Critical Behavior of the Surface Magnetization of an Isotropic Heisenberg Ferromagnet: EuS(111) on Si(111)

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Magneto-optic Kerr-effect measurements (bulk sensitive) and spin-polarized low-energy electron diffraction (surface sensitive) reveal that the surface and the bulk Curie temperatures of epitaxial EuS(111) on Si(111) are identical to within ±0.04 K. In the critical region of the bulk, the surface magnetization m_1 of this model substance for an isotropic Heisenberg ferromagnet decreases as $m_1 \sim (1-T/T_C)^{\beta_1}$, with $\beta_1 = 0.72 \pm 0.03$. This differs significantly from current theoretical estimates of β_1, possibly pointing to a very strong surface anisotropy.

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The interest in critical phenomena near surfaces and in thin films, i.e., the role of interface effects on phase transitions, has significantly increased recently because of theoretical advances[1] as well as the advent of novel experimental methods.[2] Comparing with the success of the activities dealing with solids in the sixties and seventies,[3] one realizes that experimental progress with interfaces has by far not reached comparable magnitude. Even fundamental questions like melting near clean surfaces of real solids like Pb(100) are a current matter of controversy.[4]

Magnetic solids have played a central role in elucidating the physics of continuous phase transitions both in three and in two dimensions. Because of the technological progress in *in situ* preparation [primarily molecular-beam epitaxy (MBE)] and characterization, the possibility of also preparing well defined surfaces and ultrathin films as well as novel metastable structures and superlattices of magnetic solids is becoming reality.[5] When this progress is coupled to the simultaneous use of surface- and bulk-sensitive magnetic probes, novel possibilities open up in the field of surface magnetism. Among such probes we have successfully used *in situ* the magneto-optic Kerr effect and spin-polarized low-energy electron diffraction (SPLEED) to determine the bulk and surface magnetic behavior of the prototype itinerant-electron system[6] Ni and of the metallic, localized-magnetic-moment system Gd.[7] The (0001) Gd surface shows, surprisingly, magnetic reconstruction and enhanced surface Curie temperature T_{Cs} (up to 22 K) with respect to the bulk $T_{Cb} = 293$K. The question concerning the "ordinary" surface magnetic behavior of a model substance for an isotropic Heisenberg ferromagnet was therefore of special interest. The aim of this paper is to report the first experimental study of the surface magnetic order near the critical point of EuS(111). Our results strongly suggest an *ordinary* surface transition[1] for this system.

The experimental setup used is similar to the one described previously.[6,7] We have, however, used a novel, specially designed UHV-compatible cryostat,[8] allowing electron scattering and optical experiments to be performed *in situ* in the range 6–1060 K. In the range 6 K $< T <$ 20 K the temperature of the sample can be stabilized to within 10 mK with the use of a microcomputer. EuS(111) films with thicknesses of about 500 Å were grown *in situ* by MBE on a clean "buffer" layer of EuS(111). This buffer layer was also prepared by MBE on a clean Si(111) surface in a separate, dedicated MBE apparatus.[9] The growth rate of the EuS film was about 1 Å/s and the substrate temperature about 1020 K. Such growth parameters are known to yield nearly ideal stochiometry.[10] Annealing at 1020 K for one hour results in sharp LEED reflexes, indicating an ordered surface. Auger-electron spectroscopy indicates a topmost Eu-rich (111) plane of the surface but no surface contamination to within the spectrometer sensitivity. Subsequent Rutherford backscattering studies showed that the crystal perfection and the stochiometry of our EuS(111) samples are comparable to those of typical films grown by Saftic *et al.*[9,11] Results of the preparation and detailed characterization by ion scattering of EuS epitaxial films will be presented elsewhere.[12]

SPLEED probes the magnetization of the surface region to a depth of a few atomic layers. This magnetization generates the so-called magnetic exchange-scattering asymmetry[6]

$$A_{\mathrm{ex}}(E) = (I_{\uparrow\uparrow} - I_{\uparrow\downarrow})/(I_{\uparrow\uparrow} + I_{\uparrow\downarrow}),$$

where $I_{\uparrow\uparrow}$ ($I_{\uparrow\downarrow}$) is the scattered intensity for parallel (antiparallel) orientation of the incident electrons with respect to the direction of the $4f^7$ spins of the EuS layer. E is the kinetic energy of the incident polarized electrons from a GaAlAs photocathode. The $A_{ex}(E)$ spectra of the 00 beam collected at a scattering angle $\theta=24°$ (with respect to the crystal normal) and at a temperature $T=7$ K in the range 20 eV $< E <$ 200 eV are characterized by asymmetry values in the range of a few percent with two flat peaks (~ 10 eV FWHM) centered at $E=128$ and 170 eV, of asymmetry 4% and 3% respectively.[12] Feder and Pleyer[13] discussed—within the frame of dynamical LEED theory—the temperature range in which the relation $A_{ex} \sim m_1(T)$ is expected to be valid, with $m_1(T)$ being the long-range magnetic-order parameter of the region probed by SPLEED. They suggest that the linear relationship holds near the critical temperature T_C. The results of SPLEED experiments on Ni single-crystal surfaces showed that the relationship is indeed valid near T_C, the main reason being that in the critical region $\xi_\perp$, the magnetic coherence length perpendicular to the ferromagnetic surface, is much larger than the depth probed by SPLEED.[14] Here we note that this linear relationship could be affected by a change of the electron affinity observed in highly doped samples[15] which might induce an energy shift of the SPLEED spectral features. For EuS this shift is only a few tenths of an electronvolt,[16] i.e., much smaller than the width of the asymmetry features used to determine the surface magnetization, and thus causes a negligible perturbation. The critical exponent for the surface magnetization β_1 and the Curie temperature of the surface T_{Cs} are obtained by fitting the SPLEED data by a function of the type $A_{ex} \sim t^{\beta_1}$, where $t=(1-T/T_{Cs})$, as previously done.[17] Figures 1 and 2 show typical experimental SPLEED results. Measurements were performed at energies of 129, 126.5, 167, and 168 eV at an angle of incidence of 24° with respect to the surface normal. The results do not show a dependence on E of the fitted parameters within the statistical error. The mean value of the parameters obtained for all the measurements is $\beta_1=0.72 \pm 0.03$, in the range $0.002 < t \leq 0.16$, and $T_{Cs}=16.70 \pm 0.04$ K. The uncertainty in the determination of T_{Cs} from a given data set is typically ± 0.04 K. The high precision in the determination of the critical point is clearly illustrated in Fig. 2. The error in the determination of β_1 arises from the statistical error and is correlated to the uncertainty in the determination of T_{Cs}. The average value determined by the magneto-optic Kerr effect for the bulk Curie temperature is 16.71 ± 0.05 K. Note that the extinction length of the light used for the present magneto-optic Kerr effect ($h\nu=1.96$ eV) is about 1000 Å in EuS. Thus this measurement is sensitive to the *bulk* magnetization of the films only and surface effects, confined to the first few surface layers, can be *neglected* in these measurements.[18]

The values of T_{Cs} and T_{Cb} suggest that we observe an *ordinary* surface transition on EuS(111).[1] β_1 is, however, *clearly* smaller than the value *predicted* by the best theoretical estimates, $\beta_1=0.84 \pm 0.01$,[19] for the semi-infinite isotropic Heisenberg model. The discrepancy between experiment and theory amounts to 12%, roughly three times larger than the discrepancy between theory and the results obtained for the Ni surfaces,[6] where $\beta_1(\text{Ni})=0.8$ is found. A possible explanation for this discrepancy related to the fitting procedure's including data points at temperatures outside the asymptotic regime seems unlikely. Bulk EuS experiments indicate that the critical regime starts at $t \leq 0.14$.[20-22] Surface anisotropies of the type discussed by Selzer and Majlis, Costa, Mariz, and Tsallis, and dos Santos, Sarmento, and Tsallis[23] could cause a reduction of the temperature range of critical behavior.[24] In order to explore these

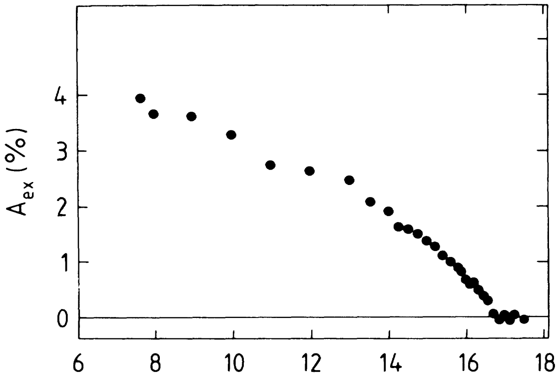

FIG. 1. Experimental SPLEED exchange asymmetry A_{ex} of the 00 beam vs temperature. The scattering angle for the polarized electrons with respect to the surface normal was $\theta=24°$.

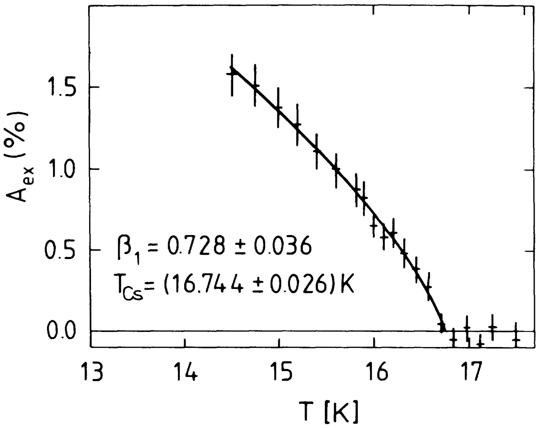

FIG. 2. Critical region of the data of Fig. 1 with the fitting of the functional form $A_{ex} \sim t^{\beta_1}$. The error bars represent statistical errors (one standard deviation).

possibilities we determined how the fitting parameters vary with the temperature range of the fitting. The results for three different data sets are collected in Fig. 3, which shows the value of β_1 as a function of the fitting range $0 < t < 0.3$. Within the present accuracy, the asymptotic regime seems to begin at $t \leq 0.16$.

We admittedly cannot fully rule out crossover effects since the analysis of Fig. 3 encompasses slightly less than two decades in reduced temperature. In the presence of anisotropic exchange coupling at the surface the effective exponent (in the crossover region) might be close to the critical exponent expected for the Ising-bulk, Ising-surface system $\beta_1 = 0.78 \pm 0.02$.[1,24-27] SPLEED experiments[7] on Gd have shown that surface couplings on rare earths can be different than in the bulk. The observations made in these experiments could therefore be rationalized by the assumption of different surface couplings J_1 between nearest neighbors *in* the surface layer and J_{12} between nearest neighbors of the first and second atomic layers. For J_1 values close to the critical value J_c—at which the "special" multicritical surface transition, with $T_{Cb} = T_{Cs}$, is expected to occur with $\beta_1^{\mathrm{sp}} \sim 0.25$ (cf. Refs. 1 and 24)—one indeed obtains a small effective β_1 in the crossover region. An exact determination of the critical exponents, however, is still pending, especially for the case of long-range magnetic anisotropies at the surface. It is interesting to note that by spin-polarized photoelectron emission on EuO(001) surfaces,[28,29] an indication of a nonsaturated surface layer was found. This layer could be magnetized only at very high magnetic fields. As suggested by the present study, those results can also be interpreted by the existence of a strong surface-induced anisotropy.

Experimental work on other magnetic surfaces as models of semi-infinite systems as well as on well-characterized two-dimensional systems[30] should shed further light on the origin of these discrepancies between theory and experiment. Finally, the observation of inelastic processes with high energy resolution, currently under way at our laboratory, should open up the possibility of also studying dynamical effects.

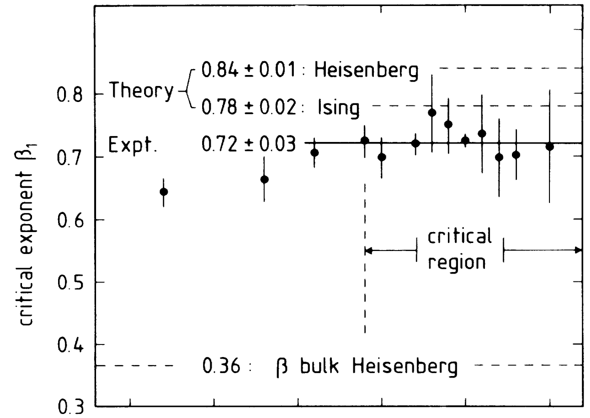

FIG. 3. Experimental mean value of β_1 from three different data sets determined by fitting of each one in the range $0 < t \leq 0.16$. The theoretical data are from Refs. 1 and 19.

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