Ferromagnetic Order and the Critical Exponent γ for a Gd Monolayer: An Electron-Spin-Resonance Study

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A monolayer of Gd(0001) on W(110) is measured by electron-spin resonance from $T=240$ to 360 K in UHV. The ferromagnetic Curie temperature lies ≈ 20 K below the bulk Curie point. The measured temperature dependence of the static susceptibility—deduced from the ESR intensity—follows a power law $\chi \sim t^{-\gamma}$ with $\gamma \approx 1.8$ for a monolayer and $\gamma \approx 1.25$ for an 80-Å film. This agrees well with the theoretical γ of 2D and 3D Ising systems. The experiment represents the first *in situ* UHV ESR study with full surface analysis allowing the measurement of magnetic phase transitions on single-crystal surfaces.

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The critical behavior of the paramagnetic susceptibility and the magnetization of thin surface layers has been the subject of many investigations.[1] Gadolinium is a prominent candidate for such studies.[2-5] Many questions concerning the magnetic behavior of a thin Gd film are still open to be answered: Does a Gd monolayer (ML) show a ferromagnetic phase transition at all? If it does, will the transition temperature T_{Cs} be shifted to higher or lower temperatures? Is it possible to detect finite-size effects in thin Gd films, i.e., is there a thickness-dependent shift of T_{Cs}? What is the difference between a magnetic monolayer [i.e., Gd(0001) on W(110)] and a semi-infinite system (i.e., the surface layer of Gd on Gd metal)? Answers to these questions contribute to the theoretical understanding of the local-moment spin-spin coupling and of the influence of the local-moment–conduction-electron interaction. Since the ferromagnetic properties of rare-earth–transition-metal compounds are essentially determined by those interactions, the answers to the above questions have direct applications to the "engineering" of thin-film ferromagnets.

First studies on polycrystalline Gd films prepared in ultrahigh vacuum (UHV) by Rau[2] (using electron-capture spectroscopy) and by Cerri, Mauri, and Landolt[3] (using spin-resolved photoemission) showed a deviation of the film magnetization from the bulk one and a shift of the surface-layer ordering temperature to higher values than the bulk Curie temperature $T_{Cb}=292.5$ K. The magnetic ordering of the topmost layear of a 140-Å epitaxial Gd(0001) film on W(110) 22 K above T_{Cb} was reported by Campagna and co-workers.[4a] They measured in zero applied field using spin-polarized low-energy electron diffraction (SPLEED) and spin-resolved photoemission. A Gd monolayer on Fe(100) was investigated by Taborelli *et al.*[5] using spin-polarized Auger-electron spectroscopy (SPAES).

In the present work we will demonstrate the usefulness of UHV ESR in the field of surface magnetism. As has been shown,[6,7] the high sensitivity of ESR (10^{12} spins) allows us to detect a fraction of a monolayer Gd on a metal surface in the paramagnetic regime. The ferromagnetic resonance (FMR) which was also recorded here (below T_{Cs}) will be discussed elsewhere. While the FMR of ultrathin magnetic layers (Fe, Ni, CO) has been reported previously,[8-10] the present work is the first ESR measurement of a well-characterized magnetic monolayer far above the transition temperature. At present, UHV ESR seems to be the only technique able to collect magnetic data above T_{Cs} for a clean magnetic ML. Even the torque measurements of the surface magnetization of Gradmann[11] are restricted to the magnetically ordered state.

It is known[4b,12] that a Gd(0001) ML grows epitaxially on W(110), provided that extreme care is taken to clean the substrate and the evaporant. The growth modus is controlled by Auger-electron spectroscopy (AES) and LEED. Figure 1(a) shows the linear increase of the Gd(138/140 eV) peak amplitude till the monolayer (defined as an adsorbate coverage $\theta_A=1.0$) is complete after an evaporation time of 16 min. Longer evaporation times lead to a Stranski-Krastanov–type growth modus (formation of Gd islands on one or two epitaxial layers) in agreement with Ref. 12. We chose deliberately a coverage of $\theta_A=0.8$ (i.e., 80% of a monolayer) to have at most one monolayer or less on the surfce (approximately 40 mm^2). A sample with $\theta_A=1.6$ and a 80-Å thick film were also measured. For the latter thickness it is assumed that the film has formed a smooth surface again, and a LEED structure is detected.[4b] After preparation and characterization of the epitaxial Gd film, the sample is moved *in situ* into a quartz finger of the UHV chamber.[6,7] Rotation of the sample allows ESR experiments with the Zeeman field **H** applied perpendicular and parallel to the surface plane.

Experimental ESR recorder traces at approximately 50 K above the ordering temperature are shown in Fig. 1(b). First, we see that the ESR is sensitive to $\frac{1}{10}$ of a

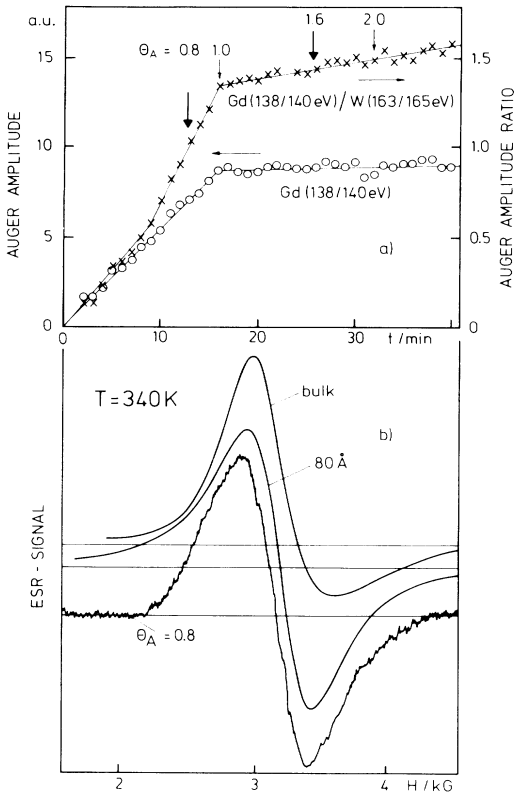

FIG. 1. (a) The Auger amplitude of Gd and the Auger amplitude ratio Gd/W as functions of evaporation time. An adsorbate coverage of $\theta_A=1$ corresponds to a hcp Gd(0001) close-packed monolayer on W(110). The substrate temperature during evaporation was $T_s=450\,^{\circ}$C. (b) ESR absorption spectra for a 18-μm (bulk), 80-Å, and $\theta_A=0.8$ Gd adsorbate layer, far in the paramagnetic regime. The microwave frequency is 9.30 GHz and H_0 lies in the surface plane. We detect a Dysonian profile (Ref. 13) for a layer thicker than the skin depth (few μm); whereas for $d\ll 1$ μm a symmetric Lorentzian line shape is recorded. The linewidths are given in Fig. 2.

monolayer and less: The signal-to-noise ratio of the 0.8-ML sample is ≈ 100. This coverage corresponds to 3.1×10^{14} Gd atoms (see Table I). Taking demagnetization effects into account a g factor, $g=1.97$, and a Korringa rate of $\Delta H/\Delta T\approx 5$ G/K are determined, in agreement with previous bulk ESR studies on Gd.[13] An analysis of the resonance shift, the linewidth, and the FMR will be published elsewhere. Here, we will focus on the two most important questions. (i) Does the monolayer Gd undergo a ferromagnetic phase transition and how does the surface Curie temperature T_{Cs} shift with respect to T_{Cb}? (ii) Does the experimental susceptibility χ_0 follow a power law,

$$\chi_0 \propto t^{-\gamma} \tag{1}$$

and what is the critical exponent γ for a 3D and a 2D Gd film?

TABLE I. The number of atoms is estimated from the sample volume. In particular for the bulk sample an error of a factor of 2 is possible. In the third column the same values normalized to 1 for $\theta_A=0.8$ are listed. In column 4 we list the maximal values of the ESR intensity (Fig. 2) of the four samples also normalized with respect to $\theta_A=0.8$. The ferromagnetic Curie temperature T_C is taken from the inflection points of the ESR intensity curves in Fig. 2 and is in agreement with the fit in Fig. 3. The critical exponent is determined from Fig. 3.

Layer	No. of atoms Absolute	Relative	χ_0^{max} (a.u.)	T_C (K)	γ
Bulk	6×10^{17}	1935	1240	292.5 ± 2	1.25 ± 0.2
80 Å	1.1×10^{16}	35	44	288 ± 2	1.25 ± 0.2
$\theta_A=1.6$	6.2×10^{14}	2	2.3	281 ± 2	1.74 ± 0.2
$\theta_A=0.8$	3.1×10^{14}	1	1	271 ± 2	1.90 ± 0.2

The static susceptibility is proportional to the area under the paramagnetic resonance curve[14]

$$\chi(0)=(g\mu_B/\pi h\nu)\int_0^\infty \chi''(H)\,dH. \tag{2}$$

An absolute calibration of the ESR intensity is rather difficult, but for the present question a relative comparison is satisfactory. In Fig. 2 this ESR intensity is plot-

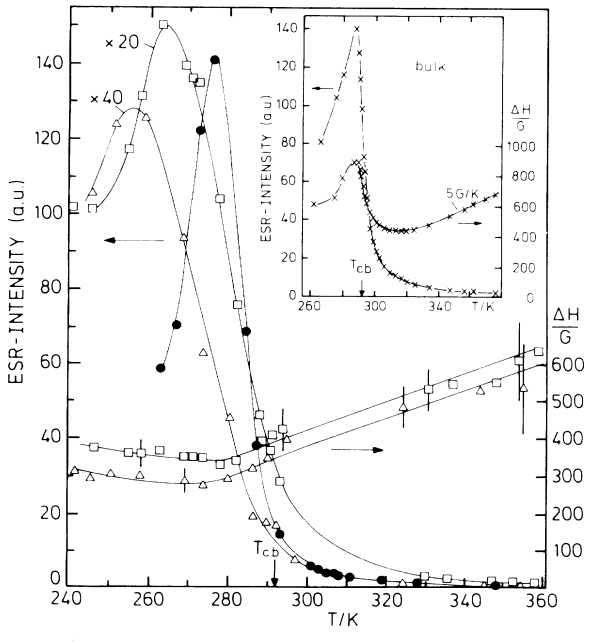

FIG. 2. ESR intensity and linewidth as functions of temperature for 80 Å (closed circles), $\theta_A=1.6$ (open squares), and $\theta_A=0.8$ (open triangles) of Gd/W(110). The gain factors for $\theta_A=0.8$ and $\theta_A=1.6$ are 40 and 20 with respect to the 80-Å data. For comparison the corresponding data of a bulk foil (crosses) are shown in the inset. Solid lines are guides to the eye; $T_{Cb}=292.5$ K.

ted as a function of T for all three layers. The inset shows the same diagram for an 18-μm bulk foil. As one can see from the inset $\chi(0)$ increases strongly in the vicinity of T_{Cb}. The inflection point of the experimental curve coincides with the known bulk ordering temperature $T_{Cb}=292.5$ K better than within one degree kelvin. The same type of analysis for the 80-Å (closed circles), the 1.6-ML (open squares), and the 0.8-ML data (open triangles) yields the layer ordering temperatures listed in Table I. Thus we find *strong evidence for a ferromagnetic ordering of the monolayer.* Furthermore, we notice a broadening of the transition for smaller coverages (open triangles). The consistency of our ESR intensity analysis can be seen by comparing the intensity maxima χ^{max} in Fig. 2 with the nominal number of spins for each sample (Table I). The corresponding relative values scale within 30% to 50% (Table I).

Now we turn to the second question. Do our experimental data for $T\rightarrow T_C^+$ follow Eq. (1)? What is γ determined from our experiment for the thick film and the 2D layer? In Fig. 3 we have plotted $\chi(0)$ on a log-log scale as a function of $t=(T-T_C)/T_C$. Taking T_C for each sample fixed and to be the inflection-point temperature or keeping it as a variable parameter in a least-squares fit yields the same result within ± 1 K(!). For the two 3D samples (crosses and closed circles) one gets $\gamma=1.25$ and for the two 2D layers $\gamma\approx 1.8$ (see Table I). This is in good agreement with theoretical predictions for

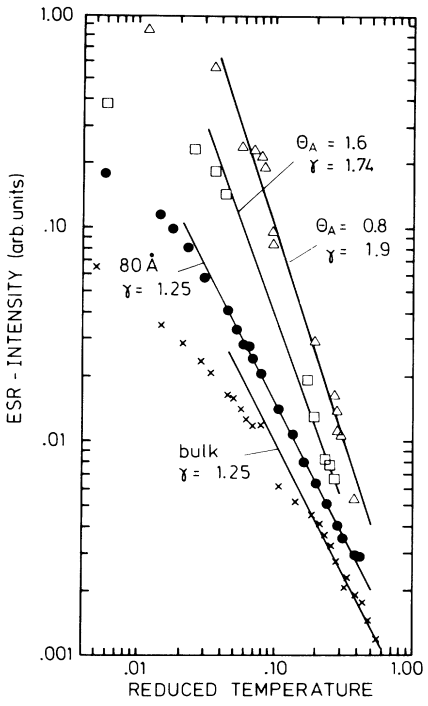

FIG. 3. Log-log plot of the ESR intensity [$\propto\chi(0)$] of Gd on W(110) for $T>T_{Cs}$. Same symbols as in Fig. 2. Straight lines are best fits by a power law $\chi(0)\propto t^{-\gamma}$.

3D and 2D Ising systems,[15] namely, 1.24 and $\frac{7}{4}$. It also agrees with the γ value of bulk Gd.[16] Consequently the "real" Gd monolayer cannot be seen as a 2D Heisenberg ferromagnet. Some anisotropic interaction—present in bulk Gd, too—turns it into an Ising-like behavior.[4a] The deviation from a power law below $t<0.03$ may be due to the applied field or a crossover behavior. The data for the bulk foil are of poor quality, because of its polycrystalline character. The error bars of the experimentally determined critical exponent (Table I) are large and chosen reasonably. However, the change from a 3D to a 2D system is unambiguous.

Concerning the ferromagnetic ordering temperature of Gd, the situation is as follows: Bulk Gd with nearest-neighbor coordination of $N=12$ orders at $T_{Cb}=292.5$ K.[4a,16] In a semi-infinite system the topmost Gd layer has $N=9$ and orders ≈ 22 K *higher.*[2-4a] A single monolayer with $N=6$ has a *lower* T_{Cs} of ≈ 271 K. At first glance one might expect that T_C scales with the coordination number. The present experiment suggests the importance of the conduction electrons for rare-earth magnets. The surface density of states for a Gd ML, the interplay of the Gd $5d$ levels with the partially filled $5d$ band of tungsten, and a mismatch between Gd bulk lattice constant and the W-W distance on a (110) surface have a competing influence on the Ruderman-Kittel-Kasuya-Yosida coupling.

We noticed that for the 80-Å samples (≈ 27 layers) the T_C is already reduced, but has the same γ within the experimental error bars. Finite-size effects do change the T_C, detectable in the experiment. A layer-dependent $T_C(l)$ should scale according to[17]

$$[T_C(l)-T_{Cs}]/T_C\propto b\times l^{-\lambda}. \quad (3)$$

For a 3D Ising layer system with free surfaces, Capehart and Fisher[18] estimate that $\lambda\approx 1.56$ and for a planar Ising square lattice the prefactor is negative, $b\approx -1$. For $l\approx 27$ layers, Eq. (3) yields a temperature reduction of approximately 2 K. Also for 30-Å-thick Gd films on Fe[5] a reduction of T_C was found, in agreement with our result. A comparison of correlation-length effects measured by SPAES[5] and ESR will be of interest. The former measures the ξ in the surface plane. The latter is not layer sensitive, it detects the averaged 3D ξ.[1]

In summary, we have demonstrated one further technique for the investigation of surface magnetism. It offers a submonolayer sensitivity and—at present—is the only method which probes the paramagnetic phase as well as the magnetic ordered state. Experiments at low temperatures far in the ferromagnetic phase are under current investigation. The result will give information on the spin-wave excitations of the 2D Gd ML and the orientation of its magnetization with respect to the surface normal.

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[1]K. Binder, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. Lebowitz (Academic, London, 1983), Vol. 8.

[2]C. Rau, J. Magn. Magn. Mater. **31–34**, 874 (1983).

[3]A. Cerri, D. Mauri, and M. Landolt, Phys. Rev. B **27**, 6526 (1983).

[4a]D. Weller, S. F. Alvarado, W. Gudat, K. Schröder, and M. Campagna, Phys. Rev. Lett. **54**, 1555 (1985); M. Campagna, J. Vac. Sci. Technol. A **3**, 1491 (1985); D. Weller and S. F. Alvarado, Z. Phys. B **58**, 261 (1985).

[4b]D. Weller and S. F. Alvarado, J. Appl. Phys. **59**, 2908 (1986); D. Weller, Ph.D. thesis, University of Cologne, 1985 (unpublished).

[5]M. Taborelli, R. Allenspach, G. Boffa, and M. Landolt, Phys. Rev. Lett. **56**, 2869 (1986).

[6]M. Farle, M. Zomack, and K. Baberschke, Surf. Sci. **160**, 205 (1985).

[7]M. Zomack and K. Baberschke, in Proceedings of the Eighth European Conference on Surface Science (ECOSS-8) [Surf. Sci. (to be published)].

[8]G. A. Prinz, G. T. Rado, and J. J. Krebs, J. Appl. Phys. **53**, 2087 (1982).

[9]W. Göpel, Surf. Sci. **85**, 400 (1979).

[10]M. Pomerantz, J. C. Slonczewski, and E. Spiller, J. Magn. Magn. Mater. **54–57**, 781 (1986).

[11]W. Gradmann, J. Magn. Magn. Mater. **54–57**, 733 (1986).

[12]J. Kolaczkiewicz and E. Bauer, Surf. Sci. **175**, 487 (1986).

[13]R. H. Taylor, Adv. Phys. **24**, 681 (1975).

[14]R. M. White, *Quantum Theory of Magnetism* (Springer-Verlag, New York, 1983), p. 97.

[15]M. E. Fisher, Rep. Prog. Phys. **30**, 615 (1967); H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Clarendon, Oxford, 1971); J. H. Chen, M. E. Fisher, and B. G. Nickel, Phys. Rev. Lett. **48**, 630 (1982).

[16]M. N. Deschizeaux and G. Develey, J. Phys. (Paris) **32**, 319 (1971); C. D. Graham, Jr., J. Appl. Phys. **36**, 1135 (1965).

[17]M. N. Barber, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. Lebowitz (Academic, London, 1983), Vol. 8.

[18]T. W. Capehart and M. E. Fisher, Phys. Rev. B **13**, 5021 (1976).