T)$ behavior is correlated with the magnetocrystalline anisotropy, drawn from our annealing experiments, is corroborated by the similarities of $K_{eff}(T)$ for bulk Gd and $H_c(T)$ for thick films.

With regard to this similarity between $K_{eff}(T)$ and $H_c(T)$, one might now argue that the unusual $H_c(T)$ dependence we observe, is simply proportional to the unusual temperature dependence of the effective anisotropy in Gd. Furthermore, the thickness dependence of this effect could be explained by a thickness-dependent anisotropy due to the residual strain which is present in thinner films. Although the coercive field generally increases with increasing anisotropy, the equivalence between $H_c(T)$ and $K_{eff}(T)$ proposed above is too simple. First, the observed thickness dependence would then suggest that the anisotropy constants even for a well annealed 80 nm thick film are substantially different from bulk Gd. This seems unlikely in light of measurements of the thickness-dependent anisotropy constants for other epitaxially grown magnetic films.[34] In these studies, a strongly thickness-dependent magnetocrystalline anisotropy has been found, but only in a thickness range of a few nm, which is far below the thickness range of interest here. In addition, one has to keep in mind that the magnetocrystalline easy axis is perpendicular to the film plane

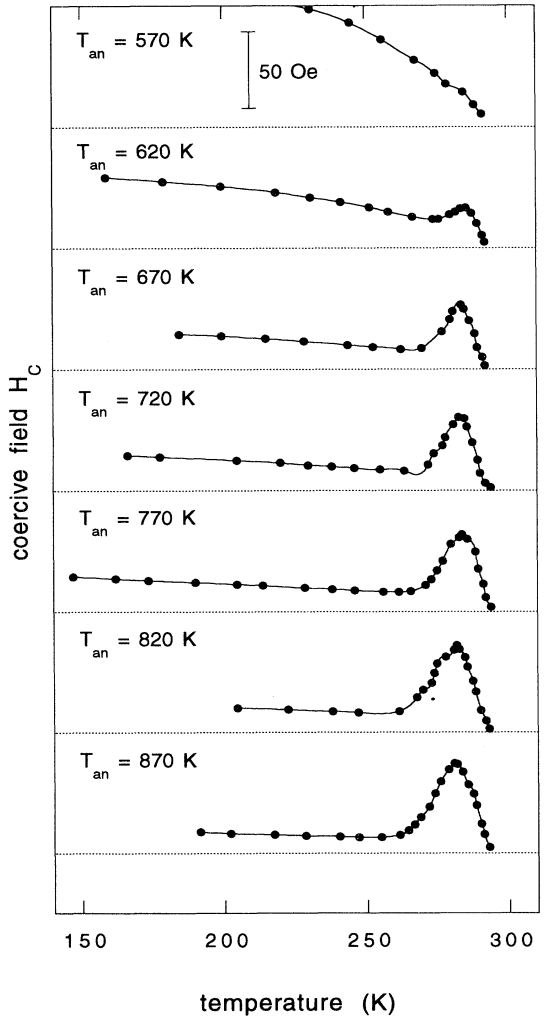


FIG. 3. Coercive field as a function of temperature for a 50 nm thick Gd film after subsequent annealing steps (annealing temperature $T_{an}$ as indicated, dotted lines correspond to $H_c=0$ for each set of data).

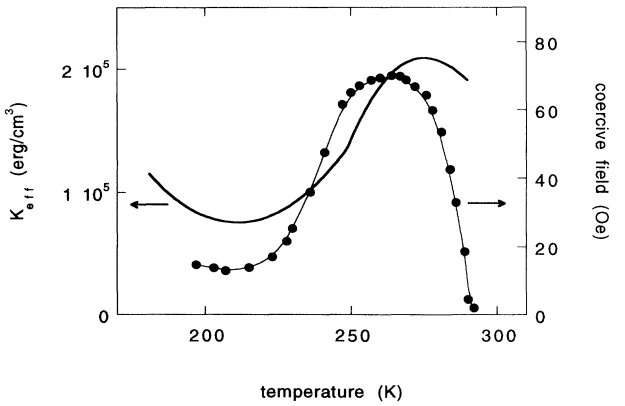


FIG. 4. Comparison between the coercive field $H_c(T)$ for a 130 nm thick Gd film and the effective anisotropy $K_{eff}(T)$ (Ref. 33) for bulk Gd.

(at least for $T_s > 235$ K.) Thus, if one assumes the magnetization to lie in the film plane, a domain wall associated with the nucleus of reversed magnetization, occurring during the magnetization reversal, would actually turn the magnetization through an easy axis (in the case of a Bloch wall). In such a case, the above-mentioned assumption, that the domain-wall energy and also the coercive field is increasing with increasing magnetocrystalline anisotropy, breaks down. So, there is no reason to believe that the coercive field in Gd films should simply exhibit exactly the same temperature dependence as the magnetocrystalline anisotropy $K_{\text{eff}}$. But nevertheless, our experiments show a strong correlation between the magnetocrystalline anisotropy $K_{\text{eff}}(T)$ and the anomalous $H_c(T)$ behavior.

### B. Remanent magnetization

Figure 5 shows the transverse Kerr signal in remanence as a function of temperature for films of different thickness. The curves for thin films ($d = 25$ and 35 nm) exhibit a temperature dependence one would expect for the saturation magnetization of a typical ferromagnet. Thus, these experiments are consistent with the assumption that thin Gd films exhibit a uniform magnetic state with in-plane magnetization. In contrast to this simple behavior of thin films, the remanent magnetization of thicker films shows anomalies. For film thicknesses $d = 40$–$65$ nm, the $M_r(T)$ curves show an abrupt change in slope at a temperature $T_d$ below the Curie temperature ($T_d \approx 275$ K for $d = 40$ nm, $T_d \approx 260$ K for $d = 55$ nm, $T_d \approx 255$ for $d = 65$ nm). For temperatures above this characteristic temperature $T_d$ up to the immediate vicinity of $T_C$, the remanent magnetization decreases more rapidly with increasing temperature than one would expect from the saturation magnetization. In even thicker films, this $M_r(T)$ slope anomaly develops into a bump structure with the characteristic onset temperature $T_d$ further decreasing with increasing film thickness. In addition, thicker films also show a positive slope for $M_r(T)$ at low temperatures, i.e., exhibit an increasing remanent magnetization with increasing temperature. Thus, thick Gd films show a temperature dependence of the remanent magnetization which is very different from thin films and very different from the temperature dependence one would expect for the saturation magnetization. So, our experimental data for the in-plane remanent magnetization $M_r(T)$ clearly indicate that in thick films, $M_r(T)$ is smaller than the saturation magnetization $M_s(T)$, at least in a certain temperature range just below the Curie temperature. This can be confirmed by a measurement of the squareness $S$, which is defined as the ratio of the magnetization in remanence to the saturation magnetization. Thus, in the case of a uniform magnetization state, one expects $S$ to be very close to 1 for all temperatures except in the immediate vicinity of $T_C$.[35] In our experiment, the magnetic field was restricted to less than 200 Oe. Therefore, our experimentally determined quantity $S_{\text{exp}}$ is given by $S_{\text{exp}} = M(H = 0\ \text{Oe})/M(H = 100\ \text{Oe})$ and can be regarded as an upper limit for the true squareness $S$. This quantity is shown in Fig. 6 for a 130 nm thick film as a function of temperature and is additionally compared to the measured temperature dependence of the remanent magnetization $M_r(T)$ for this particular film. It is obvious from Fig. 6 that $S_{\text{exp}}$ is significantly smaller than 1 with a pronounced minimum at $T \approx 240$ K. Thus, the comparison of $S_{\text{exp}}(T)$ and $M_r(T)$ clearly shows, that the strong reduction of $M_r(T)$, starting at $T \approx 220$ K is associated with a reduction of the squareness in this particular temperature range. This has also been found for all other films with $d \geq 40$ nm, which exhibit the anomalous $M_r(T)$ behavior shown in Fig. 5. Therefore, we have confirmed that remanent and saturation magnetization are not identical for sufficiently thick Gd films at elevated temperatures. Thus, thick Gd films ($d \geq 40$ nm) cannot exhibit a state of uniform in-plane magnetization in the entire temperature range.[36]

The anomalous temperature dependence implies that Gd films of sufficient thickness undergo a reorientation transition as a function of temperature. Such a reorienta-

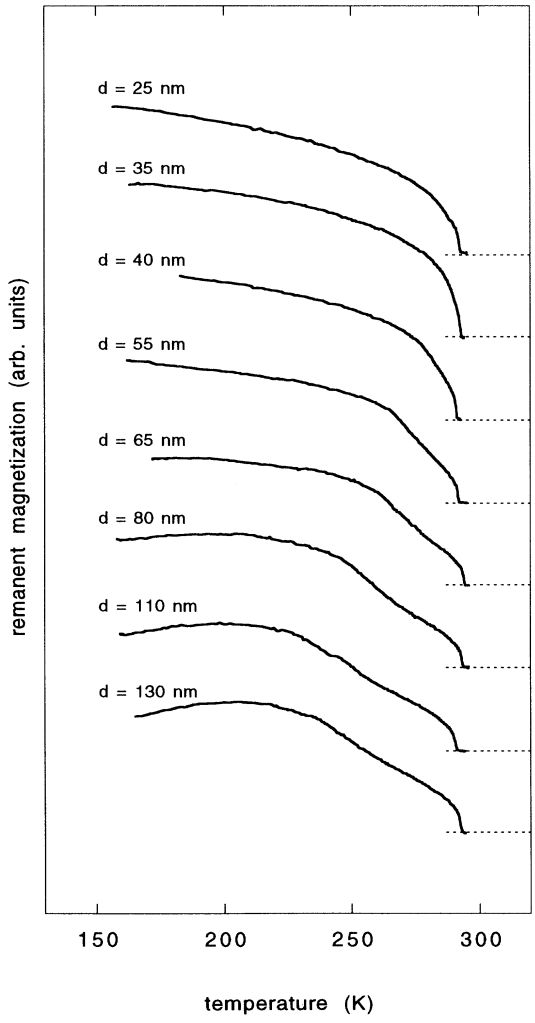


FIG. 5. Remanent magnetization as a function of temperature for Gd/W(110) films (thickness $d$ as indicated, dotted lines correspond to $M_r = 0$ for each set of data).

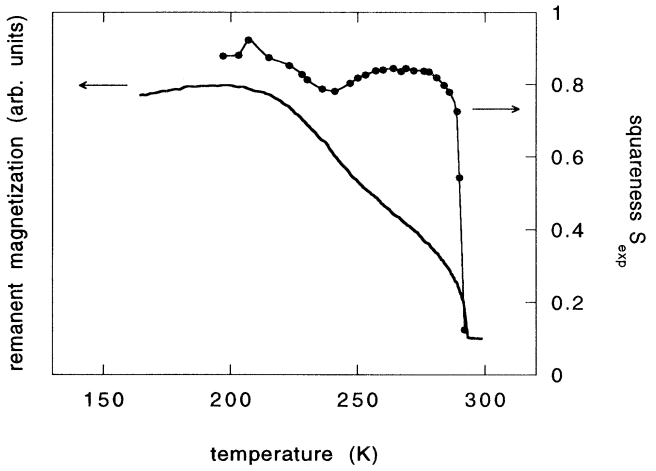


FIG. 6. Comparison between the temperature dependent squareness $S_{exp}(T)$ and the temperature-dependent remanent magnetization $M_r(T)$ for a 130 nm thick Gd film.

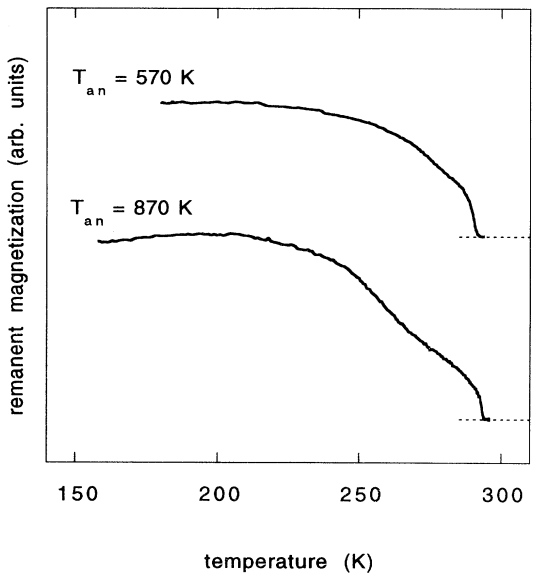


FIG. 7. Remanent magnetization $M_r(T)$ for a 80 nm thick Gd film after two subsequent annealing steps (annealing temperature $T_{an}$ as indicated, dotted lines correspond to $M_r=0$ for each set of data).

tion of the magnetization is well known from bulk Gd, but it was generally assumed that this transition is suppressed in films by the dominating effect of the demagnetizing field.[21] In bulk Gd, the reorientation is due to a very pronounced temperature dependence of the anisotropy constants $K_1$ and $K_2$.[33] For other materials, temperature-dependent reorientation transitions have also been observed in ultrathin films, where strong interface anisotropy contributions stabilize an out-of-plane magnetization state at low temperatures.[37] With respect to the assumption that the anomalous $M_r(T)$ behavior in Gd films represents a reorientation of the magnetization caused by the anisotropy, one would expect the $M_r(T)$ characteristic to change substantially with the anisotropy. As previously discussed in connection with the hysteresis loop measurements, one can vary the anisotropy in these Gd films to a certain extent by varying the annealing temperature $T_{an}$. Thus, we have performed $M_r(T)$ measurements for films of various thicknesses as a function of the annealing temperature. Figure 7 shows an example for $M_r(T)$ measurements performed on a 80 nm thick film after annealing to $T_{an}=570$ K and $T_{an}=870$ K, respectively. The lower curve in Fig. 7, representing the well-annealed film, exhibits the previously described anomalous behavior with a very pronounced bump in a temperature range between $T\approx 245$ and 290 K. The $M_r(T)$ curve for $T_{an}=570$ K looks very different with respect to this anomaly. Although there still is a deviation from the expected saturation magnetization behavior detectable in these data for $T_{an}=570$ K, it is hardly visible and much weaker than in the case of the well-annealed film. The same characteristic changes have also been found for other thick Gd films. So, these annealing experiments clearly demonstrate that the anomalous $M_r(T)$ dependence found in thick Gd films is strongly influenced by the crystal quality and is therefore strongly correlated with the magnetocrystalline anisotropy in these films.

Because of the quite low anisotropies of bulk Gd, a coherent rotation of the magnetization out of the film plane at the reorientation transition is not very likely. Therefore, our results also imply that the reorientation transition at a temperature $T_r$ is taking place via domain formation. This assumption is also supported by the strong thickness dependence of the anomalous $H_c(T)$ and $M_r(T)$ effect, indicating a strong thickness dependence of $T_r$. One would not expect any thickness dependence for the reorientation transition in the case of coherent rotation, assuming thickness-independent material constants. Thus, a thickness-dependent $T_r$ could only be explained in this model by an equally strong thickness dependence of the anisotropy or saturation magnetization which is very unlikely in the thickness range investigated here. In contrast to this picture of a coherent rotation, a reorientation transition via domain nucleation will exhibit a strong thickness dependence, due to the fact that the exchange energy associated with a domain structure can be substantially lowered by an increased film thickness.[38] Therefore, one would expect that the region of stability for a domain state is enhanced with film thickness. This is exactly what we observe in our experiments. With increasing thickness, the onset temperatures $T_0$ and $T_d$ are lowered and therefore the temperature range exhibiting anomalous behavior is increased. Thus, the anomalous properties of Gd(0001)/W(110) films we have found in our experimental investigation, can be fully understood by the assumption of a reorientation transition via domain formation in these films.

### C. Susceptibility measurements

To confirm our interpretation of the anomalous $M_r(T)$ and $H_c(T)$ behavior in thick Gd films, we have performed susceptibility measurements on these films. Due to the fact that the suggested magnetization reorientation is a phase transition, one would expect a peak in the susceptibility in addition to the peak caused by the ferromagnetic-paramagnetic phase transition. This is exactly what we have observed, as shown in Fig. 8. Here, the magneto-optical response to a small in-plane ac field, i.e., the in-plane susceptibility is shown as a function of temperature for Gd films of various thicknesses.

For thin films ($d \leq 25$ nm), only one sharp peak is observed at $T \approx 292$ K, caused by the ferromagnetic-paramagnetic phase transition at the Curie temperature $T_C$. This result is in excellent agreement with $H_c(T)$ and $M_r(T)$ measurements, in which no anomalous properties and therefore no indication for a second phase transition has been found in this thickness range. In contrast to this simple behavior of thin films, all measurements on thicker films do indeed show a second susceptibility peak, verifying the existence of a second phase transition in thick Gd(0001)/W(110) films. For $d = 35$ nm this second peak occurs only 5 K below the dominating peak at the Curie temperature. This measurement also demonstrates the superiority of susceptibility measurements for the detection of phase transitions. $M_r(T)$ for this film did not show any obvious anomalies (see Fig. 5 for comparison), whereas the occurrence of a second peak in the case of the susceptibility measurement can be easily distinguished from the usual critical behavior in the vicinity of $T_C$. With increasing film thickness, the position of this second susceptibility peak is shifted towards lower temperatures and levels off at about $T \approx 220$ K for $d \geq 95$ nm. In addition, one also observes that the peak becomes very wide in this thickness range and can hardly be associated with a sharp phase transition.

Thus, our results show that thick Gd films undergo a transition from a state with uniform in-plane magnetization at low temperatures to a state with a locally varying out-of-plane magnetization component, i.e., a multidomain state at elevated temperatures up to the Curie temperature.[39] The fact, that the domain phase is the high-temperature phase, cannot easily be deduced from the susceptibility data alone, but is obvious from the $M_r(T)$ measurements, where a reduced remanent magnetization has been observed in a certain temperature range just below $T_C$. The phase transition does also substantially alter the magnetic properties of these Gd films resulting in the observed anomalous $M_r(T)$ and $H_c(T)$ behavior. The correlation of these effects becomes obvious from Fig. 9, where the reorientation transition temperature $T_r$, deduced from the susceptibility maximum, is compared to the characteristic onset temperatures $T_d$

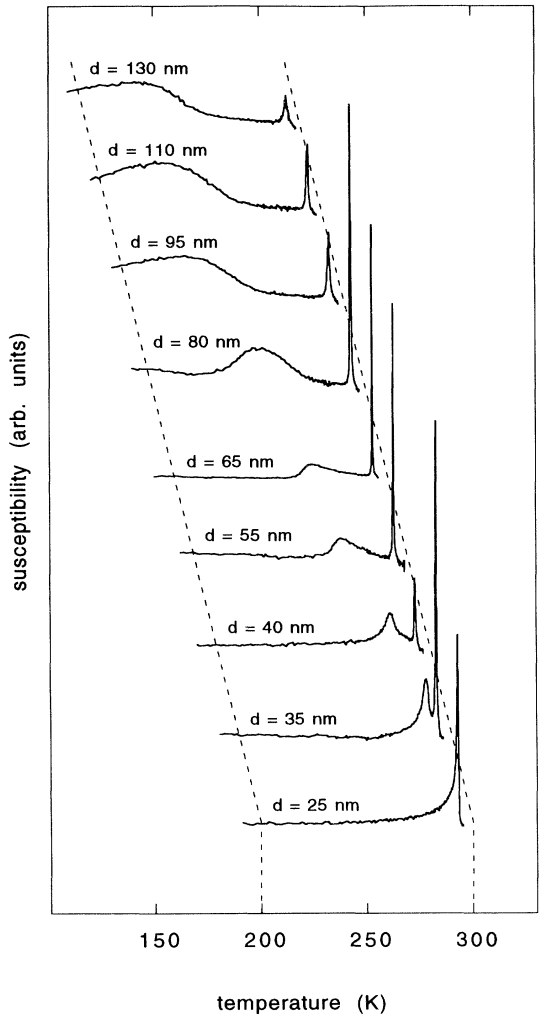


FIG. 8. Temperature-dependent susceptibility, measured for Gd/W(110) films of different thickness $d$.

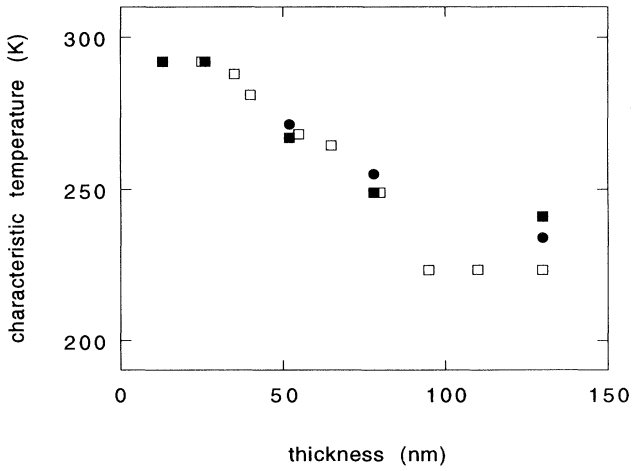


FIG. 9. Thickness dependence of the characteristic temperatures $T_0$(●), $T_d$(■), and $T_r$(□) observed for $H_c(T)$, $M_r(T)$, and $\chi(T)$ measurements.

and $T_0$ for the $M_r(T)$ and $H_c(T)$ anomalies. All temperatures are essentially the same and exhibit the same pronounced thickness dependence.

In accordance to the outlined model of an anisotropy induced reorientation transition in thick Gd films, the $\chi(T)$ characteristic should be altered by a variation of the annealing temperature. Figure 10 shows one example of an annealing study, measured on a 65 nm thick film. At low annealing temperatures, equivalent to very poor crystallographic order, there is hardly any coherent anisotropy present in these films. Thus, the demagnetizing field is dominating the thin-film behavior and no reorientation transition occurs, i.e., only one susceptibility peak at $T_C$ is observed (see Fig. 10, $T_{an}=570-620$ K). With improved crystalline quality due to higher annealing temperatures, the effect of magnetocrystalline anisotropy is increased and it becomes finally large enough to overcome the demagnetizing field and cause a reorientation transition. This can be clearly seen in Fig. 10, where the susceptibility exhibits a second maximum only after annealing to at least $T_{an}=720$ K. With subsequent annealing steps, this second susceptibility maximum is only slightly shifted towards lower temperatures, but becomes significantly sharper which can be associated with a narrowing of the anisotropy distribution during the annealing process. Thus, these measurements indicate that we are able to modify the width of the anisotropy distribution only, i.e., we cannot really tune the anisotropy by varying the annealing temperature. But, in general this annealing experiment demonstrates that a reduction of the sample anisotropy, or more precisely anisotropy coherence, results in a disappearance of the second susceptibility peak, which is in full agreement with our interpretation as a reorientation phase transition.

All the previously shown results were measurements of in-plane magnetic properties. From these measurements, we have deduced the existence of a reorientation transition in thick Gd films with a nonvanishing out-of-plane magnetization component in the high-temperature phase. Therefore, one should also be able to detect this reorientation transition as a peak in the out-of-plane susceptibility. Figure 11 shows a susceptibility measurement perpendicular to the surface for a 130 nm thick film, detected via the polar Kerr effect and compared to the equivalent in-plane measurement. As one can easily see, the out-of-plane susceptibility does exhibit an extremely strong peak at $T\approx 220-230$ K, exactly in the same temperature range where the in-plane measurement has a pronounced peak. Furthermore, polar Kerr effect measurements show that even for the very thick films there is no remanent out-of-plane magnetization component

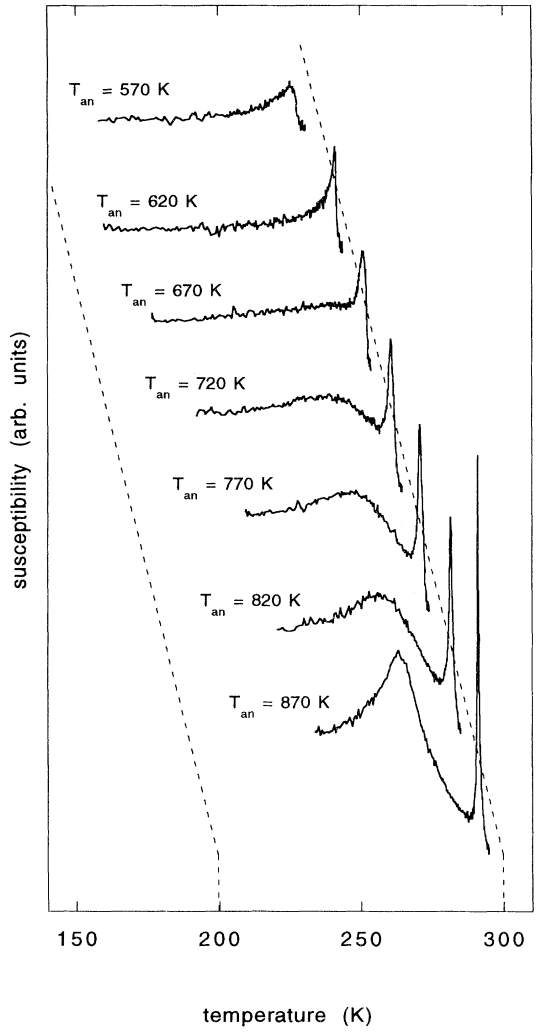


FIG. 10. Temperature-dependent susceptibility, measured for a 65 nm thick Gd film after subsequent annealing steps (annealing temperature $T_{an}$ as indicated).

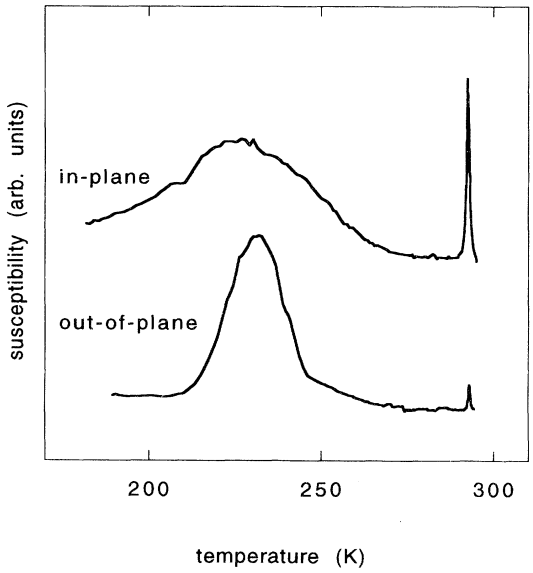


FIG. 11. Comparison between the temperature-dependent in-plane and out-of-plane susceptibility for a 130 nm thick Gd film.

detectable in the entire temperature range. The absence of a remanent out-of-plane magnetization is fully consistent with the existence of a multidomain state at high temperatures, i.e., the occurrence of a locally alternating out-of-plane magnetization component. Thus, the performed polar Kerr effect measurements also confirm our picture of a reorientation transition via domain nucleation, occurring in thick Gd films.

## IV. DISCUSSION

In this study, we have investigated the temperature-dependent magnetic properties of Gd(0001)/W(110) films *in situ* under UHV conditions. Using the transverse, as well as the polar Kerr effect, we have performed measurements of hysteresis loops, remanent magnetization and susceptibility as a function of temperature and film thickness ($d = 10-130$ nm). For well-annealed films of sufficient thickness ($d \geq 40$ nm), we observe an anomalous temperature dependence of the coercive field $H_c(T)$ and the remanent magnetization $M_r(T)$. In addition, susceptibility measurements on these films showed a second peak with the magnetic field applied parallel as well as perpendicular to the film plane. All results on thick films can consistently be explained by the occurrence of a reorientation transition between a low-temperature phase with uniform in-plane magnetization and a high-temperature phase with a nonvanishing out-of-plane magnetization component. For thinner films ($d < 30$ nm), no anomalous behavior and no second susceptibility peak was observed. Thus, in thin Gd films the reorientation transition seems to be suppressed by the dominating influence of the demagnetizing field. Furthermore, the strong thickness dependence of the transition temperature $T_r$ implies that the reorientation transition is associated with the formation of a domain structure. This is additionally confirmed by a vanishing remanent out-of-plane magnetization in the entire temperature range.

Although previous experimental studies on Gd films assumed the magnetization to lie in the film plane, the occurrence of a reorientation transition with increasing film thickness for a material with the magnetocrystalline easy axis perpendicular to the film plane is no surprising result. As early as 1946, Kittel deduced the appearance of such a transition from a total-energy calculation using a domain theory approach.[38] Later, this problem of a reorientation transition in thin films was even rigorously solved within micromagnetic theory.[40] Furthermore, calculations have been performed on the dynamic properties of thin films in the vicinity of the reorientation transition.[41] Experimentally, stripe domains have been observed for Gd films in transmission electron microscopy investigations, but were ascribed to magnetoelastic effects.[42] The unusual result of our study is the temperature dependence of this reorientation transition. In general, the magnetocrystalline anisotropy decreases with increasing temperature much more rapidly than the order parameter, i.e., the saturation magnetization.[43] Therefore, one expects the anisotropy to influence the low temperature, but not the high-temperature behavior of magnetic materials. Thin films with an easy axis perpendicular to the film plane should exhibit an anisotropy determined stripe domain state with locally varying out-of-plane magnetization as the low-temperature phase. At elevated temperatures, the demagnetizing energy, associated with the saturation magnetization should dominate the thin-film properties and cause a transition to a state with uniform in-plane magnetization. Our experiments on Gd(0001) films show exactly the opposite behavior with an out-of-plane magnetization state at high temperatures and a uniform in-plane magnetization state at low temperatures. This surprising result can be fully understood by the unusual anisotropy properties of gadolinium. Micromagnetic calculations, using material constants for bulk gadolinium are in excellent agreement with the observed experimental data and will be discussed in a subsequent paper.[27] It can also be seen from this comparison that the unusual anisotropy properties of Gd at high temperatures, which stabilize the out-of-plane magnetization state, are due to the strong dipolar anisotropy contribution of the nonideal hcp lattice structure.[27] To our knowledge, such a reorientation transition in the vicinity of the Curie temperature has not been observed before.

The results for very thick films ($d \geq 95$ nm) show certain features which differ slightly from the outlined phase transition picture and have to be explained separately. First, the susceptibility peak at low temperatures becomes very broad (see Fig. 8) and secondly, the remanent magnetization decreases with decreasing temperature in the low-temperature region (see Fig. 5). This behavior can be explained by the fact that there is no real reorientation phase transition for films thicker than a certain critical thickness $d_c$. As shown in Fig. 4, $K_{\text{eff}}(T)$ has a pronounced minimum at $T \approx 210$ K, but is always positive, i.e., $K_{\text{eff}}(T)$ favors an out-of-plane magnetization component for all temperatures. So, there exists a critical thickness $d_c$ for which even the minimum value of $K_{\text{eff}}(T)$ is sufficient to stabilize a multidomain state with an out-of-plane magnetization component. For $d \geq d_c$, the multidomain state is stable in the entire temperature range and no reorientation phase transition occurs. Thus, the susceptibility peak at $T \approx 220$ K for $d \geq 95$ nm corresponds to the minimum of $K_{\text{eff}}(T)$ and is not associated with a true phase transition anymore, which explains the observed broadening. Furthermore, the increasing $K_{\text{eff}}(T)$ at low temperatures causes an increasing out-of-plane magnetization component which is consistent with the experimentally determined reduction of the remanent in-plane magnetization for decreasing temperatures in this thickness range.

Our measurements do not show any indication, that the magnetization is switching back into the film plane in the vicinity of $T_C$. It seems that the anisotropy determined multidomain state persists up to the Curie temperature. In the case of thin Gd films, no second susceptibility peak was observed. Thus, no transition into a static multidomain structure occurs, at least not in an experimentally detectable temperature region up to about 1 K below the Curie temperature. Therefore, the phase transition at the Curie temperature is of particular interest, due to the fact that the $T_C$ region might be determined

by two overlapping phase transitions and exhibits a crossover characteristic. Unfortunately, our present experimental data do not have sufficient precision to analyze the critical behavior unambiguously.

Regarding the phase diagram of thick Gd films, it is obvious why we applied a bias field during the cooling procedure prior to susceptibility measurements. During the cooling process, thick Gd films undergo two phase transitions. At the Curie temperature, these films form a multidomain state with an alternating out-of-plane magnetization component first. Afterwards, the magnetization flips into the film plane at the transition temperature $T_r < T_C$. But this reorientation transition does not necessarily guarantee that the in-plane magnetization state at low temperatures is a single-domain state. Instead, the low-temperature phase might be a metastable multidomain state. The occurrence of such a metastable multidomain state with in-plane magnetization in the vicinity of the reorientation transition has actually been observed by Allenspach, Stampanoni, and Bischof for ultrathin Co/Au(111) films.[44] Such a multidomain state causes a nonvanishing susceptibility at low temperatures due to a field-dependent domain-wall displacement. To avoid these complications and produce a single domain state in the low-temperature phase, we have applied a magnetic field of 100 Oe parallel to the film plane during the cooling procedure. Figure 12 shows a comparative study for the in-plane susceptibility after cooling with and without bias field. Both measurements were performed under identical conditions on a 100 nm thick film. The susceptibility data clearly show a strong enhancement in the low-temperature region when no bias field was applied during the cooling procedure in excellent agreement with the assumption of domain-wall contributions to the susceptibility in a metastable multidomain state.

In addition to measurements in a temperature range between 160 and 300 K, we have also performed several attempts to detect the surface enhanced magnetic order above the bulk Curie temperature. For this purpose, we measured the magneto-optical properties immediately after film preparation and surface quality check during the cool down from about $T \approx 350-290$ K. We did not find any indication of surface magnetic order or other unusual magnetic properties above the bulk Curie temperature, although an estimate of our sensitivity showed that we would have been able to detect ferromagnetic order in one monolayer, assuming bulklike magnetic and magneto-optical properties. However, this results does not necessarily imply that there is no surface-enhanced magnetic order present in our samples. For instance, a reduced magneto-optical coupling constant might have reduced the signal level to values below our detection limit.

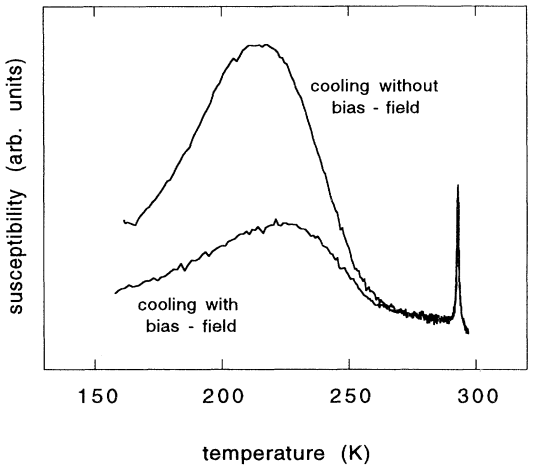


FIG. 12. Comparison between two temperature-dependent in-plane susceptibility measurements for a 100 nm thick Gd film: (a) after cooling with bias field, (b) after cooling without bias field.

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