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## The influence of enhanced surface magnetism on finite-size scaling

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**Abstract.** – The thickness-dependent spin-polarized electronic structure of strained ultrathin and thin films of Gd has been investigated. The surface magnetic structure dominates the magnetic ordering of the ultrathin Gd films. With decreasing thickness some bulk bands exhibit increasingly more "passive" magnetic behavior. These bulk bands resemble a paramagnet over an increasing volume of the bulk Brillouin zone with decreasing film thickness.

Deviations from finite-size scaling of the Curie temperature could, in principle, result if surface magnetism has a crucial "active" role in the magnetic ordering of ultrathin films. This issue has been widely ignored in finite-size scaling experiments to date [1], [2], though recently deviations from finite-size scaling have been observed in the thin-film limit [3]. The finite-size scaling behavior in thin films of gadolinium has considerable experimental support [4]-[6]. Recent experimental evidence of magnetically "alive" monolayer and even submonolayer films [7]-[9] suggests that scaling behavior may however be quite complex. The deviations from expected scaling behavior have been attributed to interface-strain and surface-tension-induced anisotropies [10]-[12] but both band structure and the surface can play a role as well.

We studied the influence of the surface magnetic behavior on increasingly thinner films of strained Gd films. Expansive strain of 4% within the hexagonal closed-packed system substantially alters the electronic and magnetic valence band structure [13]. The magnetic structure of the system is strongly influenced by the intra-atomic  $4f$ -( $5d, 6s$ ) wave function overlap and  $5d, 6s$  electron itinerancy [14], [15]. This is manifested in a strong wave vector dependence [16] with an enhanced Curie temperature as compared to the unstrained Gd(0001) films grown on W(110) [4].

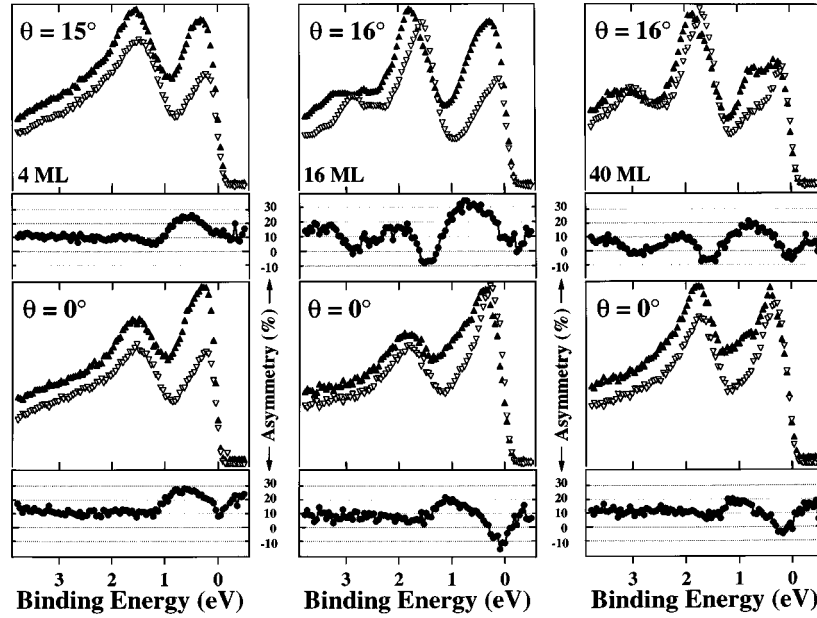


Fig. 1. – Spin-polarized emission angle-dependent valence band photoemission spectra for a 4 ML (left), a 12 ML (center) and a 40 ML (right) thick strained Gd film. The spectra have been acquired at the zone center (bottom) and near the high symmetry points  $\bar{M}$  and  $\bar{Y}''$  (top) of the hexagonal (12 and 40 ML) and rectangular (4 ML) surface Brillouin zones, respectively. Spin majority and spin minority components are indicated by ( $\blacktriangle$ ) and ( $\blacktriangledown$ ), respectively. The spin asymmetries are displayed directly underneath each set of spin-resolved spectra.

The temperature-dependent spin-resolved electronic structure of Gd films on Mo(112) was studied at the new U5UA undulator beamline at the National Synchrotron Light Source (NSLS) using a spin- and angle-resolved photoemission analyzer, as described in detail elsewhere [17], [18]. Ultrathin (3 monolayers  $< d < 10$  monolayers) and thin ( $d > 10$  monolayers) films of Gd were grown that resembled Gd(10 $\bar{1}2$ ) and Gd(0001), respectively, both with a 4% expansive strain [13]. The Gd films were magnetized in plane along the substrate corrugation lines and spin-polarized photoemission spectra were acquired in remanence. The surface and bulk character of the bands has been determined from chemisorption studies and photon energy dependence, while the symmetry of the bands has been ascertained from the light polarization dependence as described in detail elsewhere [13].

The thickness-dependent spin-resolved electronic structure of the valence band of strained Gd is illustrated in fig. 1, which shows three sets of valence band spectra that were acquired for film thicknesses of 4 monolayers (4 ML, left), 16 monolayers (16 ML, center) and 40 monolayers (40 ML, right), each at the surface Brillouin zone center  $\bar{\Gamma}$  (bottom) and near the zone edge  $\bar{Y}''$  (4 ML) and  $\bar{M}$  (16 ML and 40 ML) (top), respectively. The spectra are taken for a reduced temperature of  $T/T_c = 0.8$  (4 ML) and  $T/T_c = 0.5$  (16 ML and 40 ML) where the Curie temperature based on the bulk band structure is approximately 120 K, 290 K and 340 K, respectively. There are compelling differences in the spin-resolved spectra for the three film thicknesses.

The  $5d_{xz,yz}$  or  $5d_{x^2-y^2}$  bulk bands at 1.8 eV binding energy [13] of the 4 ML thick strained Gd film exhibit very little dispersion [13]. The lack of dispersion is associated with spin

majority and minority bands of very similar binding energy and a spin asymmetry which is comparable to the background polarization of approximately 10%. This is indicative of insignificant Stoner exchange splitting and minimal spin mixing behavior (four sub-bands that tend toward equal weight at the Curie temperature but with little shift in binding energy). The behavior of this bulk band is similar to that of a paramagnetic band that is polarized by the surface or the background [16].

This behavior in the ultrathin strained Gd films is in contrast to the 16 ML and 40 ML strained Gd films which are characterized by substantial dispersion of the  $5d_{xz,yz}$  or  $5d_{x^2-y^2}$  bulk bands [13], [16]. For the thicker films the bulk spin majority and minority bands are also of similar binding energy at 1.8 eV at  $\bar{\Gamma}$ . Approximately half-way through the zone along  $\bar{\Gamma}$   $\bar{\Sigma}$   $\bar{M}$  these bulk bands split into two branches and exhibit substantial exchange splitting of approximately 0.27 eV for  $T/T_C \approx 0.5$  at the zone edge  $\bar{M}$  with a characteristic Stoner-like exchange splitting collapse ( $\Delta E_{\text{ex}} \rightarrow 0$  as  $T \rightarrow T_C$ ). The wave-vector-dependent exchange splitting reflects a gradual change from predominantly paramagnetic behavior at the Brillouin zone center to Stoner-like magnetism near the zone edge for these bulk bands [16].

The spin-resolved valence band spectra of the thicker strained Gd(0001) films (fig. 1) reveal three features in the region near the Fermi level (0–1 eV binding energy), which is most clearly resolved for the 40 ML thick film. They have been assigned as i) a  $5d_{z^2-r^2}$ ,  $6s$  spin majority state with substantial surface character at approximately 0.75 eV below  $E_F$  for  $T/T_C \approx 0.5$ , ii) the corresponding spin minority state of surface character at approximately 0.2 eV below  $E_F$  for  $T/T_C \approx 0.5$  and iii) a spin majority state of bulk character at approximately 0.4 eV binding energy. The spin majority and minority bands of the surface-sensitive state exhibit little dispersion [16] and retain substantial exchange splitting throughout the studied thickness range at both high-symmetry points zone center and zone edge (fig. 1).

Our results demonstrate that some bulk bands have distinct thickness-dependent spin-resolved band structure. This thickness dependence of the band structure is as significant for the surface states. The dispersive  $5d_{xz,yz}$  or  $5d_{x^2-y^2}$  bulk bands at 1.8 eV below  $E_F$  at  $\bar{\Gamma}$  of the 40 ML thick Gd films are characterized by wave-vector-dependent exchange splittings. For films thinner than approximately 10 ML these bulk bands are localized with diminishing exchange splitting over a larger volume of the Brillouin zone. These bulk bands share the same polarization as the background for film thicknesses of less than 5 ML.

Despite the “complicated” spin-polarized band structure which is dependent on wave vector, band symmetry and electron localization (fig. 1), the thickness-dependent critical temperature decreases with decreasing film thickness. The thickness-dependent Curie temperature resembles the behavior of “unstrained” Gd grown on W(110) [6] (fig. 2), with the exception of the markedly increased surface  $T_C$  for films thicker than approximately 15 ML. The surface has a Curie temperature of  $370 \pm 25$  K for the strained Gd(0001) on Mo(112) compared to an enhanced Curie temperature of 310 to 315 K for unstrained Gd(0001) [4]. Strain has also increased the bulk Curie temperature from 293 K (unstrained) to  $340 \pm 20$  K.

Both strained and “unstrained” gadolinium films follow, at first glance, the expected scaling behavior [6]. Clearly the surface Curie temperature drops with decreasing film thickness, as expected from finite-size scaling theory. Considering the fact that both strained (Gd/Mo(112)) and unstrained (Gd/W(110)) films are dominated by enhanced surface magnetism (a surface with a Curie temperature higher than the bulk) for the thicker films and a distinct surface band structure one would expect surface contributions to affect the magnetic properties. Even more surprising is that the structural change from the strained Gd(0001) to the strained Gd(10 $\bar{1}$ 2) at a film thickness of approximately 10 ML also does not substantially affect the superficial finite-size scaling behavior measured from the surface (fig. 2), although significant changes in the electronic and magnetic structure of the bulk bands occur (fig. 1).

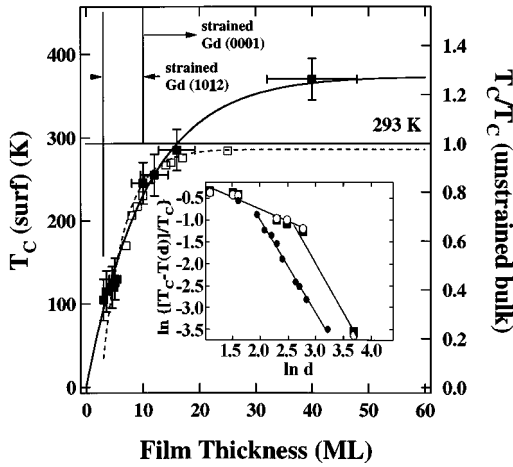


Fig. 2

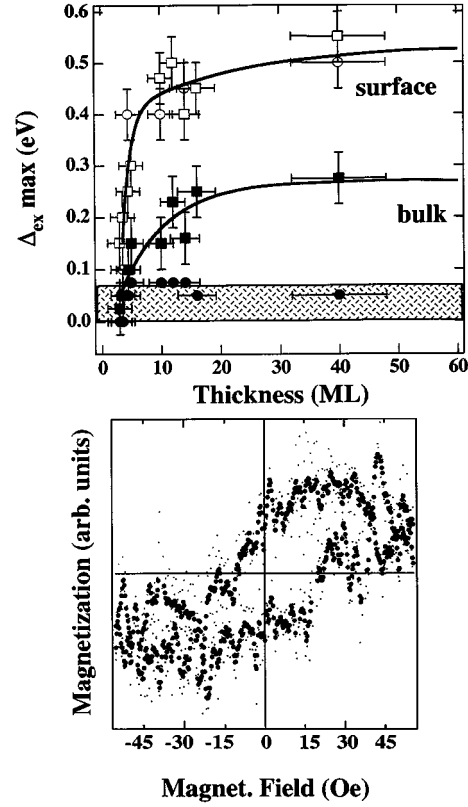


Fig. 3

Fig. 2. – The surface Curie temperatures for strained Gd films of various thicknesses (■) as determined from the temperature-dependent spin-resolved photoemission measurements. The right axis labels the measured surface Curie temperature of the strained Gd films relative to the unstrained bulk Curie temperature (293 K). The solid line shows the expected power law dependence of finite-size scaling (see text). The thickness-dependent critical temperatures of Gd(0001) (ref. [5], [6]) are indicated by the empty boxes and compared to a calculation (ref. [6]) following the finite-scaling theory (dashed line). The inset shows the measurements plotted on a log scale for both Gd(0001) on W(110) (●) and on Mo(112) (◻). Note that for the thicker films of Gd on both W(110) and Mo(112) the critical exponents are very similar.

Fig. 3. – Top: The maximum measured exchange splitting for the surface (open symbols) and bulk (filled symbols) as a function of film thickness. The data points were determined from spin-polarized photoemission spectra that were acquired near the zone edge (boxes) and at the zone center (circles). The exchange splitting plotted for the bulk bands at zone center is the upper bound. Bottom: The hysteresis loop of a ten-monolayer film at 130 K obtained using magneto-optic Kerr effect.

For films thicker than approximately 15 ML the experimentally determined critical exponent  $\beta$ , in

$$\frac{T_C - T_C(d)}{T_C} = C \cdot d^{-\beta} \quad (1)$$

compares to that of unstrained Gd [3] and the 3-dimensional Ising critical exponent of the correlation length. In the thinner films ( $d < 15$  ML) the critical exponent for strained Gd is

reduced by a factor of approximately 4 as seen in the insert to fig. 2. We do not have sufficient data to establish that as  $d \rightarrow 1$ ,  $\beta \rightarrow 0$ .

We have plotted the thickness-dependent maximum observed exchange splittings for the  $5d_{z^2-r^2}$ ,  $6s$  surface state and the  $5d_{xz,yz}$  or  $5d_{x^2-y^2}$  bulk bands at 1.8 eV below  $E_F$  (at  $\bar{\Gamma}$ ) for two wave vectors, corresponding to the zone center and zone edge (fig. 3a). For film thicknesses of more than 10 ML the bulk bands exhibit substantial exchange splittings at the zone edge of approximately 0.27 eV at  $T/T_C \approx 0.5$  (for a 40 ML thick film). With decreasing film thickness the maximum observed exchange splitting drops rapidly to 0.20 eV for the 16 ML film ( $T/T_C \approx 0.5$ ) and less than 0.05 eV for the 4 ML film ( $T/T_C \approx 0.8$ ) as illustrated in fig. 3a. The exchange splitting of the  $5d_{z^2-r^2}$ ,  $6s$  surface state at the zone edge (0.55 eV at  $T/T_C \approx 0.5$ ) is approximately twice that of the bulk which is indicative of an enhanced  $5d$  magnetic moment at the surface. The surface exchange splitting decreases with film thickness but less than the  $5d_{xz,yz}$  or  $5d_{x^2-y^2}$  bulk bands.

At the zone center the exchange splitting of the  $5d_{xz,yz}$  or  $5d_{x^2-y^2}$  bulk bands is negligible. The upper bound of this bulk band exchange splitting at  $\bar{\Gamma}$  is of the same order as the resolution of our experimental measurements (fig. 3a) and the possible exchange splittings are small. For the surface there is little difference between the exchange splitting at the zone center and at the zone edge. The thickness- and wave-vector-dependent exchange splitting suggests that with reduced film thickness increasingly more volume in  $k$ -space is occupied with weakly dispersive bulk bands that exhibit paramagnetic behavior. This results in a dominance of the surface magnetic ordering at the Brillouin zone center. This is supported by the hysteresis loop, shown in fig. 3b, of a 10 ML thick strained Gd film that exhibits a negative slope in the high-field region possibly indicating underlayer demagnetization effects [18].

An enhanced surface Curie temperature implies that there is an enhanced magnetic coupling at the surface as compared to the bulk ( $J_s > J_b$ ) [19]. Based on statistical models on semi-infinite systems, theory predicts an enhanced surface Curie temperature, when the coupling strength within the surface exceeds the critical value of  $J_s \geq 1.5 J_b$ . A different coupling at the surface as compared to the bulk is expected for strained Gd, based upon the distinct surface electronic structure. The magnetic correlation length of the surface is thus substantially shorter than that of the bulk. For increasingly thinner films, the film is considerably influenced by the surface, which has a higher  $T_C$  and can polarize the “paramagnetic” bulk [19], [20]. This polarization of an increasing volume of the film bulk by the surface should have a considerable effect on the critical exponent  $\beta$ , which changes with film thickness. Such changes in the film-thickness-dependent critical exponent have been seen before in Ni(111) films [3].

Our results extract crucial aspects of the magnetic ordering in ultrathin films that are based on the electronic structure and not typically considered in the finite size-scaling formalism. Using the analogy of M. Donath [21] we can say that for the ultrathin films of strained Gd some bulk bands are “spectators” and some are “actors” while the surface is consistently a very lively “actor”. The “actors” may play different roles at different points in the Brillouin zone. This work provides further support for the argument that electronic structure plays a key role in determining the Curie temperature of gadolinium, as has been suggested [22]. The statistical approach, typical of finite-size scaling models, fails to account for these important factors.

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