Spin-polarized photoemission study of epitaxial Gd(0001) films on W(110)

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Surface magnetic properties of epitaxial Gd(0001) films grown on W(110) were studied with spin-polarized resonant $4f$ photoemission. Films grown at room temperature and subsequently annealed to 550 °C show 66% polarization at 150 K, whereas films grown at 400 °C show only 46% polarization. Both types of films exhibit surface enhanced magnetic order, with the highest observed surface critical temperature exceeding the bulk value by as much as 60 K. For the first time, a rich variety of novel surface magnetic phenomena, i.e., sizeable perpendicular polarization component and unusual temperature hysteresis in the spin polarization, are observed, demonstrating that surface magnetic reconstruction is present in Gd(0001) films.

Magnetism at surfaces and in ultrathin films offers a great variety of phenomena different from the corresponding bulk behavior.[1] Surface enhanced magnetic order (SEMO) and surface magnetic reconstruction (SMR) are among the most intriguing phenomena in surface magnetism. While extensive studies on $3d$ transition-metal surfaces have failed to demonstrate the existence of such exotic surface effects, rare-earth ferromagnetic materials, Gd in particular, have proven to be promising candidates. Evidence of SEMO was first found by Rau and Eichner[2] on polycrystalline Gd surfaces with spin-polarized electron-capture spectroscopy. Using spin-polarized low-energy electron diffraction (SPLEED), Weller *et al.*[3] later convincingly demonstrated this effect on single-crystalline Gd(0001) surfaces and showed that the surface Curie temperature (T_{CS}) may exceed the bulk value ($T_{CB}=293$ K) by as much as 22 K. In the same study, the authors reported evidence of SMR on Gd(0001) which they interpreted as antiferromagnetic coupling between the surface and bulk moments. This conjecture was strongly supported by a recent electronic structure calculation on Gd(0001) by Wu *et al.*,[4] who found that the state of antiferromagnetic surface-bulk coupling is energetically favored over that of ferromagnetic coupling. On the experimental side, however, no progress has been made in clarifying the surface magnetic structure of Gd(0001). Moreover, confirmation of SEMO on Gd(0001) with other experimental techniques has yet to be achieved. Further studies of the surface magnetism of Gd(0001) are therefore highly desirable.

We have employed spin-polarized $4f$ photoemission to study the surface magnetism of Gd(0001) films. The high surface sensitivity coupled with the relative simplicity in interpreting the data, i.e., the spin polarization of the $4f$ electrons can be straightforwardly translated into the magnetic moment, makes this technique ideally suited. The photoemission experiments were performed at the Stanford Synchrotron Radiation Laboratory in a UHV system (base pressure 2×10^{-10} Torr) described elsewhere.[5] Gd was evaporated from an e-beam heated W crucible onto a single-crystal W(110) substrate at a rate of 0.7–1.5 Å/sec to a thickness of 400–450 Å. Low-energy electron diffraction and Auger electron spectroscopy were used to verify the structure and surface cleanliness of the films. Two types of Gd films were made under different growth conditions: one at an elevated substrate temperature of 400 °C, and the other at room temperature followed by annealing to 550 °C. Both types of films showed sharp LEED patterns and AES showed only trace amount of oxygen contamination (O/Gd < 5%). After preparation each film was cooled down to about 150 K and simultaneously magnetized with a nominally in-plane magnetic field of a few hundred oersted whose direction was about 15° off one of the three symmetry axes in the hcp(0001) basal plane. Photoelectrons were excited with ultraviolet light incident on the surface at 45° off normal and collected along the normal direction. The photoelectrons were energy selected through a 90° spherical analyzer and transported to a spin analysis chamber where all three components of the spin vector were measured using two medium-energy retarding field Mott detectors. All photoemission measurements were performed on samples in their remanent state.

Most of our measurements were performed using 149 eV photons. At this photon energy, photoemission of the $4f$ electrons in Gd may occur not only through the direct photoexcitation channel but also through the $4d$-$4f$ resonant excitation channel.[6] The latter process is known as resonant $4f$ photoemission and may significantly enhance the emission cross section. This enhancement is crucial to our experiments since it allows for the measurement of the polarization spectrum of the $4f$ emission peak with sufficient statistics within a relatively short period of time (~ 1 h) before the film surface gets contaminated by residual

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gases, which can otherwise be a severe problem because of the high chemical reactivity of Gd. As resonant photoemission may involve the spin-flip processes,[7] which tend to reduce the polarization of the photoelectrons; however, it is important to examine if the polarization of the resonantly emitted $4f$ electrons remains the same as that of the directly emitted $4f$ electrons. We thus performed a "control" experiment with 110 eV photons such that $4f$ photoemission may occur only through the direct photoexcitation channel. The results with respect to the polarizations of the $4f$ photoelectrons in the two cases were identical within experimental accuracy. This finding is in agreement with the results reported in Ref. 8, where experiments were done on monolayer thick polycrystalline Gd films on Fe. With the confirmation of this identity, spin-polarized resonant $4f$ photoemission can then be simply applied to study the surface magnetic properties of Gd(0001) films.

Figure 1 shows the intensity and polarization spectra across the $4f$ peak at 150 K from a room-temperature grown sample subsequently annealed for 5 min at 550 °C. The polarization has only an in-plane component in the direction of the applied field. The lack of sizeable variations in the polarization across the $4f$ peak and the large polarization values strongly indicate that surface and bulk moments are primarily ferromagnetically aligned in these films. The observed 66% polarization of the Gd $4f$ electrons at 150 K is considerably larger than previously reported results.[8,9] In order to better understand the data we have employed the deconvolution procedures described in Refs. 3 and 10 and fitted simultaneously the intensity and polarization spectra by using a bulk and a surface $4f$ component, both of Doniach-Sunjic line shape, and a background, convoluted with a Gaussian profile to take into account instrumental broadening. The surface $4f$ binding energy shift with respect to the bulk $4f$ component is taken to be 0.48 eV (Ref. 10) and fixed during the fit. Assuming ferromagnetic surface-bulk coupling, the best fit yields 68% and 61% polarizations for the bulk and surface $4f$ electrons, respectively, and a surface to bulk intensity ratio of 0.4. The smaller polarization for the surface $4f$ component is consistent with the well-known result that spin waves are more easily excited at a surface than in the bulk. On the other hand, it may also indicate the existence of possible SMR (see discussions below) which will also result in a reduced polarization. Clearly, however, the scenario of antiferromagnetic surface-bulk coupling can be excluded in these films solely based on the large polarization value which translates into nearly 100% at 0 K. The intensity and polarization spectra from films grown at 400 °C are virtually identical to those of the room-temperature grown films except that the polarization is only about 46% at 150 K, which is considerably smaller than that of the room-temperature grown films. Presumably, the difference in the polarization can be attributed to different surface morphologies of the two types of films since Gd is known to grow in a layer-by-layer mode at room temperature whereas at elevated substrate temperatures three-dimensional growth occurs and results in rougher surfaces.[11,12]

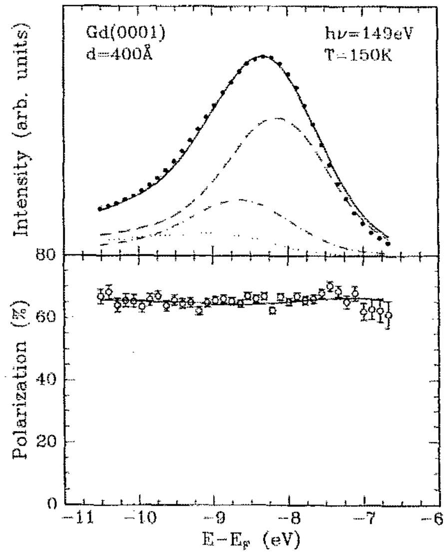

FIG. 1. Intensity (upper panel) and in-plane polarization (lower panel) spectra across the $4f$ emission peak. The various curves are obtained from the best fit: broken—bulk, dot-broken—surface, dotted—background.

In order to use spin-polarized $4f$ photoemission as surface magnetometry for Gd, we relaxed the energy resolution of the analyzer to 2 eV so that the entire $4f$ emission peak was measured. This led to further improvement in counting rates, making it possible to take densely spaced (in terms of temperature) polarization data with an accuracy of better than 1%. Figure 2 shows the temperature dependence of the spin polarization for a room-temperature grown and annealed film. A large SEMO of about 60 K over T_{CB} is readily recognizable. Although the magnitude of the polarization above T_{CB} is relatively small, the excellent statistics warrants the authenticity of the effect. This giant SEMO is significantly larger than the highest SEMO of 22 K previously reported.[3] It is important to stress, however, that in the present experiment the sample was left in its remanent magnetic state during the entire temperature scan whereas in the previous SPLEED experiment the sample was remagnetized at each temperature step. Therefore, the present data contain in its sign the added information on the relative directions of the polarizations below and above T_{CB}. The lack of a sign reversal of the polarization on going through T_{CB} supports our earlier conclusion that the surface and bulk moments are primarily ferromagnetically coupled.

More results of temperature-dependent measurements are shown in Fig. 3. These data are all obtained from films grown at 400 °C. To take the data shown in Fig. 3(a), the sample was first cooled from above 300 K in a nominally in-plane external field to about 250 K, then the spin polarization was measured on cooling down to below 150 K and warming up to 350 K. While the appearance of an anomalous outward perpendicular polarization component in excess of 10% around 225 K in the cooling cycle is striking, the temperature hysteresive behavior in the in-plane

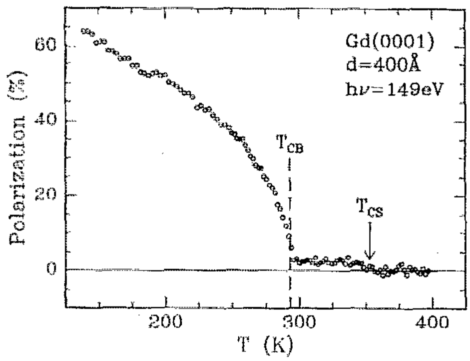

FIG. 2. Temperature dependence of the spin polarization of the $4f$ electrons showing giant SEMO.

component along the field direction over a temperature range of about 40 K below 200 K is equally intriguing. The appearance of the perpendicular component is clearly a surface effect since the dominant shape anisotropy of the film, which is about 450 Å thick, would easily force the bulk magnetization into the film plane even though the bulk magneto-crystalline anisotropy may favor a perpendicular magnetization.[13] The temperature hysteresis in the in-plane component is also likely due to surface effect as no such behavior has been observed in bulk Gd. The data shown in Fig. 3(b) were obtained during the warming cycle from a sample which was first field cooled to 250 K and subsequently zero field cooled to below 150 K. In contrast to the data in Fig. 3(a), a finite, albeit small, inward perpendicular component persisted in the warming cycle to 25 K above T_{CB}. The direction of the perpendicular component seems to depend on stray magnetic field at the sample. Figure 3(c) displays another example in which a large SEMO of about 33 K is observed.

Both of the observed surface magnetic behaviors, perpendicular polarization component and temperature hysteresis, are in fact manifestations of the more general phenomenon known as SMR. Our more recent experimental studies by means of spin-polarized secondary electron emission revealed that similar SMR phenomena are also shared by films grown at room temperature and subsequently annealed.[14] Therefore, such surface magnetic reconstructions are intrinsic to Gd(0001) films and the method of sample preparation is not critical. The size of the effects of SEMO and SMR, however, depends on the thermal and magnetic history of a sample.

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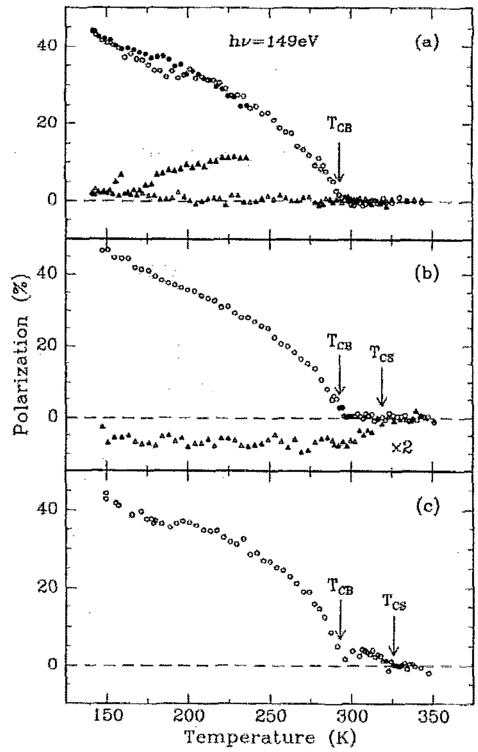

FIG. 3. Results of polarization versus temperature for several 450-Å-thick Gd(0001) films: Filled symbols—cooling; open symbols—warming; circles—in-plane component; triangles—perpendicular component. See text for sequence of experimental procedures during data taking.

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