Critical Adsorption and Dimensional Crossover in Epitaxial FeCo Films

B. Nickel, W. Donner, and H. Dosch
Max-Planck-Institut für Metallforschung, D-70569 Stuttgart, Germany
and Universität Stuttgart, Institut für Theoretische und Angewandte Physik, D-70569 Stuttgart, Germany

C. Detlefs and G. Grübel
European Synchrotron Radiation Facility (ESRF), F-38042 Grenoble, France
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The critical behavior of thin FeCo films grown on MgO has been studied using phase-sensitive synchrotron x-ray diffraction. These studies unravel several novel features of criticality in thin films, as the simultaneous appearance of the 3D-2D crossover and the truncation of the correlation length normal to the film at approximately $1/3$ of the film thickness. Above the critical temperature of the film we observe a pronounced pinning of the order parameter at the MgO-FeCo interface, which indicates a novel critical adsorption behavior.

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The understanding of critical phenomena in thin films is one milestone in handling cooperative phenomena in partly reduced dimensions. It is predicted by theory [1,2] that the finite thickness (L) of the film gives rise to several universal deviations from the well-established bulk criticality governed by a diverging correlation length

$$\xi = \xi_0 t^{-\nu} \tag{1}$$

[with $t = (T - T_c)/T_c$ as the reduced temperature, $\nu = 2/3$ as a universal exponent, and ξ_0 as a microscopic length]. Within the concept of "finite size scaling" one expects that the critical temperature T_c is shifted and smeared out in temperature ("rounding") according to universal laws [2]. One fascination of critical thin films results from the predicted crossover from three-dimensional to two-dimensional criticality which should occur as the reduced temperature t crosses a characteristic value t_{cr}. This scenario has indeed been observed experimentally in various systems, as in binary fluids [3,4] and in thin Ni [5] and Fe films [6], by monitoring the average order parameter of the film with temperature.

Several key features of critical thin films are crucial for the theoretical concepts: (a) The power-law growth (1) of the correlation length ξ normal to the film is necessarily truncated at a finite value ξ^* which is of the order of the film thickness L. According to renormalization group theory $\xi^* = L/2.89$ [1], furthermore, the associated reduced temperature $t^* = (L/2.89\xi_0)^\nu$ should be identical to the crossover temperature t_{cr}. (b) The order parameter m of the system must exhibit—as all structural quantities of finite systems—a pronounced depth profile across the film; following [7] we thus write the thin film order parameter as a function of depth z in the form

$$m(z,t) = m_{\mathrm{hom}}(t) + \Delta m_A(t) e^{-z/\xi} + \Delta m_B(t) e^{(z-L)/\xi}, \tag{2}$$

with $m_{\mathrm{hom}}(t)$ as the homogeneous part of the order parameter, and $\Delta m_A(t)$ and $\Delta m_B(t)$ as order-parameter deviations adjacent to the walls A and B, respectively (see Fig. 1). Notice that the decay of the profile is governed by the system-inherent correlation length $\xi(t)$ across the film which should show the aforementioned truncation effect. The values of the wall-induced order-parameter changes Δm_A and Δm_B are nonuniversal and depend upon the microscopic details of the wall-films interactions. This raises several questions, in particular the following: (a) Can the relationship between the dimensional crossover and the truncation of the correlation length be confirmed experimentally? (b) Which order-parameter profiles evolve upon approaching $t = 0$ and which of the order-parameter contributions in (2) do show the universal crossover behavior?

We have performed a detailed x-ray scattering study of a thin film of FeCo which undergoes a continuous ($B2$-$A2$) order-disorder transition in the bulk [8,9]. As we will show in what follows, we obtain, by exploiting special two-beam interferences, the temperature-dependent order-parameter profiles across the film on an absolute scale (including the phase of the order) which allow us to give clear-cut answers to the open questions addressed above and, in particular,

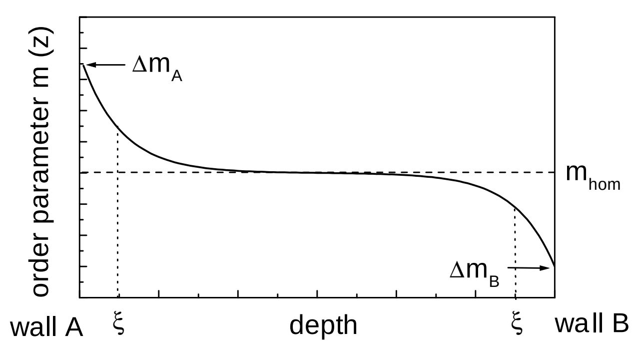

FIG. 1. Possible order-parameter profile emerging in a thin critical film of thickness L enclosed between walls A and B. $\Delta m_{A,B}$ are the wall enhancements and ξ is the (bulk) correlation length.

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to measure the temperature dependence of the quantities $m_{\text{hom}}(t)$, Δm_A, $\Delta m_B(t)$, and $\xi(t)$ which determine the total order-parameter profile within the thin film.

The FeCo(001) single crystal film has been grown by molecular beam epitaxy techniques using a growth rate of 0.5 Å/s on a MgO(001) which was kept at 250 °C during the growth. The evaporation rates of Fe and Co have been determined prior to the growth with a quartz balance placed at the sample position. Low energy electron diffraction studies revealed a single crystal film grown with a 45° epitaxy with respect to the substrate lattice. The actual mosaic spread of the film has been determined by x-ray diffraction to be 0.3°. Directly after growth the FeCo film does not exhibit any *B*2 order which emerges only after annealing the film at 600 °C. Prior to the x-ray experiment the sample has been cycled several times through the order-disorder temperature to assure homogeneity. The actual composition of the FeCo film has been determined by x-ray fluorescence spectroscopy to be $Fe_{44}Co_{56}$. After growth and characterization the sample is transferred into a mobile ultrahigh vacuum diffraction chamber equipped with a Be window and a radiation heating stage. The sample temperature is measured via thermocouple and optical pyrometry to a relative accuracy of ±50 mK. The x-ray experiments have been performed at the TROIKA beam line (ID10A) of the ESRF in Grenoble which is supplied with highly brilliant undulator radiation. A monochromatic 12 keV x-ray beam is selected by a diamond (111) monochromator and cleaned from higher harmonics by a Rh-coated mirror.

X-ray diffraction from thin films gives rise to characteristic Laue thickness fringes which contain the information on the structural properties across the film. The scattering geometry and the relevant part of the reciprocal lattice is shown schematically in Fig. 2 depicting the center of the bcc-related (002) and the order-related (001) Laue fringes of the FeCo film as well as the MgO(002) reflection and the MgO(00L) Bragg rod amplitude which emanates from the MgO interface and interferes coherently with the film Laue fringes. The amplitude of the MgO(00L) Bragg rod is given by

$$A_{\text{MgO}} = F_{\text{MgO}}(q_z)\,\frac{e^{-(q_z\sigma_{\text{MgO}})^2}}{1 - e^{-i(q_z a_{\text{MgO}})}} \tag{3}$$

(F_{MgO} is the form factor of the MgO unit cell, q_z is the momentum transfer normal to the film, and σ_{MgO} is the substrate roughness), while the Laue fringes of the homogeneously ordered FeCo film of thickness N read

$$A_{\text{FeCo}} = F_{\text{FeCo}}(z,q_z)\,\frac{1 - e^{i(q_z a_{\text{FeCo}}N)}}{1 - e^{i(q_z a_{\text{FeCo}})}}\,e^{-(q_z\sigma_{\text{FeCo}})^2}, \tag{4}$$

and contain in the case of the (001) reflection the information on the order parameter $m(z)$ within the film which enters the $F_{\text{FeCo}}(z,q_z)$ by

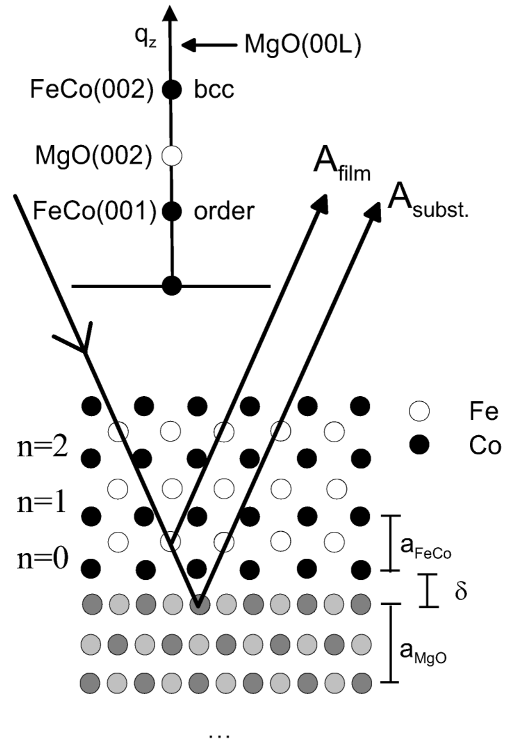

FIG. 2. Side view on the sample: the FeCo film is grown in registry with the MgO substrate (below), separated by a distance δ. In the ordered *B*2 state the film consists of alternating lattice planes with Fe and Co occupation. The upper part depicts the section of reciprocal space relevant to this study (for further explanation see main text).

$$\begin{aligned} F_{\text{FeCo}}(z,q_z) = {} & f_{\text{Fe}}\Big(\frac{m(z)}{2} + c_{\text{Fe}}\Big) + f_{\text{Co}}\Big(\frac{-m(z)}{2} + c_{\text{Co}}\Big) \\ & + \Big[f_{\text{Co}}\Big(\frac{m(z)}{2} + c_{\text{Co}}\Big) \\ & + f_{\text{Fe}}\Big(\frac{-m(z)}{2} + c_{\text{Fe}}\Big)\Big]e^{i(q_z a_{\text{FeCo}}/2)}, \end{aligned} \tag{5}$$

where c_X denotes the concentration of the species X and f_X its atomic form factor. In Eq. (4) σ_{FeCo} denotes the surface roughness of the film.

We explicitly exploit the interference between the MgO rod amplitude and the FeCo(001) Laue amplitude to extract the $m(z,T)$ directly, including the (phase) information on the site occupancy by Fe and Co. The total intensity across the FeCo(001) order-related Laue fringes thus reads

$$\begin{aligned} I(q_z) &= |A_{\text{MgO}} + e^{iq_z\delta}A_{\text{FeCo}}|^2 \\ &= I_{\text{MgO}}(q_z) + I_{\text{FeCo}}(q_z) \\ &\quad + 2\text{Re}\{A_{\text{FeCo}}A^*_{\text{MgO}}e^{iq_z\delta}\}. \end{aligned} \tag{6}$$

Note that the interference term retains, also in the presence of roughness, all the information on the epitaxial distance δ between the laterally averaged first FeCo layer and the laterally averaged MgO substrate layer (see Fig. 2) as well as on the absolute value of the order-parameter profile $m(z)$.

Figure 3a shows typical scans across FeCo(001) as obtained for several different temperatures below and above T_c. Not shown is the (temperature-independent) fundamental FeCo(002) Laue fringes which have simultaneously been measured and used to calibrate the intercepted intensity on absolute scale [allowing one to access the order-parameter profile $m(z,T)$ on an absolute scale] and to determine all structural parameters of the film: A least squares fit to the data provides the layer thickness $L = 121$ unit cells, the lattice constant perpendicular to the film $a_{\mathrm{FeCo}} = 2.873$ Å, and the roughnesses $\sigma_{\mathrm{MgO}} = 1.3$ Å and $\sigma_{\mathrm{FeCo}} = 2.9$ Å.

Inspection of the order-related (001) Laue fringes reveals a pronounced asymmetry in the scattering distribution which is a direct action of the interference term in Eq. (6). The full lines in Fig. 3a are least squares fits according to Eqs. (2) and (6), the resulting order-parameter profiles $m(z,T)$ are shown in Fig. 3b for some selected temperatures below and above T_c: They exhibit a temperature-dependent homogeneous part m_{hom} and a noticeable profile Δm_A at the MgO-FeCo interface which is slowly developing and spatially expanding upon approaching T_c. Within our experimental error we do not observe any noticeable profile at the FeCo-vacuum interface (i.e., we set $\Delta m_A = 0$ for all temperatures). The peculiar shapes of the Bragg profile for $T > T_c$ can completely be understood (full lines) by assuming $m_{\mathrm{hom}} = 0$ and an exponentially decaying order-parameter profile pinned at the MgO-FeCo interface with a temperature-dependent correlation length ξ [10].

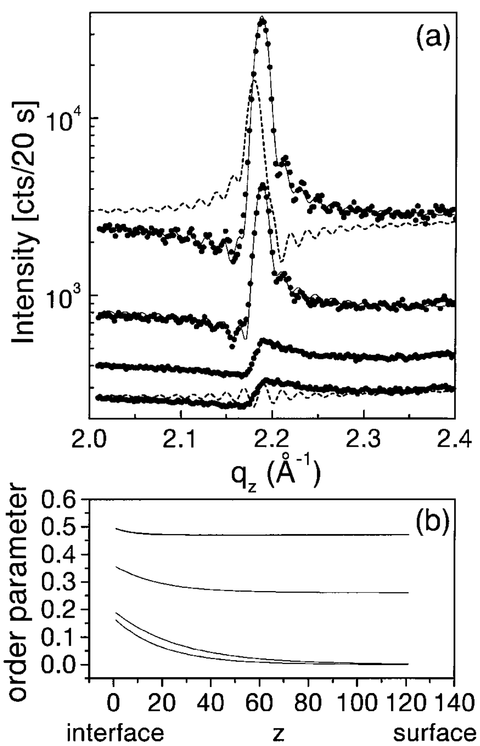

FIG. 3. (a) q_z scans through the (001) reciprocal lattice point at different temperatures above T_c (from top to bottom: 10 and 3 K below T_c and 1.6 and 3 K above T_c). Symbols represent measured data, and solid lines are fits as described in the text. The dashed lines in (a) are simulations according to alternative models (see text). The upper curves are shifted by factors 2.5, 5, and 10 for clarity. (b) Order-parameter profiles underlying the fits in the upper part in the same temperature sequence as in (a).

We note at this point the merit of the phase-sensitive diffraction mode. The interference term in Eq. (6) unambiguously fixes (a) the film-substrate spacing to be $\delta = 2.30$ Å, (b) the Co-rich termination at the FeCo-MgO interface, and (c) the pinning of the high-temperature order at the MgO-FeCo interface. For comparison and for the demonstration of the phase sensitivity of the recorded scattering profile we show by way of example in Fig. 3a two calculated profiles (dashed lines) for $T < T_c$ and $T > T_c$ associated with an Fe-enriched layer at the MgO-FeCo interface and with a pinning of the high-temperature order at the FeCo-vacuum interfaces, respectively (which is apparently not observed). Note also that the observable order-parameter profile at the MgO-FeCo interface maps out the correlation length ξ directly in real space. This effect will be used later to measure the temperature dependence of ξ normal to the film. The high quality of our data together with the absolute calibration and phase sensitivity recovers directly the temperature dependence of the order-parameter profiles $m(z,T)$ and its contributions $m_{\mathrm{hom}}(t)$, $\Delta m_A(t)$, and $\xi(t)$ and allow us by this to get a detailed picture of the temperature-dependent ordering phenomena in the film which will be discussed in what follows.

Figure 4a shows the temperature dependence of the homogeneous part m_{hom} of the order-parameter profile versus temperature. It disappears continuously at a temperature which we define as the critical temperature T_c of the film. The inset depicts a double-logarithmic plot of m_{hom} versus reduced temperature with T_c suggesting a crossover behavior from a three-dimensional to a two-dimensional critical behavior: Away from $t = 0$ we observe a power-law behavior with 3D-power-law exponent $\beta = 0.347 \pm 0.016$, below a reduced temperature $t_{\mathrm{cr}} = 6 \times 10^{-4}$ the 2D-power-law exponent $\beta = 0.125$ becomes visible. Since m_{hom} is obtained on an absolute scale, we also can deduce the order-parameter amplitude $(m_{\mathrm{hom}})_0$ which enters the the power-law $m_{\mathrm{hom}} = (m_{\mathrm{hom}})_0 t^{\beta}$. Within the 3D-criticality regime we obtain $(m_{\mathrm{hom}})_0 = 1.79 \pm 0.1$ which has to be compared with the theoretical prediction of $(m_{\mathrm{hom}})_0 = 1.49$ [11]. The slight discrepancy may be reconciled by adjusting the range of interactions used in the theory.

The wall-induced enhancement of the order parameter, Δm_A, is also shown in Fig. 4a as a function of temperature. It exhibits a rather complicated behavior: for $T < T_c$ it is almost constant followed by a pronounced increase around T_c with a maximum achieved slightly above T_c. Upon

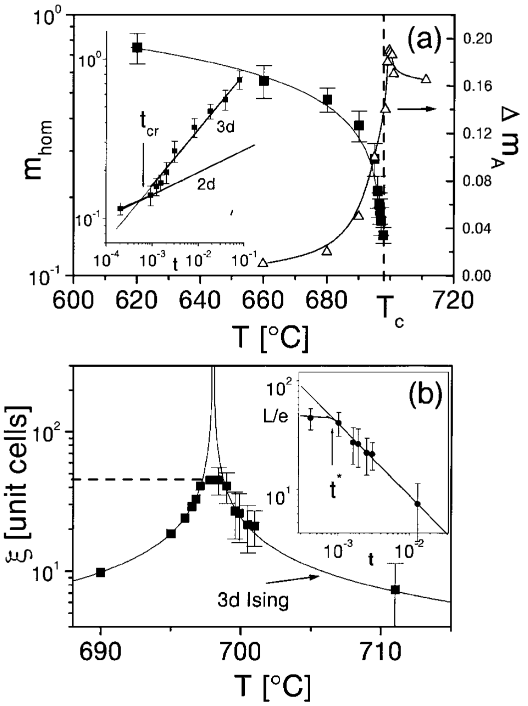

FIG. 4. (a) Order parameter vs temperature for the vertically homogeneous part m_{hom} (filled squares) and the first-layer enhancement Δm_A (open triangles). The full line through the m_{hom} data is a fit, the curve through Δm_A is a guide to the eye. The inset is plotted on a double-logarithmic scale and shows the crossover of the two slopes for m_{hom}. (b) Decay length of the order-parameter profile as a function of temperature together with the expected 3D Ising behavior. The horizontal dashed line denotes 1/3 of the film thickness. Inset: double-logarithmic representation.

further heating Δm_A decreases slowly giving rise to a nonzero order pinned at the MgO-FeCo interface. The maximum order enhancement is $\Delta m_A = 0.19$ equivalent to an average Co enhancement at the first layer of $\Delta c_{\mathrm{Co}} = 8.7$ at. %. While the microscopic origin of the Co segregation at the MgO interface is not clear at the moment, the mechanism which leads to a segregation-induced order pinning is well understood and mediated by the symmetry break of the FeCo(001) interface [9,12].

Finally we come to the temperature dependence of the correlation length ξ normal to the film which is deduced from the exponential decay of the order-parameter profiles at the MgO-FeCo interface below and above T_c. The results are summarized in Fig. 4b together with the bulk behavior of the critical correlation length (full line). We find that our data are fully consistent with a truncation of the correlation length at approximately $\xi^* = L/3$ which is in perfect agreement with the aforementioned theoretical prediction ($L/2.89$ [1]). Most interestingly, however, the truncation takes place at a reduced temperature $t^* = 8 \times 10^{-4}$ being—within the experimental error—almost exactly identical with t_{cr} discussed above which governs the 3D-2D crossover of the homogeneous order parameter. Thus, these experiments give the first direct experimental evidence that the dimensional crossover in thin films is mediated by the truncation of the correlation length. As a further observation we note that, within the 3D regime, the correlation length matches perfectly the bulk behavior including the prefactor ξ_0, which is found in this study to be $\xi_0 = 0.43 \pm 0.15$ in very good agreement with the theoretical value $\xi_0 = 0.448$ [11].

In conclusion, we have presented a phase-sensitive and absolutely calibrated x-ray diffraction study of the critical order-parameter profiles in thin FeCo(001) films grown on MgO(001). We find that the order parameter develops a pronounced profile at the MgO-FeCo interfaces upon approaching T_c. According to our study the homogeneous part of the profile exhibits universal behavior which matches quantitatively the theoretical predictions. In particular, we find a crossover from 3D to 2D criticality at a reduced temperature $t_{\mathrm{cr}} = 6 \times 10^{-4}$ which turns out to be virtually identical to the reduced temperature which governs the truncation of the correlation length perpendicular to the film. The truncation of the correlation length occurs when ξ reaches the value $\xi^* = L/3$ in agreement with theory. Above T_c a remaining order is discovered which is pinned at the MgO-FeCo interfaces and sparked by a Co segregation at the first FeCo layer adjacent to the MgO interface.

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