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LETTER TO THE EDITOR

Evidence for imperfect ferromagnetic coupling between the Gd(0001) surface and the bulk

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Abstract. We have investigated ordered Gd(0001) films deposited on W(110) by spin-polarized photoemission at temperatures well below the Curie temperature. The gadolinium valence band including the 5d surface state, the 5d bulk, 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 provide strong evidence that the surface 4f polarization differs from the bulk 4f polarization for Gd(0001). We attribute the differences between surface and bulk polarizations to a component of magnetization normal to the surface in the surface magnetization which is not present in the bulk. Dilute amounts of contamination at the surface result in a more perfect ferromagnetic coupling between the surface and the bulk.

The magnetic coupling of the surface and bulk is an issue that has attracted considerable attention. Antiferromagnetic coupling on Gd(0001) has been predicted by recent calculations [1]. Nonetheless, the surface is seen to be able to maintain an in-plane remanent magnetization [2–5] in many thin gadolinium film studies, and there is little evidence for antiferromagnetic coupling [4]. Recent spin-polarized photoemission data of 4f levels demonstrate the presence of a sizeable perpendicular polarization for the surface layer indicating that perfect ferromagnetic coupling between surface and bulk does not exist [4].

Our recent photoemission studies [6–8] of the Gd(0001) band structure have demonstrated the existence of a surface state at $\bar{\Gamma}$ consistent with the theoretical band structure [1]. The surface state is well localized at the surface [8, 9], and is therefore a unique probe of the surface magnetic order of gadolinium without the complexities of separating signals from the surface and the bulk. In this paper we show that the surface is ferromagnetically coupled with the bulk from a comparison of the gadolinium 5d surface state with the other gadolinium photoemission features in spin-polarized photoemission. The spectra also show that the surface moments cannot be perfectly ferromagnetically coupled with the moments in the bulk, when free of contamination, consistent with the earlier results of Tang *et al* [4].

The experiments were undertaken at the U5 beamline at the National Synchrotron Light Source (NSLS). The spectra were taken at low photon energies (40–44 eV) using a gold-target spin polarimater (described in detail elsewhere [11, 12]) attached to a

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commercial angle-resolved hemispherical analyser. The spin-polarized photoemission spectra were obtained from remanently magnetized, thick (200–400 Å), gadolinium films deposited on W(110) substrate. The pulsing field (100–200 Oe) was applied to the films in the plane of the film before each scan and its direction was reversed each time to cancel out the possible instrumental asymmetry. The incident light from the undulator source was dispersed by a 6m toroidal grating monochromator and the photon energy for much of this work was 44 eV (an energy close to the second-harmonic output of the undulator). The 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 centred at $\bar{\Gamma}$ [1, 6–9]. The combined energy resolution of the spectra is 300 meV.

The gadolinium films were evaporated from a tungsten basket following well established procedures that are known to give well ordered clean gadolinium films [8, 13]. Following expansion the films were annealed at 450 °C to reduce the number of defects [14].

As seen in figure 1, the 5d surface state near the Fermi energy is highly spin-polarized as are the photoemission background and the Gd 4f levels for the film at 100 K (the bulk Curie temperature is 293 K). The polarization for these films is 33–52% in the plane of the film. The sign of the spin polarization can be reversed by reversing the direction of the magnetic field, which rules out the possibility of any non-magnetic artifact. The nearly constant polarization around 33% of both surface photoemission features (the surface state near E_F) and the 4f photoemission feature with a strong contribution from the gadolinium bulk excludes 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 [4, 15].

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 of 300–400 meV greater than that of the spin-majority peak for gadolinium films at 100 K (as seen in figures 1 and 2). This binding-energy difference diminishes with the adsorption of a very small amount of contamination as seen in figure 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 [16, 17]. For the 4f photoemission feature the surface component has a binding energy at least 0.37±0.02 eV greater than the bulk [16]. The different surface-layer and bulk contributions to the Gd 4f photoemission signal can easily be seen from the difference spectrum between the spectrum obtained for 1 ML films and the spectra obtained for thicker gadolinium films as seen in the inset to figure 2. As can be seen from the inset to figure 2, the intensity and binding energy of the surface (1 ML film) contribution to the 4f levels closely resemble the intensity and peak position of the spin-minority contribution to the spin-resolved photoemission spectra for gadolinium films at 100 K ($T/T_c = 0.34$). 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 the surface is slightly less polarized in the plane of the film when compared to the bulk for the films which are free of contaminations.

The 4f moment may be safely assumed to be the same for the surface and the bulk as they are large localized moments [1]. Contaminants tend to reduce the surface-state

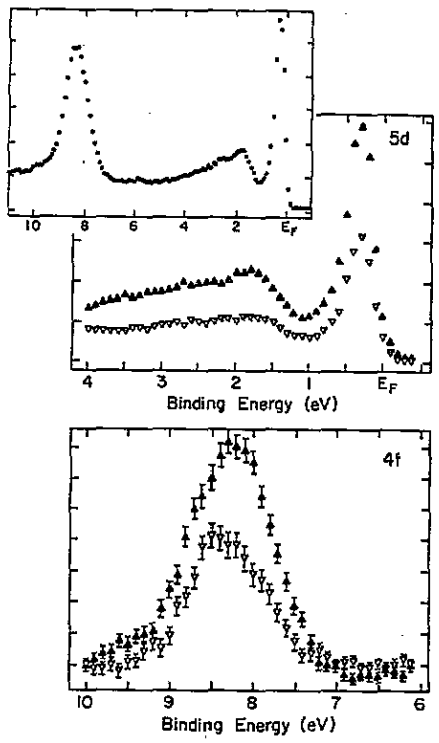

Figure 1. The spin-polarized photoemission spectra for a 300 Å thick gadolinium film on W(110) at 100 K. The spin-majority signal (△) and spin-minority signal (▽) were measured in the plane of the film. The photon energy is 44 eV and the photoelectrons were collected normal to the film so the surface 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.

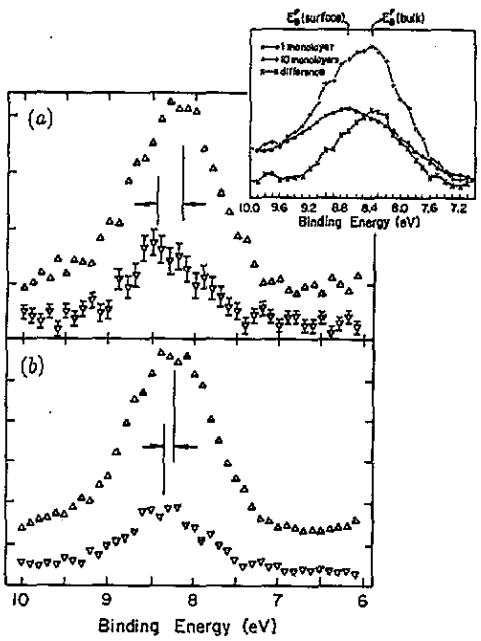

Figure 2. The in-plane spin-polarized photoemission spectra for a 300 Å thick gadolinium film on W(110) at 100 K across the Gd 4f levels. (*a*) the spectra of the freshly deposited film and (*b*) following exposure to a small amount of contamination. Spin-majority (△) and spin-minority (▽) are indicated, the photon energy is 44 eV. The inset shows the different Gd 4f photoemission spectra for 1 ML and a 10 ML film illuminating the different surface (1 ML) and bulk (the difference spectrum) contributions to the Gd film (this time deposited on Cu(100)). Data for the insert are described in [16] and taken with a photon energy of 40 eV.

intensity [7] and are believed to reduce the surface magnetization. It is unlikely given that the spin-minority component binding energy decreases that these small amounts of contamination would increase the surface 4f moments. Our results, therefore, suggest that there exists a surface magnetic-moment component normal to the surface.

These results are consistent with other evidence for an out-of-plane component to the magnetization. Spin-polarized photoemission investigations of the gadolinium 4f and secondary-electron background demonstrate that there exists a normal component of the magnetization [4]. Since this normal spin component to the magnetization was observed to have a Curie temperature greater than the bulk Curie temperature of gadolinium [4], it can be attributed to the surface alone.

These results indicating the presence of a normal component of the surface magnetization are substantially altered by contamination. Very small amounts of contamination $\ll 1\%$ of a monolayer result in a reduction of the out-of-plane component

of magnetization in the surface. As seen in figure 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 10 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 anisotropy of the magnetization at the surface is more sensitive to surface conditions than the surface state.

In conclusion, with spin-polarized photoemission, we have shown that the 5d surface state near E_F is a magnetic surface state. The magnetic moment at the Gd(0001) surface is mainly ferromagnetic coupled with those in the bulk. The 'in-plane' surface magnetization is consistent with the surface moments oriented with a tilt away from the surface plane, indicating a surface anisotropy. Since the surface state provides the magnetic information purely from the surface, it will be interesting to study the temperature dependence of its spin polarization to gain more understanding of this unique enhanced surface magnetic ordering, such as critical behaviour.

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