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ABSTRACT

With spin polarized electron spectroscopies, we have investigated ordered Gd(0001) films deposited on W(110). The photoemission features of the gadolinium 5d surface state, the 4f levels, and the background exhibit considerable spin polarization along the same direction in the plane of the film, indicative of ferromagnetic coupling between the surface and the bulk. The 4f spin polarized photoemission data provides strong evidence that the surface 4f polarization differs from the bulk 4f polarization for Gd(0001). Our temperature dependent measurements with spin polarized secondary electron spectroscopy conclusively establishes that the surface of clean Gd(0001) possesses a perpendicular polarization component which persists to an enhanced surface Curie temperature. Small amounts of contamination at the surface result in the disappearance of the perpendicular component and, therefore, a more perfect ferromagnetic coupling between the surface and the bulk.

INTRODUCTION

The issues of surface magnetic anisotropy and the magnetic coupling between the surface and the bulk have attracted considerable attention. The Gd(0001) surface appears to exhibit enhanced magnetic order, i.e., higher Curie temperature at the surface than in the bulk [1]. It is seen to be able to maintain an in-plane remanent magnetization [1, 2, 3, 4], although there is indication that an perpendicular anisotropy exists for ultrathin Gd films[4]. Calculations [5] supported the altered exchange coupling in the surface layer, and the postulate put forward earlier on the basis of experiment [1] that the surface is antiferromagnetically coupled with the bulk. In this paper, we provide evidence that the surface is <u>not</u> antiferromagnetically coupled with the bulk but rather an imperfect ferromagnetic coupling exists between the surface and the bulk as a result of a sizable perpendicular polarization of the surface layer, consistent with earlier work [3, 6, 7]. This effect undoubtedly results in phenomena consistent with the earlier experiment [1] but challenges the simpler models of perfect antiferromagnetic [1, 5] or perfect ferromagnetic [8] coupling between the surface and the bulk.

Our recent photoemission studies [9, 10, 11] of the Gd(0001) band structure have demonstrated the existence of a surface state at $\overline{\Gamma}$ consistent with the theoretical band structure[5]. The surface state is well localized at the surface[11, 12], and is therefore a unique probe of the surface magnetic order without requiring the complex task of separating surface and bulk signals. In addition, the temperature dependence of both the parallel

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and perpendicular components can be measured explicitly.

EXPERIMENTAL

The photoemission experiments were undertaken at the U5 beamline at the National Synchrotron Light Source (NSLS). The spectra were taken at low photon energies (typically 43 eV in this work) using a low energy spin polarimeter (described in detail elsewhere[13, 14]) attached to a commercial angle-resolved hemispherical analyzer. The light polarization of the incident light was perpendicular to the applied field and a mixture of s- and p-polarized light (55° incidence angle). All photoelectrons were collected normal to the surface since the Gd surface state is centered at $\bar{\Gamma}$ [5, 9, 10, 11, 12]. The combined energy resolution of the spectra was 300 meV. Spin polarized secondary electron emission experiments were conducted in a separate UHV system[15] equipped with two medium-energy retarding field Mott polarimeters capable of measuring all three components of the spin polarization vector. The secondary electrons were collected around normal emission with the sample negatively biased at -30 V. All spin polarization measurements were performed on remanantly magnetized samples.

The gadolinium films were evaporated from a tungsten basket, or crucible, at a rate of 0.5 – 2 Å/min. onto single crystal W(110), following well established procedures that are known to give coherent ordered clean gadolinium films[11, 16]. Following the deposition onto the room temperature substrate to allow layer-by-layer growth, the films were annealed at 450-550°C to reduce the number of defects [17, 18]. The films prepared in this way are flat and yield better remanent magnetization, in contrast to the "island" films grown at the elevated temperature of 450°C. The thickness of the films used in this work was typically 200 – 500 Å.

RESULTS AND DISCUSSION

As seen in Figure 1, the 5d surface state near the Fermi energy is highly spin-polarized as is the photoemission background and the Gd 4f levels for Gd(0001) films at 100K (the bulk Curie temperature is 293 K). The polarization for these films is 33 to 52% in the plane of the film. The polarization of both the surface state near E_F which contains only the surface contribution and the 4f photoemission feature with strong contributions from the gadolinium bulk are nearly constant at around 33%. The spin polarization of the 4f feature does not change much (±10%) with photon energy of 43 – 100 eV, while the surface sensitivity changes because the electron mean free path changes with electron kinetic energy. These facts exclude any possibility that the surface is antiferromagnetically coupled to the bulk. These results are consistent with the previously reported results from spin polarized photoemission of the shallow 4f gadolinium core levels and the spin-polarized secondary electrons[3, 7].

The "in plane" 4f spin-polarized spectra for clean, well ordered gadolinium films have a maximum for the spin minority peak at a binding energy 300 to 400 meV greater than that of the spin majority peak for gadolinium films at 100K (as seen in Fig. 1 & 2). This binding energy difference diminishes with the adsorption of a very small amount of contamination as seen in Fig. 2. Continued adsorption of contamination leads to a decrease in the net polarization and a diminution of the surface state intensity.

It has been well established that there exists a surface to bulk core level shift for gadolinium[19, 20]. For the 4f photoemission feature the surface component has a binding

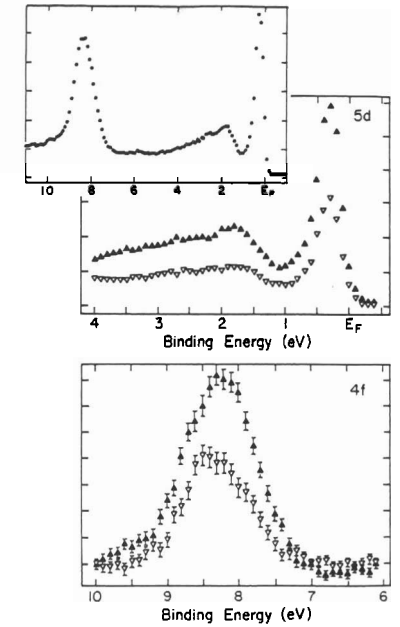

Fig. 1. The spin polarized photoemission spectra for a 300Åthick gadolinium film on W(110) at 100K. The spin majority signal (△) and spin minority signal (▽) were measured in the plane of the film. The photon energy is 43 eV and the photoelectrons were collected normal to the film so the surface state near E_F (200 meV binding energy) can be distinguished from the bulk Gd bands (1 – 4 eV binding energy). The insert shows the spin integrated Gd photoemission spectrum.

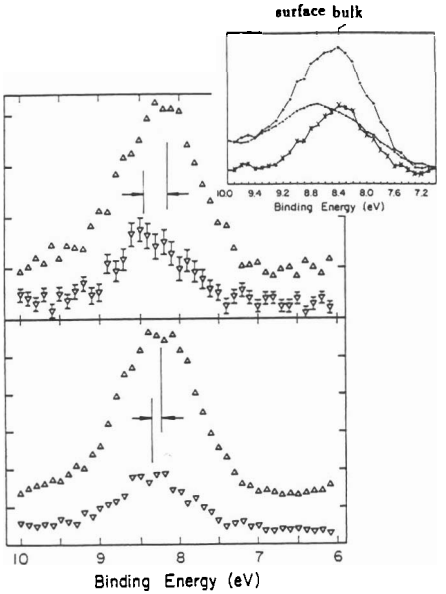

Fig. 2. The in-plane spin polarized photoemission spectra for a 300Åthick gadolinium film on W(110) at 100K across the Gd 4f levels. At top are shown the spectra of the freshly deposited film and below following exposure to a small amount of contamination. Spin majority (△) and spin minority (▽) are indicated, the photon energy is 43 eV. The insert shows the different Gd 4f photoemission spectra for an one monolayer and a ten monolayer film illuminating the different surface (one monolayer, •–•) and bulk (the difference spectrum, × – ×) contributions to the Gd film (this time deposited on Cu(100)). Data for the insert is described in Ref. 16 and taken with a photon energy of 40 eV.

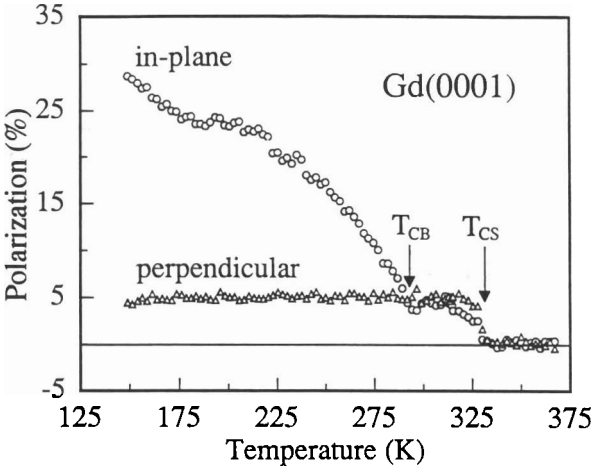

Figure 3: Temperature dependence of the spin polarization of 2 eV secondary electrons from a 400 ÅGd(0001) film grown at 300K annealed to 825K for 3.5 minutes. T_{cs} and T_{cb} indicate the surface and bulk magnetic ordering temperatures, respectively.

energy at least 0.37±0.02 eV greater than the bulk[19]. The different surface layer and bulk contributions to the Gd 4f photoemission signal can be easily seen from the difference spectrum between the spectrum obtained for a 1 monolayer films and the spectra obtained for thicker gadolinium films as seen in insert to Fig. 2. As can be seen from the insert to Fig. 2, the intensity and binding energy of the surface (1 monolayer film) contribution to the 4f levels closely resembles the intensity and peak position of the spin minority contribution to the spin resolved photoemission spectra for gadolinium films at 100K (T/T_c=0.34). While the level of polarization (>30%) and the spin majority polarization of the surface state excludes antiferromagnetic coupling between the surface and the bulk as noted above, this apparently slightly greater binding energy of the in-plane spin minority gadolinium 4f peak indicates that, for the films that are free of contaminations, the surface is slightly less polarized in the plane of the film when compared to the bulk. The Gd 4f moment may be safely assumed to be the same for the surface and the bulk as they are large localized moments [5]. Our results, therefore, suggest that there exists a surface magnetic moment component normal to the surface.

The existence of a perpendicular component in the surface magnetization is clearly shown in the temperature dependent secondary electron polarization data in Fig. 3. The secondary electrons are highly polarized due to significant contributions from the 4f core electrons[3, 7], and clearly show both perpendicular and in-plane polarization. Since the perpendicular component persists to a critical temperature well above ($\sim$ 40 K) the bulk Curie temperature (T_{CB}), it can only be identified with the surface polarization, as is the non-vanishing in-plane component above T_{CB}. The complete insensitivity of the perpendicular magnetization to the bulk magnetic transition suggests that the Gd(0001) surface behaves as an independent magnetic entity. We further note that, as best revealed in the perpendicular component, the surface magnetic transition is extremely abrupt, reminiscent of a two-dimensional system or possibly even a first order phase transition[21]. It is important to stress, however, that in the present experiment the sample was left in its remanent state throughout the entire temperature sweep, whereas in the previous

SPLEED[1] experiment the sample was remagnetized at each temperature step. Therefore, the present data contains, in the sign of the polarization, the added information on the relative directions of the in-plane polarizations above and below T_{CB}. The lack of a sign reversal on going through T_{CB} shows that the *in-plane* surface and bulk moments are ferromagnetically coupled.

The presence of a normal component of the surface magnetization are substantially altered by contamination. Very small amounts of contamination result in a reduction of the out of plane component of magnetization in the surface. As seen in Fig. 2, the binding energies for the spin minority and spin majority contributions to the Gd 4f signal differ by 350±50 meV for a clean Gd film but differ by less than 100 meV following the adsorption of contamination. We noted that the surface state still exists with the level of contamination that results in near perfect ferromagnetic ordering. So this perpendicular component of the magnetization at the surface is more sensitive to surface conditions than the surface state.

CONCLUSION

With spin-polarized spectroscopies, we have shown that the 5d surface state near E_f is a magnetic surface state. The surface magnetization of clean Gd(0001) is canted out of the plane, with its in-plane component ferromagnetically coupled with the bulk magnetization. The existence of a perpendicular component in the surface magnetization indicates an altered surface-bulk coupling and possibly a strong surface anisotropy.

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