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Possible evidence for a first-order magnetic phase transition on the Gd(0001) surface

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The magnetic phase transition at the Gd(0001) surface is found to be strongly dependent on the presence of an external magnetic field during cooling across the surface critical temperature $T_{C,s}$. Surface enhanced magnetic order on Gd(0001), present at up to 22 K above the bulk Curie temperature $T_{C,b}$, can be almost quenched if cooling is performed under zero magnetic field conditions. These observations are found on the surface of bulklike epitaxial, remanently magnetized Gd(0001) films probed with spin-polarized low-energy electron diffraction. In connection with recent theoretical work of Sanchez and Morán-Lopez, who predict magnetic-field-dependent surface first-order phase transitions on close-packed (111) surfaces of fcc spin-$\frac{1}{2}$ Ising ferromagnets, the above observations provide perhaps evidence for the existence of surface first-order phase transitions on the hcp(0001) surface of Gd.

INTRODUCTION

The surface magnetism of Gd(0001) has gained considerable attention since convincing evidence for the existence of surface ferromagnetic order at temperatures up to 22 K above the bulk critical temperature $T_{C,b}=293$ K was provided.[1-3] Those results, obtained in magnetic-field-free conditions on epitaxial thin films (thickness: several 10 nm) represent the behavior of the surface of a bulk crystal (so-called semi-infinite system) and show that in Gd(0001) the surface-atom magnetic coupling J_1 is enhanced above a critical value J_c. Rau and Eichner[4] have earlier reported on the observation of surface magnetic order at $T > T_{C,b}$ on polycrystalline Gd films by means of electron-capture spectroscopy performed in nonzero magnetic field conditions. An appropriate scaling ansatz to extrapolate their data to zero magnetic field recently performed by Eichner[5,6] indeed shows that an enhancement of the surface magnetic coupling can be derived from those earlier experiments (see also Ref. 7).

Interesting electron-spin resonance (ESR) measurements on epitaxial Gd films on W(110) a few atomic layers thick have been lately performed by Farle and Baberschke.[8] They show that the Curie temperature of monolayer thin Gd films is reduced below that of bulk Gd. The bulk sensitive ESR experiment is consequently interpreted in terms of a lowering of the nearest-neighbor coordination and of the size effect present in very thin films. We mention that there has also been intense activities in the theoretical aspects of order-disorder phenomena of semi-infinite systems.[9-12]

Both types of experiments, surface and bulk sensitive, are therefore complementary and show that the magnetic coupling in Gd strongly depends on the sample size and can both be enhanced [surface of bulk Gd(0001) crystal] or reduced [monolayer thin Gd(0001) film].

A new magnetic surface phenomenon is reported in the present paper. We find that the surface Curie point of atomically clean Gd(0001) depends on the thermal treatment of the sample, especially if an external magnetic field is applied while cooling across $T_{C,s}$. This observation can be seen in connection with recent theoretical work performed by Sanchez and Morán-Lopez,[13,14] who report on the possibility of a surface first-order phase transition in Ising ferromagnets. In the frame of their model one would then expect the surface transition temperature $T_{C,s}$ to depend on the external magnetic field present while changing the temperature across the Curie point. In the light of those new theoretical predictions our findings suggest that the magnetic surface phase transition in Gd(0001) might be of first order under certain experimental conditions. It is characterized by an abrupt decrease of the surface magnetization at $T_{C,s}$ in qualitative agreement with some of our experimental observations. The experimental details of the apparatus have been described elsewhere.[2,3] For the sake of a clear understanding of the nature of the surface transition experimentally observed we here give a more detailed description of the technique used to determine the surface magnetization of Gd(0001).

EXPERIMENT

The present spin-polarized low-energy electron-diffraction (SPLEED) experiment relies on the determination of I_σ^μ, the diffracted electron beam intensities corresponding to the four possible combinations of the relative orientation of the incident electron spin polarization σ and the magnetic moments μ of the target. Low-energy electrons strongly interact via Coulomb scattering $V_{\text{Coul}}=V_0$ with electrons in solids; their mean free path is only a few angstroms. When dealing with magnetic materials, in addition to the Coulomb and spin-orbit interaction V_{so}, magnetic exchange V_{ex} contributes to the scattering amplitude. The magnetization of the surface region is related to the exchange asymmetry A_{ex} which can

be determined from I_σ^μ according to

$$A_{ex}=\frac{1}{|\mathbf{P}_0|}\frac{I_+^+-I_-^++I_+^--I_-^-}{I_+^++I_-^++I_+^-+I_-^-} . \quad (1)$$

In the above expression $\sigma=\pm 1$ represents the spin states of the incident electrons with respect to the normal of the scattering plane defined by

$$n=\mathbf{k}_1\times\mathbf{k}_2/|\mathbf{k}_1\times\mathbf{k}_2| , \quad (2)$$

where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the incident and scattered electron momenta. $\mathbf{P}_0$ is the spin-polarization vector of the incident electrons. The quantity $\mu=\pm 1$ stands for the magnetization vector (**M**) antiparallel respectively parallel to **n**. In the present experimental geometry $\mathbf{P}_0$ is normal to the scattering plane. In order to eliminate systematic errors it is convenient to measure the scattering asymmetries

$$A^\mu=(I_+^\mu-I_-^\mu)/|\mathbf{P}_0|(I_+^\mu+I_-^\mu) . \quad (3)$$

Thus, for instance, A^+ is generated by reversing the polarization of the incident electrons (at frequencies of about 7 Hz) while keeping the magnetization of the sample constant. After a predetermined number of flips of the polarization of the incident electron beam, the magnetization of the target is reversed by a short magnetic field pulse and the corresponding asymmetry is determined (A^-). The cycle is repeated until satisfactory statistics are achieved. The magnetically induced scattering asymmetry is then calculated by taking

$$A_{ex}=(A^+-A^-)/2 . \quad (4)$$

It is important to note that during data collection (I_σ^μ) at a given temperature T the external magnetic field is turned off, i.e., the measurements reflect the remanent magnetization of the surface region of the sample.

RESULTS

Figures 1 and 2 show typical results obtained from freshly *in situ* prepared Gd(0001) epitaxial films on W(110). In Fig. 1 measurements of a 50-nm-thick film in the temperature range between 210 and 320 K (scattering parameters as indicated in the figure) are shown. The sample was cooled from deposition temperature 720 K down to about 210 K without applying an external magnetic field. The subsequently measured exchange asymmetry A_{ex} as a function of (increasing) temperature (filled stars) in the presence of a pulsed, switching magnetic field of ± 4 kA/m results in a surface Curie temperature $T_{C,s}=295\pm 1$ K which is slightly (2 K) higher than the bulk Curie temperature $T_{C,b}$. Note the abrupt decrease of the surface magnetization at $T_{C,s}$. Upon further heating A_{ex} remains zero. Cooling and simultaneously pulsing the magnetic field on and off as described in the previous section now results in the second curve (filled dots) clearly revealing surface-enhanced magnetic ordering in the presence of a paramagnetic bulk crystal in the temperature range $T_{C,s}=312\pm 1$ K $> T_{C,b}=293\pm 1$ K as has been reported earlier.[1-3] The feature marked T_{Comp} and the corresponding local maximum of A_{ex} at $T_{C,b}$ have earlier been interpreted and experimentally confirmed to be due to at least partial antiparallel spin orientation of the topmost surface layer and the underlying bulk layers.[1]

The above experimental observations show that the surface magnetic order present in remanently ferromagnetic Gd(0001) films is strongly dependent on the presence of an external magnetic field when cooling across $T_{C,s}$ (see also Ref. 3). We observe an increase of $T_{C,s}$ of 17 K

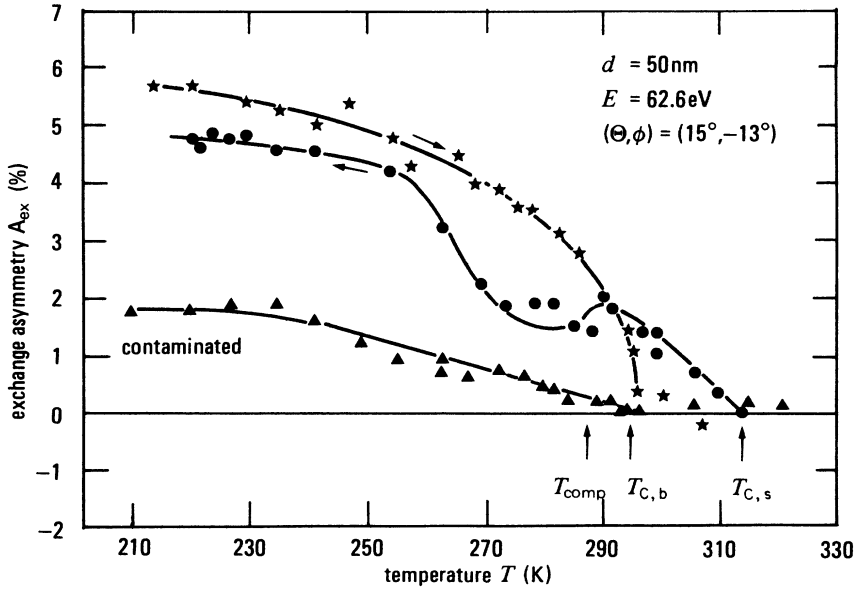

FIG. 1. Exchange asymmetry A_{ex} as a function of increasing temperature of a 50-nm-thick clean Gd(0001) film measured after first cooling to 215 K (from epitaxy growth conditions) in the absence of an external magnetic field. Filled dots represent the A_{ex}-vs-T curve measured after subsequent cooling in a pulsed switching magnetic field of ± 4 kA/m.

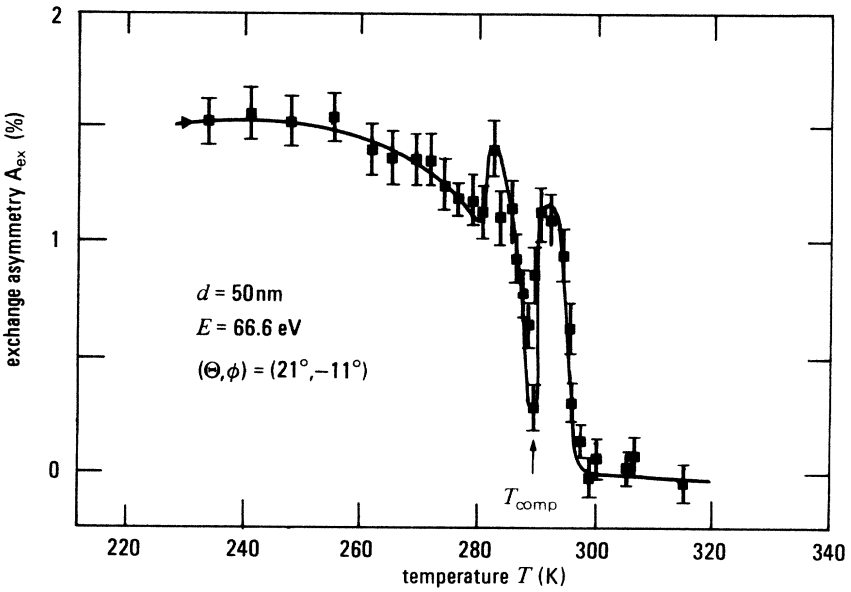

FIG. 2. As in Fig. 1. The sample was first cooled in the presence of a pulsed, switching magnetic field of ± 4 kA/m to 235 K. Surface magnetic order is clearly observed up to $T_{C,s} = 296$ K. The abrupt transition within 2–3 K at $T_{C,s}$ is a direct indication on the presence of a first-order surface phase transition.

(from 295 to 312 K) in the present case by application of an external magnetic field of ± 4 kA/m. Furthermore the T_{Comp} feature evolves, indicating a compensation between nonparallel surface layer spins and underlying bulk spins. In other words, the surface-enhanced magnetic order can almost completely be quenched (within 2 ± 1 K) when cooling is performed under field-free conditions. It is important to note that $T_{C,s}$ is also affected by residual gas adsorbates. Also shown in Fig. 1 is the case of Gd(0001) surface contaminated by residual gases, mainly CO and CO_2, which reveals $T_{C,s} = 293 \pm 1$ K $= T_{C,b}$. These tend to steadily decrease A_{ex} and $T_{C,s}$ so that the maximum value of $T_{C,s}$ might be somewhat higher than the measured 312 K. The present result was obtained after leaving the sample for about 24 h at 1×10^{-10} mbar base pressure.

Data of another so far unpublished measurement are shown in Fig. 2. The freshly prepared 50-nm-thick Gd(0001) film was cooled from its deposition temperature of 720 K down to 235 K in the presence of an external magnetic field of ± 4 kA/m and then measured up to 320 K. The exchange asymmetry shows an extremely sharp structure at $T_{Comp} = 290$ K a pronounced maximum at 293 K, which is exactly the bulk Curie temperature $T_{C,b}$ and falls off abruptly to zero within 2–3 K above $T_{C,b}$. This clearly shows the presence of surface magnetic order different from the bulk in this sample. $T_{C,s}$ can be extrapolated to 296 K, which is only 3 K but significantly higher than $T_{C,b}$. The abrupt decrease of A_{ex} near $T_{C,s}$ might be a direct indication for the presence of a first-order magnetic surface phase transition. We note, however, that this abrupt decrease is atypically not reproduced near $T_{C,s}$ for the data of Fig. 1. The data of Fig. 2 were measured with a much better statistics than those of Fig. 1 in order to establish the sharpness of the discussed features. This explains the much lower surface Curie temperature $T_{C,s} = 296$ K in that case, because measuring time was longer, and consequently the surface contamination level was higher in the case of Fig. 2.

CONCLUSIONS

The results presented here confirm the existence of a magnetic surface phase transition in the Gd(0001) surface and show that it can be almost quenched by cooling across the surface Curie point $T_{C,s}$ in the absence of an external magnetic field. The experimental results suggest the existence of a surface first-order transition, as predicted in the frame of a simple model by Sanchez and Morán-Lopez. More experimental work has to be done to further characterize the field dependence of $T_{C,s}$.

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