

Magnetic Moments in Ultrathin Cr Films on Fe(100)

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Absolute magnetic moments of Cr are determined from *in situ* magnetometer measurements during growth of ultrathin Cr films on Fe(100). Cr moments in the first atomic layer are aligned antiparallel to the Fe moments and amount to $\geq 4\mu_B$ for submonolayer coverage and $3\mu_B$ for the first complete monolayer. This is the first experimental evidence of the huge moment enhancement predicted for Cr interfaces by band theory. Monte Carlo simulations indicate a drastic deviation from layer-antiferromagnetic order within the first 2–3 Cr atomic layers.

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Magnetic order in Cr layers on ferromagnetic Fe has been widely discussed since *ab initio* band calculations [1,2] predicted strongly enhanced magnetic moments for a Cr overlayer [$3.1\mu_B/\text{atom}$ [2] or $3.6\mu_B/\text{atom}$ [1] for 1 monolayer (ML) Cr on Fe(100) compared to the bulk moment of $0.59\mu_B$] and an antiparallel orientation of the Cr moments relative to the Fe magnetization. This interest was further stimulated by the discovery of an oscillatory exchange coupling between Fe layers through an intermediate Cr layer [3] and a giant magnetoresistance effect (GMR) in the case of antiferromagnetic coupling [4]. A strong influence of ordered Cr moments on both effects is to be expected; however, magnetic order in the Cr layer has not really been considered in many theoretical investigations as pointed out recently [5]. Indeed, ordered surface moments and a spin density wave antiferromagnetic order in Cr films on Fe(100) have recently been shown to persist up to 1.8 times the bulk Curie temperature ($T_N^b = 311$ K) and up to > 75 ML Cr [6]. For submonolayer Cr coverage the antiparallel alignment of the Cr moments relative to the moments of the Fe substrate has been shown by spin-resolved core level photoemission [7] and soft x-ray magnetic circular dichroism (SXMCD) [8]. The predicted giant Cr moments, however, have *not* been found experimentally up to now. Hillebrecht *et al.* [7] from their photoemission data estimate a value of 0.5 to $1\mu_B/\text{Cr atom}$ for a 1 ML Cr film; Idzerda *et al.* [8] infer a value of $(0.6 \pm 0.2)\mu_B$ (i.e., the bulk value) from their SXMCD measurements. Pierce, Celotta, and Unguris [9], however, have pointed out the difficulties in extracting absolute moment values from electron spectroscopy and scanning electron microscopy. These difficulties mainly arise from the fact that the measured spin polarization may well be proportional to the magnetic moment but the constant of proportionality is not known.

In order to clarify this situation it seems worthwhile and even necessary to measure the absolute magnetic moments of Cr films on Fe *directly* with an appropriate magnetometer. In this Letter we report on such an experiment. An *in situ* alternating gradient magnetometer (AGM) was used which allows one to continuously monitor the sample magnetic moment during growth in UHV

with submonolayer sensitivity. We find a pronounced decrease of the sample moment during the deposition of the first 4 ML of Cr. The Cr moment is found to be $\geq 3\mu_B$ for (sub)monolayer coverage and aligned antiparallel to the Fe magnetization. This is the first experimental verification of the huge moment enhancement theoretically predicted for Cr at interfaces. From Monte Carlo simulations we further conclude that large Cr moments persist in thicker layers and there is an evident deviation from layer-antiferromagnetic order in the first 2–3 Cr monolayers.

The film preparation was done by molecular beam epitaxy (MBE) in an ultrahigh vacuum (UHV) chamber equipped with low energy electron diffraction (LEED), Auger electron spectroscopy (AES), sputter cleaning, and sample annealing. During film growth the residual gas pressure was kept below 5×10^{-10} mbar. Resistively heated BeO crucibles were used for the evaporation of all materials. LiF crystals were cleaved along (100) planes and annealed in UHV at 600°C . Au films 300 to 800 Å thick were grown on these substrates at a temperature of 360°C .

Transmission electron diffraction proved that a perfect (100) orientation of the Au films resulted from a low growth rate of $0.5 \text{ \AA}/\text{min}$ and an 8 Å Cr seed layer. Bright field electron micrographs showed rectangular Au crystallites with $0.5\text{--}2 \mu\text{m}$ diameter.

Fe films of typically 7 ML ($\approx 10 \text{ \AA}$) thickness were grown on the Au base layer with a substrate temperature of 40°C and a rate of $\approx 1 \text{ \AA}/\text{min}$. Cr was deposited on top of the Fe layer at a temperature of 20°C and a rate of $\approx 0.1 \text{ \AA}/\text{min}$. Finally, the whole film was covered with a Au protective layer of several 1000 Å thickness. After each step the (100) orientation was verified by LEED.

A quartz crystal oscillator individually calibrated for each material was used to continuously monitor the thickness of the films. An independent thickness determination was carried out *ex situ* using x-ray fluorescence analysis (XFA). The mass coverage resulting from these measurements was converted into a nominal thickness in Å or ML assuming the bulk lattice constant. The resulting statistical uncertainty of the individual layer thickness

was below 0.3 ML.

Magnetization curves of the samples were continuously measured during film growth with an AGM. This instrument, which was built following the principle given by Flanders [10], to our knowledge is the first one to operate in UHV. The maximum field obtained with the UHV electromagnet is 9 kOe.

Extrapolation of the saturated part of the magnetization loop back to zero field yields the spontaneous magnetic moment, m_S , of the sample. A plot of m_S versus quartz crystal monitor frequency shift, Δf_Q , is shown in Fig. 1. The thickness of the Fe and Cr layers in ML is indicated by vertical lines.

The sample moment increases linearly with growing Fe thickness ($\Delta f_Q \leq 385 \text{ s}^{-1}$). From previous experimental studies [11] we know that the central layers of a 7 ML Fe film have essentially the bulk moment of $2.2\mu_B/\text{Fe atom}$. Therefore, the slope of the line in Fig. 1 at $t_{\text{Fe}}=7-8 \text{ ML}$ provides a calibration of the magnetometer. This calibration must be carried out for each sample because the sensitivity of the magnetometer depends on the mass and shape of the substrate. In addition, the calibration was checked independently using a calibrated superconducting quantum interference device (SQUID) magnetometer and proved to be correct with an error smaller than 5%.

When the Fe deposition is stopped and the deposition of Cr started ($\Delta f_Q=385 \text{ s}^{-1}$ in Fig. 1) an abrupt decrease of the sample magnetic moment is observed. Several tests were carried out to verify that this effect is exclusively caused by the growth of the Cr layer: The effect of surface contamination on the magnetic moment was shown to be negligible under the conditions of the experiment by operating the shutter in front of the Cr source and by an intentional increase of the residual gas pressure by several orders of magnitude.

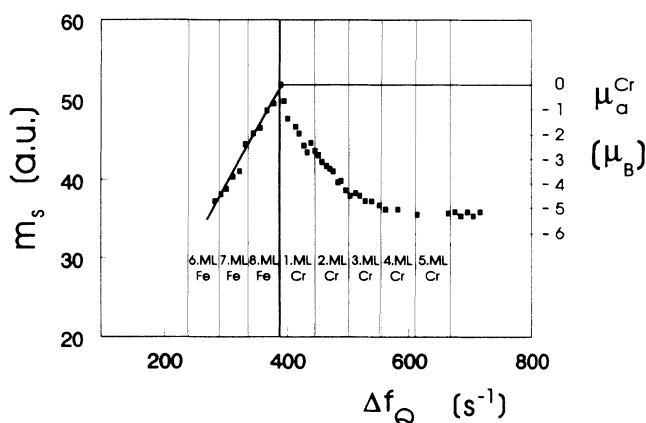


FIG. 1. Spontaneous magnetic moment at 20°C during growth of 8 ML Fe and 5.5 ML Cr as a function of frequency shift of the quartz crystal monitor. Complete monolayers are marked by vertical lines. Numbers at the right margin indicate the areal density of the Cr moments in μ_B per atom position in the film plane.

In principle, the reduction of the sample moment by covering with Cr could be a consequence of either a partial quenching of the Fe magnetic moment or of the magnetic moments of the Cr atoms themselves. There are several facts which indicate that the moment change of the Fe film due to the Cr coverage is not the main effect: Band calculations predict that the ground state interface moments of an Fe(100) layer covered by 1 ML Cr(100) is $1.96\mu_B/\text{atom}$ [1,2]. This means that the Fe moment is expected to be reduced by $1\mu_B$ compared to the free Fe(100) surface value of $2.98\mu_B$ [12,13]. However, the moment reduction observed at the beginning of Cr deposition amounts to $\geq 5\mu_B$ per Cr atom (Fig. 1).

In order to check whether the observed moment drop is connected with thermal spin excitations we have measured the temperature dependence of the spontaneous moment of a typical sample (8 ML Fe/5 ML Cr/1000 Å Au covering layer) with the SQUID magnetometer. (The *in situ* measurement must be performed at the growth temperature, i.e., 293 K.) The variation of m_S between 10 and 300 K of 8% was nearly the same as for an 8 ML Fe film without Cr.

We therefore conclude that the observed moment reduction by the Cr cover cannot be explained by a quenching of the Fe moments, but is a consequence of the ordered magnetic moments of the Cr itself. This also means that the Cr moments in the first monolayer have a net orientation antiparallel to the Fe moments in applied magnetic fields up to 8 kOe.

We first discuss the observations for a Cr coverage below a monolayer. From the slope of the curve of total moment versus Cr thickness (Fig. 1) immediately after the Cr deposition starts we deduce a Cr moment of $-(4.5 \pm 0.5)\mu_B/\text{atom}$ taking into account the moment reduction of the outermost Fe layer. The negative sign indicates the antiparallel orientation of the Cr moments relative to the positive Fe moments. The absolute value of the average Cr moment decreases to about $3\mu_B/\text{atom}$ for a nominal thickness of 1 ML.

These results are easily understood on the basis of band theory [1,2,12,13]: The average Cr moment of $-3\mu_B$ for a monolayer of Cr on Fe(100) agrees well with predicted values ($-3.1\mu_B/\text{atom}$ [2] or $-3.6\mu_B/\text{atom}$ [1]). For an incomplete Cr monolayer we may indeed expect a ground state moment which is still further enhanced relative to the bulk value due to the reduction of the coordination from a two-dimensional array to a linear chain and a single Cr atom on the Fe surface. It would be highly desirable to calculate the moment distribution for such submonolayer configurations and compare them to the present experiment.

Next, we discuss the sample moment for Cr coverages of more than 1 ML. For this purpose we introduce the areal density of magnetic moments, μ_a^{Cr} , by dividing the total magnetic moment of the Cr film, m_{Cr} , by the number of Cr atoms in one atomic plane.

In order to explain the evolution of the sample moment

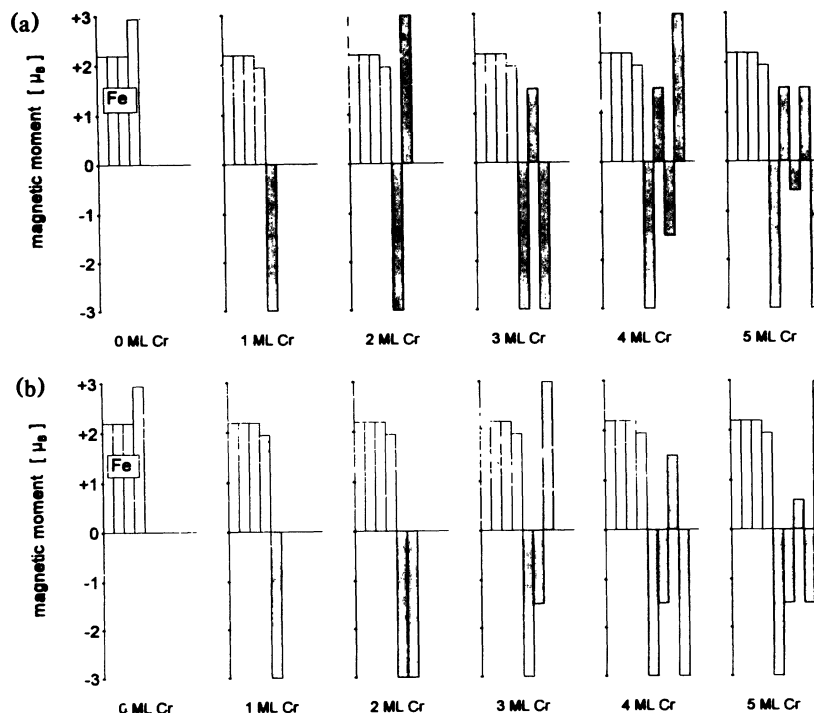


FIG. 2. Moment depth profiles for Cr films on Fe(100) derived from band calculations (see text); (a) layer-antiferromagnetic order throughout the Cr film; (b) parallel moments of the first 2 ML of Cr followed by layer antiferromagnetism.

with the growing Cr layer we have to make assumptions about (i) the depth profiles of the magnetic moments per Cr atom for each thickness of the Cr layer and (ii) the growth mode of the Cr film.

We performed Monte Carlo simulations to obtain the total moment of the Cr film as a function of the average thickness. In our calculations we assume a random arrival of Cr atoms at the surface; the growth mode is modeled by allowing a certain number of jumps on the surface with different probabilities for an upward and downward jump at the edge of a terrace. In this way, a perfect layer-by-layer growth results if a nearly infinite number of jumps is allowed and upward jumps are forbidden. A totally random growth will occur if jumps are not allowed at all.

Concerning the moment depth profiles, unfortunately, there are no band calculations available for the film structures used in our experiment. For this reason, we use the calculated result for 1 ML Cr on Fe(100) [1,2] and combine it with calculated moment depth profiles for thicker Cr layers [e.g., for a free-standing 7 ML Cr(100) film [14] or a Cr(100) film on Au(100) [15]]. The resulting depth profiles with a layer-antiferromagnetic (LAF) order throughout the Cr film as outlined in Fig. 2(a) are used together with the relative occupation numbers of the individual atomic layers for the case of a nearly perfect layer-by-layer growth to compute the areal density of the Cr moments as a function of Cr coverage. The result is plotted in Fig. 3 by round dots together with

the experimental data of Fig. 1 as a function of the average Cr thickness; it is characterized by strong oscillations with a period of 2 ML. Such oscillations are definitely absent in the experimental data. Taking into account the fact that a LAF order has been proved to exist in thicker Cr(100) films on Fe(100) [6,16], we therefore conclude

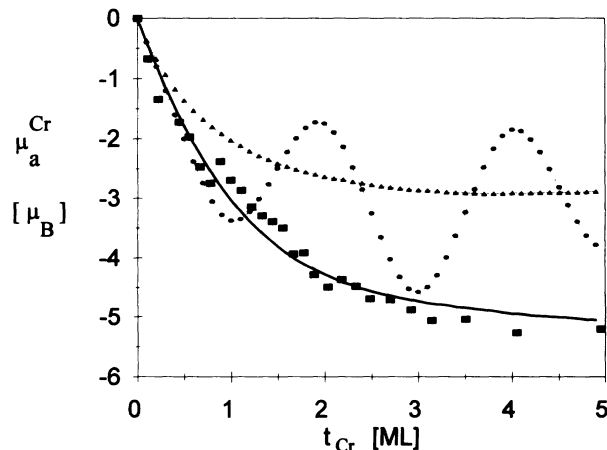


FIG. 3. Areal density of magnetic moments in Cr film on Fe(100) as a function of average Cr thickness; \blacksquare , experimental data from Fig. 1; \bullet , simulation for layer-by-layer growth and moment depth profiles according to Fig. 2(a); Δ , simulation for random growth and moment depth profiles from Fig. 2(a); solid line, simulation for random growth and moment depth profiles from Fig. 2(b).

that the growth of the Cr film does not occur layer by layer under the conditions of our experiment.

Next, we calculate the sample moment assuming a totally random growth and the same LAF moment depth profiles as before [Fig. 2(a)]. The resulting areal density of the magnetic moment is plotted in Fig. 3 by triangular symbols. We now find a qualitative agreement with the experimental data showing a continuous decrease of the sample moment and saturation around 4 ML Cr; however, the moment reduction is much smaller than observed experimentally.

This means that the moment depth profiles in our film must be different from the ones used in the simulation [Fig. 2(a)]. It can easily be shown that it is not possible to reproduce the experimental moment reduction of $5.2\mu_B$ per atomic position in the bcc(100) surface if we assume a LAF order and all Cr moments to be smaller than $5\mu_B$ which is the maximum value allowed by Hund's rule.

We therefore conclude that at least two adjacent Cr layers must have their magnetic moments parallel to each other and antiparallel to the Fe moment. A possible reason for such a behavior could be the Fe-Cr exchange coupling beyond nearest neighbors. It is then plausible to expect the parallel moment orientation for the first two Cr layers adjacent to the Fe film.

In Fig. 2(b) alternative moment depth profiles are shown which are modified according to the foregoing consideration. The sample moment calculated from these profiles for a random growth of the Cr film is plotted versus Cr thickness in Fig. 3 as a solid line. It is obvious that now the moment reduction observed in the experiment is well reproduced by the simulation.

This agreement, however, should not be overestimated because the growth mode is not exactly known at present and the moment depth profiles introduced here are not unique except the basic conclusion about the parallel magnetization of two adjacent Cr monolayers close to the Fe(100)-Cr interface.

In their recent Letter on the spin polarization of secondary electrons from a Cr film on Fe(100), Unguris, Celotta, and Pierce [6] reported a LAF order up to 75 ML Cr. However, a "defect" in the antiferromagnetic order was observed between 1 and 4 layers of Cr giving rise to a 180° phase shift of the Cr polarization (e.g., the moments of the fourth Cr layer are antiparallel to the Fe moments). In a subsequent paper [9] this phenomenon was studied in more detail and a possible correlation between this irregularity of the moment stacking and structural defects was considered to be responsible for an apparent discrepancy with data from another experiment [16], yet no explanation was given for the magnetic "stacking fault" itself. The present experimental results indicate that a fundamental deviation from layer-

antiferromagnetism within the first few monolayers of Cr on Fe(100) is the reason for the observed "irregularity" of the spin polarization.

In conclusion, in the present investigation absolute magnetic moments of ultrathin Cr layers on Fe(100) were measured *in situ* during film growth for the first time. We find a strong enhancement of the atomic moments in a Cr monolayer on Fe(100) in agreement with the results of previous band calculations [1,2]. This constitutes the first experimental verification of the predicted interface moment enhancement.

An even stronger moment enhancement found in sub-monolayer Cr films and the complex magnetic order of Cr moments close to the Fe substrate require further band and total energy calculations for corresponding model structures.

The present experiments will be extended to film preparation conditions, where a better layer-by-layer growth can be expected. The correct determination of the actual moment profiles in Cr layers on Fe including the constraint on the Cr moments by an additional Fe film will be of great value for the understanding of oscillatory exchange coupling and magnetoresistance in Fe/Cr multilayers.

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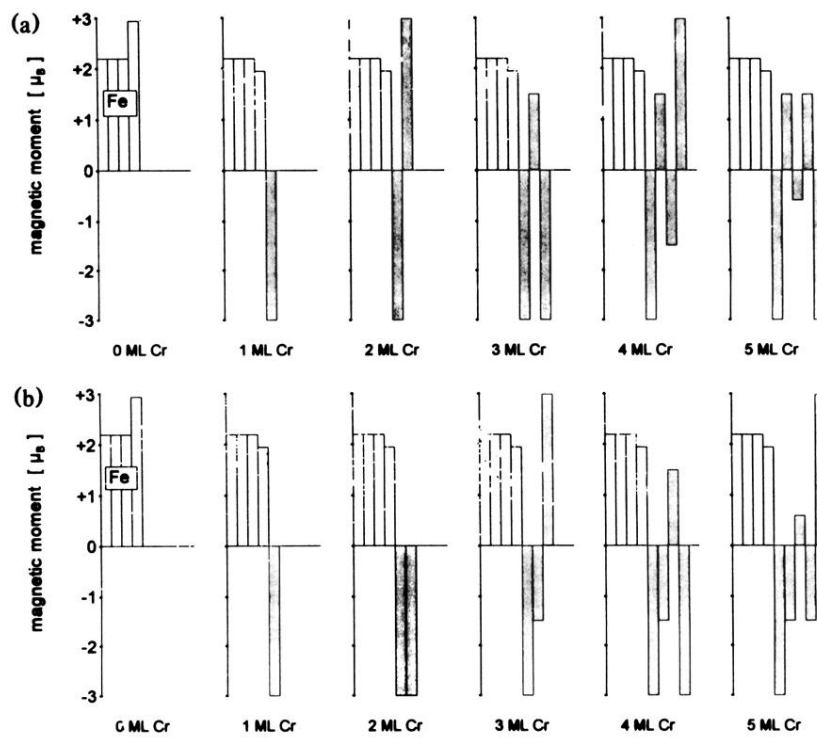


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