PHYSICAL REVIEW B VOLUME 51, NUMBER 15 15 APRIL 1995-I

Surface structure of epitaxial Gd(0001) films on W(110) studied by quantitative LEED analysis

J. Giergiel, A. W. Pang, and H. Hopster
Department of Physics and Institute for Surface and Interface Science, University of California, Irvine, California 92717

X. Guo and S. Y. Tong
Department of Physics and Laboratory for Surface Studies, University of Wisconsin–Milwaukee, Milwaukee, Wisconsin 53201

D. Weller
IBM Research Division, Almaden Research Center, San Jose, California 95120

(Received 20 October 1994)

The surface structure of thick (400 Å) Gd(0001) films, epitaxially grown on W(110), is investigated by low-energy electron-diffraction (LEED) *IV* measurements in combination with dynamical LEED calculations. A first-layer contraction of 2.4% and a second-layer spacing expansion of 1% is found. These findings are in good agreement with literature values determined for the (0001) surface of bulk Gd crystals. No significant difference in the LEED *IV* data is found between films grown at room temperature and films grown at elevated temperatures.

INTRODUCTION

The atomic structure of surfaces is in general different from the terminated bulk structure. Surface reconstruction and relaxation phenomena have been studied on many surfaces.[1] Usually one finds the first interlayer spacing to be smaller (contraction) than the bulk lattice constant. In magnetic materials, the interplay between structure and magnetic properties is a topic of great current interest.[2] Most of the magnetic studies have been performed on 3*d* transition-metal surfaces, and comparatively little work has been done on rare-earth surfaces. The (0001) surfaces of the rare-earth metals Gd and Tb have been shown to exhibit unusual magnetic properties. It was first shown that Gd has a surface-enhanced critical temperature, thus maintaining magnetic order above the bulk Curie temperature.[3,4] Later, Tb was found to show a similar effect.[5] Spin-polarized 4*f* photoemission spectra on Gd(0001) indicated the presence of a magnetically reconstructed surface with possibly antiferromagnetic (AF) coupling of the top surface layer to the bulk.[4] First-principles electronic structure calculations for Gd(0001) in a slab geometry confirmed that an AF surface alignment may be energetically favored over the ferromagnetic alignment.[6] This calculation also predicted an expansion of 6.3% of the first interlayer spacing, in contrast to most metal surfaces, where a contraction is observed. The calculations find a spin-polarized conduction-band surface state of minority-spin character. The observation of a surface state in non-spin-resolved photoemission spectra[7] seems to support this picture. However, very recent spin-polarized photoemission results on Gd(0001) surfaces have shown large 4*f* polarizations at low temperature which clearly invalidate the antiferromagnetic surface ordering.[8,9] Also, the surface state was shown to be mainly of majority-spin character.[8] Spin-polarized secondary electron spectroscopy shows a perpendicular magnetization component, which persists above the bulk Curie temperature.[9]

Thus the magnetic properties of Gd(0001) are much more complicated than anticipated, and the interplay between surface structure and magnetism needs to be studied more carefully. Structure analysis of (0001) surfaces of bulk Gd single crystals by low-energy electron diffraction (LEED) has yielded a surface contraction.[10] The discrepancy between theoretical predictions and experimental results have recently been addressed by Bylander and Kleinman, and attributed to inherent errors in the local-density approximation.[11]

Most of the magnetic studies on Gd surfaces were done on epitaxially grown films because bulk rare-earth surfaces are notoriously difficult to clean. We have therefore studied the structure of Gd(0001) films grown on W(110) in the same system and under the same conditions that have been used previously to study the surface magnetic properties of these films.[9] It is well established that good quality Gd(0001) can be grown on W(110). However, at elevated temperatures the growth proceeds as three-dimensional islands (Stranski-Krastanov growth mode). Good quality films can also be grown at low temperatures (room temperature) followed by annealing. The low-temperature growth leads to layer-by-layer growth. However, the structural quality is poor as evidenced by diffuse LEED spots. Subsequent annealing restores good structural order with sharp LEED spots. It is believed that these surfaces are smoother than those of films grown at higher temperature (450 °C). In spin-polarized photoemission and secondary electron emission[9] experiments the low-temperature grown and postannealed films clearly show higher polarizations. We therefore concentrate our studies on the surface structure of films grown at low temperature. A comparison to films grown at elevated temperatures will also be made.

EXPERIMENT

The experiments were performed in the same UHV system in which the spin-polarized photoemission and polarized secondary electron studies have previously been performed.[9] For LEED *IV* spectra measurements, a change-coupled-device camera connected to a personal computer via a frame grabber board was added. This allows for the digitization and spot intensity integration of LEED patterns observed with a commercial reverse-view LEED system. The details of the system are described elsewhere.[12] All LEED data were taken with the electron beam at normal incidence (to within 0.1°). Gd films were deposited from a W crucible at a rate of 0.5–1 Å/s onto a clean W(110) surface held at room temperature. Final thicknesses were approximately 400 Å as determined by a calibrated quartz-crystal monitor. The pressure during deposition remained below 1×10^{-9} Torr (dominated by Gd vapor). Surface cleanliness was monitored by Auger electron spectroscopy, which showed no more than a few percent of a monolayer of oxygen as the main surface contaminant. The films were subsequently annealed for 3–5 min at 825 K, resulting in a sharp (1×1) sixfold LEED pattern. Quantitative LEED *IV* data for the (10), (11), (20), and (21) (and their symmetry equivalents) were taken on these films in the energy range 40–300 eV in 2-eV increments.

THEORY

The dynamical LEED calculations were done by the real and reciprocal space symmetrized versions of the Van Hove–Tong multiple-scattering LEED code.[13,14] The phase shifts of Gd were calculated using the spherical part of a self-consistent full potential generated by a spin-polarized linearized augmented plane-wave (APW) band-structure calculation.[15] Since Gd is a strong scatterer, up to 14 phase shifts had to be used in the LEED *IV* calculation to ensure proper convergence up to 300 eV. At normal incidence, the step-averaged C_{3v} symmetry of the hexagonal-close-packed (hcp) structure is utilized and only 19 inequivalent beams are needed for proper convergence. The effect of atomic vibrations was taken into account by using a Debye temperature of 176 K, and inelastic damping was included by an optimized constant imaginary potential of 4.0 eV. The real part of the inner potential was also optimized in the calculation.

DISCUSSION

The spacing in the bulk of Gd is kept at $d=2.89$ Å. The first (d_{12}) and second (d_{23}) interlayer spacings are varied to match the calculated *IV* spectra with experiment. We also compare the calculation with the data of Quinn *et al.*[10] on a bulk Gd single crystal. Three reliability factors (Zannazi and Jona,[16] Pendry,[17] and Van Hove, Tong, and Elconin[18] are used to determine the best structure. The comparison between experimental and calculated curves is shown in Figs. 1–3. The optimal structure and the corresponding results of R factors are listed in Tables I and II.

In Fig. 1 we show a comparison between our calcula-

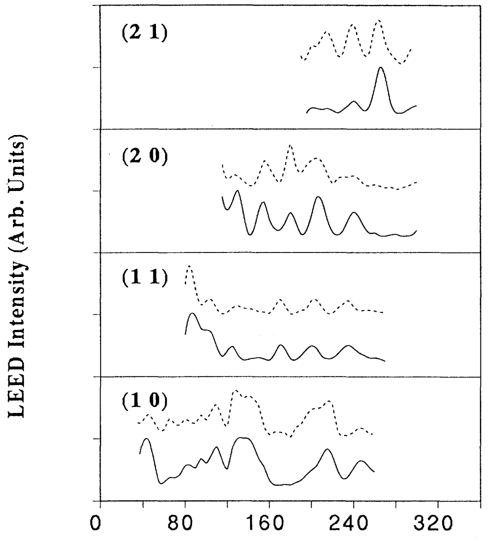

FIG. 1. Comparison between the experimental *IV* curves from Quinn *et al.* (Ref. 10) and our theoretical *IV* curves (dotted lines) on a bulk Gd single crystal.

TABLE I. Optimal structure and R-factor values for a bulk Gd single-crystal (0001) surface.

	This work			Results of Quinn *et al.*		
	R	Δd_{12} (Å)	Δd_{23} (Å)	R	Δd_{12} (Å)	Δd_{23}(Å)
R_{ZJ}	0.14	−0.12	+0.03	0.208	−0.115	+0.065
R_P	0.27	−0.01	+0.04	0.256	−0.085	+0.050
R_{VHT}	0.22	−0.12	+0.03	0.231	−0.105	+0.075

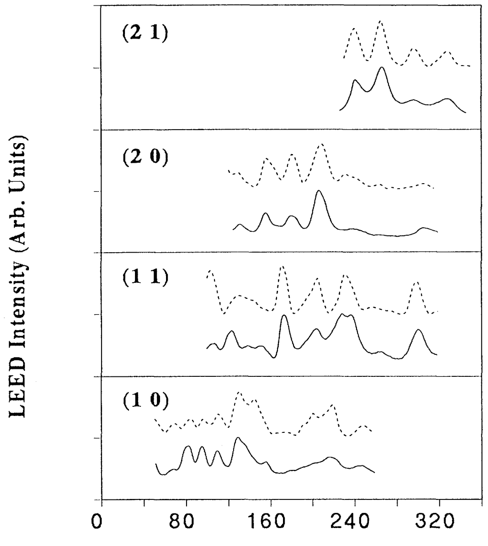

FIG. 2. The experimental (solid) and theoretical (dotted) *IV* curves for the high-temperature-grown film.

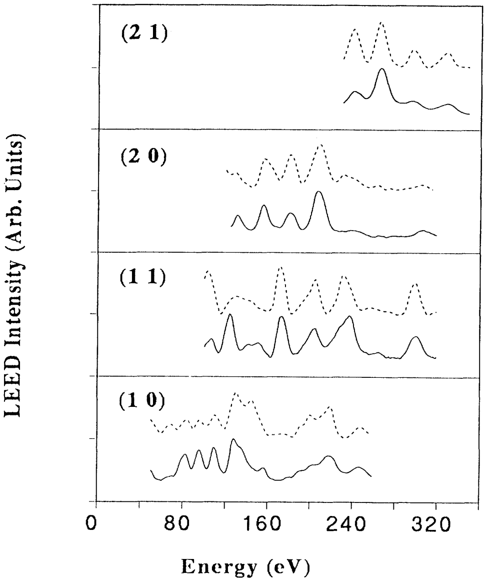

FIG. 3. The experimental (solid) and theoretical (dotted) *IV* curves for low-temperature-grown and postannealed films.

TABLE II. Optimal structure and R-factor values for 400-Å-thick Gd films grown at low temperature (300 K) and postannealed for 3 min at 875 K or grown at high temperature (825 K).

	Low temperature			High temperature		
	R	Δd_{12} (Å)	Δd_{23} (Å)	R	Δd_{12} (Å)	Δd_{23} (Å)
R_{ZJ}	0.20	−0.08	+0.03	0.20	−0.11	+0.02
R_P	0.34	−0.07	+0.03	0.37	−0.07	+0.03
R_{VHT}	0.23	−0.07	+0.03	0.27	−0.07	+0.03

tions and the measurements of Quinn *et al.*[10] on a bulk Gd single crystal. The R factors and the optimal structures thus determined are listed in Table I, together with the published results of Quinn *et al.*[10] The agreement between the two sets of structures is very good, especially in Δd_{12} (to within 0.015 Å). The scattering potential and other dynamical factors used, such as inelastic damping, inner potential, and surface vibration, are different in the two calculations. The biggest difference in the determined structures is that Δd_{23} is smaller in our result by 0.045 Å. This close agreement shows the consistency of LEED analyses, in this case better than 0.015 Å for Δd_{12} and 0.045 Å for Δd_{23}.

A comparison between measured and calculated *IV* spectra for Gd(0001) thin films, grown at high and low temperatures, respectively, is shown in Figs. 2 and 3. Our experimental data, after background correction, are quite similar to those of Quinn *et al.*,[10] especially in the position of the peaks and valleys. However, differences in peak shapes and relative intensities do exist. On the other hand, our measured *IV* spectra for low- and high-temperature growth show less of a difference, both in peak positions and relative intensities. The optimal structures and R factors obtained from these analyses are summarized in Table II. The optimized inner potential is 6 eV for both films. This value is smaller than that of the bulk crystal which is 9 eV. The R factors are also somewhat worse than those obtained in comparing with the data of Quinn *et al.*[10] This may be due to disorder in the film. By smoothing the experimental curves, we improve the Pendry R factor to better than 0.29, a number close to that in Table I. For reasons mentioned above, the low-temperature-grown films have better long-range order. This is confirmed by the better R-factor values shown in Table II for two of the three R factors. The most important result, however, is that the surface structure of the epitaxially grown films (at low or high temperature) and that of the bulk crystal are quite close. Averaging the structures over the different R factors for the low-temperature film, we obtain a best structure of $\Delta d_{12} = -0.07 \pm 0.01$ Å and $\Delta d_{23} = 0.03$ Å, i.e., a contraction of 2.4% for the first interlayer spacing and an expansion of 1% for the second interlayer spacing, compared to $\Delta d_{12} = -0.12$ Å and $\Delta d_{23} = +0.03$ Å (4.2% and 1.0%, respectively) for the bulk crystal surface. The differences in the two sets of numbers are certainly within the error bars of the LEED method.

In conclusion, we have determined by quantitative *IV* LEED analysis the interlayer spacings at the surface of epitaxial Gd(0001) films grown on W(110). A first-interlayer contraction of 2.4% is found, while a slight expansion of the second-layer spacing is deduced. These values are in good agreement with results of a previous *IV* LEED study on bulk Gd(0001) surfaces. Thus the surfaces of the epitaxial films correspond to bulk surfaces, and the magnetic surface properties should be the same for the two systems.

ACKNOWLEDGMENTS

The work of University of Wisconsin–Milwaukee is supported in part by NSF Grant No. DMR 9214054 and by DOE Grant No. DE-FG02-84ER45076. The work at the University of California, Irvine, is supported by NSF Grant No. DMR 9119815.

---

[1] See, for example, *The Structure of Surfaces III*, edited by S. Y. Tong, M. A. Van Hove, K. Takayanagi, and X.-D. Xie (Springer-Verlag, Berlin, 1991).

[2] L. M. Falicov, D. T. Pierce, S. D. Bader, R. Gransky, K. B. Hathaway, H. Hopster, D. N. Lambeth, S. S. P. Parkin, G. Prinz, M. Salamon, I. K. Schuller, and R. H. Victora, J. Mater. Res. **5**, 1299 (1990).

[3] C. Rau and S. Eichner, Phys. Rev. B **34**, 6347 (1986).

[4] D. Weller, S. F. Alvarado, W. Gudat, K. Schröder, and M. Campagna, Phys. Rev. Lett. **54**, 1555 (1985).

[5] C. Rau, C. Jin, and M. Robert, J. Appl. Phys. **63**, 3667 (1988).

[6] R. Wu, C. Li, A. J. Freeman, and C. L. Fu, Phys. Rev. B **44**,

9400 (1991).

[7] D. Li, C. W. Hutchins, P. A. Dowben, C. Hwang, R. T. Wu, M. Onellion, A. B. Andrews, and J. L. Erskine, J. Magn. Magn. Mater. **99**, 85 (1991).

[8] G. A. Mulhollan, K. Garrison, and J. L. Erskine, Phys. Rev. Lett. **69**, 3240 (1992).

[9] H. Tang, D. Weller, T. G. Walker, J. C. Scott, C. Chappert, H. Hopster, A. W. Pang, D. S. Dessau, and D. P. Pappas, Phys. Rev. Lett. **71**, 444 (1993).

[10] J. Quinn, Y. S. Li, F. Jona, and D. Fort, Phys. Rev. B **46**, 9694 (1992).

[11] D. M. Bylander and L. Kleinman, Phys. Rev. B **50**, 4996 (1994).

[12] J. Giergiel, H. Hopster, J. M. Lawrence, J. C. Hemminger, and J. Kirschner, Phys. Sci. Instrum. (to be published).

[13] M. A. Van Hove and S. Y. Tong, *Surface Crystallography by LEED* (Springer-Verlag, Berlin, 1979).

[14] S. Y. Tong, H. Huang, C. M. Wei, W. E. Packard, F. K. Men, G. Glander, and M. B. Webb, J. Vac. Sci. Technol. A **6**, 615 (1988).

[15] E. Wimmer, H. Krakauer, M. Weinert, and A. J. Freeman, Phys. Rev. B **24**, 864 (1981); H. J. Hansen and A. J. Freeman, *ibid.* **30**, 561 (1984).

[16] E. Zanazzi and F. Jona, Surf. Sci. **62**, 61 (1977).

[17] J. B. Pendry, J. Phys. C **13**, 937 (1980).

[18] M. A. Van Hove, S. Y. Tong, and M. H. Elconin, Surf. Sci. **64**, 85 (1977).