Magnetic Frustration in Ultrathin Fe Films

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In Fe(110) films prepared at 300 K on W(110), ferromagnetic long range order is suppressed in a range of coverages between 1.20 and 1.48 pseudomorphic monolayers, whereas below and above this gap of long range order, monolayer and double layer ferromagnetism is observed with Curie temperatures of 230 and roughly 450 K, respectively. Apparently, the frustration of long range order in the gap is caused by indirect interactions of electronic origin between double layer islands, mediated by the surrounding monolayer and its W substrate.

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The unusual magnetic properties of ultrathin films are caused in part by reduced dimensionality and in part by their microstructure. Scanning tunneling microscopy (STM) offers the chance to elucidate the interplay between morphology and magnetism in ultrathin films, and considerable progress has been achieved recently in understanding important aspects of ultrathin film magnetism from STM studies [1–4]. In the present Letter we report on a new coupling phenomenon, observed in uncovered pseudomorphic Fe(110) films on W(110), to be interpreted from their microstructure as observed by STM. Pseudomorphic monolayers (ML) of Fe on W(110) were used previously as model systems of 2D magnetism [4–6] because of their thermodynamic stability [7]. The uncovered ML is ferromagnetic with $T_C(\mathrm{ML}) = 230$ K [4]. The new phenomenon occurs between 1.20 and 1.48 ML, where ferromagnetic long range order is suppressed at least down to 115 K, STM images in this gap of long range order show separated double layer islands on a closed monolayer. Our data suggest that long range order is suppressed by a quasiantiferromagnetic frustrating interaction between the double layer islands, probably of indirect electronic origin. This is an analog to the frustration of magnetic order by competing interactions in spin glasses [8]. Because the islands would be superparamagnetic without interaction, the frustrated island system could then be addressed as a super spin glass [9].

Our Fe films were grown in UHV at room temperature (RT) on atomically clean W(110) substrates. Film thickness was controlled by quartz oscillator monitors and structure was checked by low-energy electron diffraction spectroscopy (LEED) and Auger electron spectroscopy (AES). The growth mode for coverages between $\theta = 0.8$ and 1.9 (pseudomorphic) ML is shown by STM images in Fig. 1. At $\theta = 1.0$, and second monolayer nucleates on a nearly complete first monolayer. Coalescence of the double layer patches proceeds between 1.4 and 1.7 ML. For $\theta \geq 1.7$ one observes substantial third layer contributions and the incipient relaxation of the misfit by dislocations. We focus on the range $\theta \leq 1.6$ where the films were pseudomorphic with the W substrate (strained by 10.4% in the plane to accommodate the misfit $f_{\mathrm{FeW}} = -9.4\%$), and were composed of monolayer and double layer patches.

For magnetic analysis, we used spin-polarized electron scattering (SPLEED), torsion oscillation magnetometry (TOM) [10], and conversion electron Mössbauer spectroscopy (CEMS) [11] in separate UHV systems, but with identical growth conditions. For SPLEED, we used scattering geometries as described previously [4], with the quantization axis of the electron spin polarization along the easy axis [$1\bar{1}0$] of the magnetic film. The magnetic signal is then the exchange asymmetry A_{ex} [4], the properly nor-

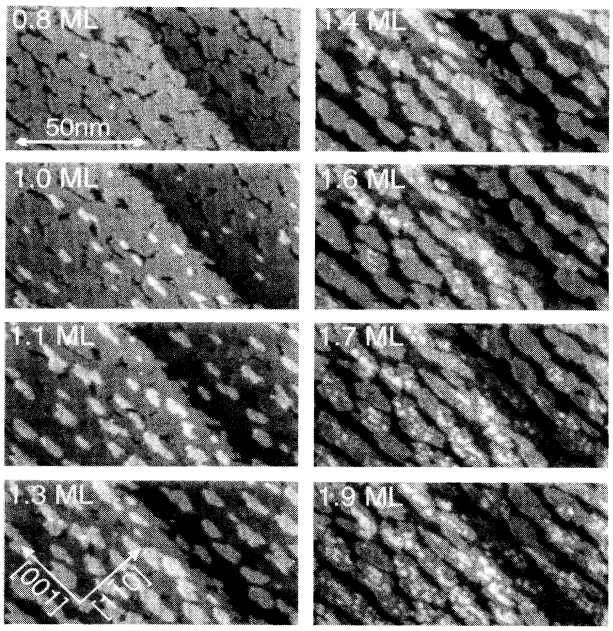

FIG. 1. STM images of Fe films prepared on W(110) at 300 K. Coverages θ in units of pseudomorphic monolayers are indicated.

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malized difference in electron reflectivities for spin parallel or antiparallel to $[1\bar{1}0]$. A_{ex} is roughly proportional to the magnetization [12]. External fields up to 200 Oe could be applied along $[1\bar{1}0]$, generated by currents through the W substrate. SPLEED was possible in fields up to 2 Oe. Immediately below T_c we obtained, even in these low fields, standard easy axis loops with coercivities down to 0.1 Oe, and perfect saturation in the remanent state, e.g., the inset in Fig. 2. This is an important consequence of the strong uniaxial in-plane anisotropy of the samples [13]. It enabled us to restrict the measurements for $T < T_c$ to the remanent state, and to take remanence as saturation. By field cooling down to 115 K in 200 Oe (applied as 20 msec pulses in order to avoid Joule heating), the samples were locked near T_c into the saturated state, in which they remained at lower temperatures, where the rapid rise of H_c beyond 200 Oe prevented any (re)magnetization. We then followed the remanent value of P_0A_{ex} (primary beam spin polarization $P_0 = 20\%$) versus T during warming up. Results are shown for some samples in Fig. 2. The temperature dependence was reversible. An extended series of samples is represented in Fig. 3(a) by the Curie temperature T_C, respectively, both versus θ. With respect to $A_{ex}(T)$, four regime of coverage θ can be distinguished. In regime I, $\theta \leq 0.58$, the films are nonmagnetic; see [4]. In regime II, $0.58 < \theta < 1.2$, represented in Fig. 2 by three samples, finite values of A_{ex} indicate long range remanent order up to roughly the Curie temperature of the monolayer, $T_C(\mathrm{ML}) = 230$ K [4]. Near the upper limit of this regime, A_{ex} decreases rapidly with increasing θ [Fig. 3(a)]. In regime III, $1.20 < \theta < 1.48$, we observe a gap of long range order, in the sense that finite values of A_{ex} could be observed neither in remanence after field cooling nor in external fields up to 2 Oe for $T \geq 115$ K, for all samples in the gap. Long range order returned quite abruptly in regime IV, $\theta \geq 1.48$. The Curie temperatures [see Fig. 3(b)] were now always above 300 K. The steepness of the transition between the gap regime III and the magnetic regime IV is documented by comparing the easy axis loop given as the inset in Fig. 2, taken from a film $\theta = 1.48$ just above the gap ($H_c = 0.1$ Oe) with the inability to obtain any magnetic signal for the last film in the gap ($\theta = 1.47$), either in remanence after field cooling or in external fields up to 2 Oe. Because the disappearance of H_c near T_c is a general phenomenon, an explanation of the gap from a mere change of coercivity is not possible. A remarkable feature of samples immediately above the gap ($\theta = 1.48$, 1.49, and 1.50; see Fig. 2) is the nonmonotonic dependence $A_{ex}(T)$. Because the upper end of the gap coincides with the coalescence of the double layer patches (see Fig. 1), we conclude that the group of T_C values approximately 375 K at this upper end belongs to an interconnected double layer network. The Curie temperature of the double layer (DL) is roughly estimated as $T_C(\mathrm{DL}) = 450$ K.

In order to understand the puzzling absence of magnetic long range order in the gap, we took CEMS spectra from pure ^{57}Fe films in the center of the gap, $\theta = 1.3$, at temperatures $T_m = 250$ and 150 K, respectively, just above and below $T_C(\mathrm{ML}) = 230$ K. They are shown in Figs. 4(a) and 4(b), respectively. At 250 K, only 20 K above $T_C(\mathrm{ML})$, we observed a purely (super)paramagnetic quadrupole doublet [Fig. 4(a)]. Below $T_C(\mathrm{ML})$ instead [Fig. 4(b)], we observed a pattern which can be fitted by a superposition of three magnetic sextets, with $B_1 = 11$ T, $B_2 = 19$ T, and $B_3 = 31$ T, without any

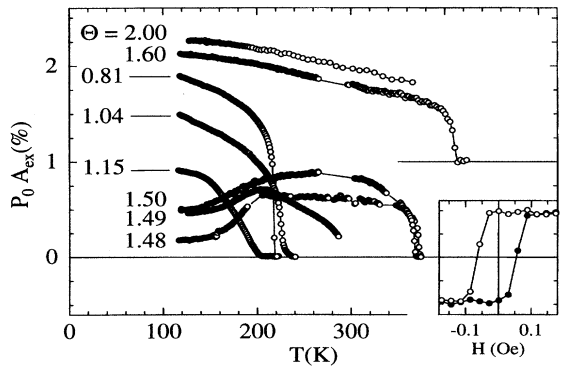

FIG. 2. Exchange asymmetry P_0A_{ex} versus temperature T for Fe(110) films on W(110), with coverage θ as a parameter. The inset shows the magnetization loop for a film $\theta = 1.48$, 10 K below T_C.

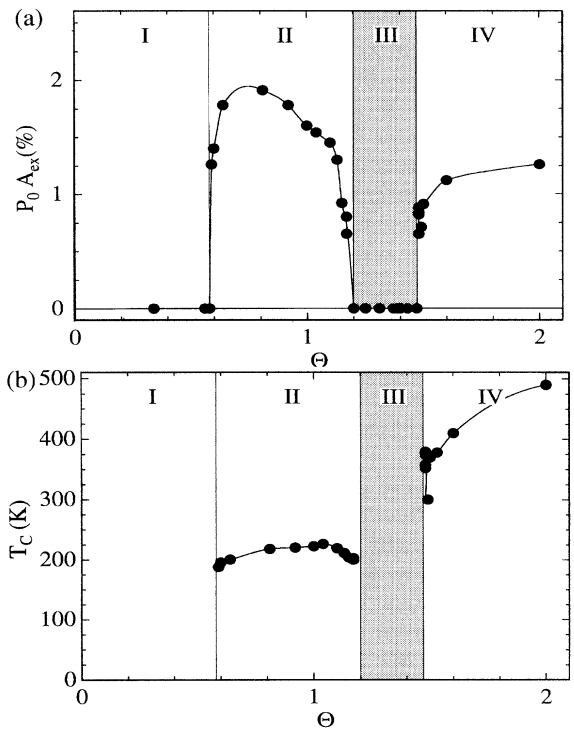

FIG. 3. (a) Maximum values of P_0A_{ex} observed for temperatures $T > 115$ K versus Fe coverage θ. (b) Curie temperatures T_C versus θ. Regimes I–IV are as discussed in the main text. The gap of long range order in regime III is obvious.

(super)paramagnetic component. The fields agree with those of the bare monolayer, 10 T [5]; of the W/Fe(110) interface, 21 T [14]; and of the free Fe surface, 31 T [11]. The spectrum thus indicates a blocked superposition of magnetic monolayer and double layer patches. This is confirmed by the relative contributions $p_1 = 56\%$, $p_2 = p_3 = 22\%$, from which one obtains $\theta = 1.28$, in excellent agreement with the value from the quartz balance, $\theta = 1.30$. The interpretation of Fig. 4(a) is obvious: At 250 K, the monolayer patches are paramagnetic because the temperature is above T_C(ML), and the double layer islands obviously are superparamagnetic, because T is below T_C(DL). A rough estimate based on the island sizes from Fig. 1 and on estimates of the uniaxial in-plane anisotropies of the islands [4,13] indicates that the blocking temperature would be well below 150 K if the double layer islands were noninteracting. Obviously, they interact by the monolayer between, and we observe in our samples a quite uncommon mode of blocking caused rather by magnetic freezing of the monolayer substrate than by the usual intrinsic slowing down of thermal fluctuations in the double layer patches. The locking of the whole sample into a microscopically ordered state near T_C(ML), as shown by Fig. 4(b), is reasonable. The absence of long range macroscopic order is surprising. Because the samples are magnetic on a nanoscale [Fig. 4(b)], they must be considered as a static superposition of uniaxial double layer islands, magnetized statistically up and down the easy axis [1$\bar{1}$0], with a complicated twisted state of the monolayer regions between. Could this compensated state be explained as a result of a strange freezing of the superparamagnetic fluctuations of the double layer islands by the freezing monolayer? The TOM experiments to be described below show that this is not possible, but that the compensation of magnetic moments apparently results from a frustrating interaction between the double layer islands.

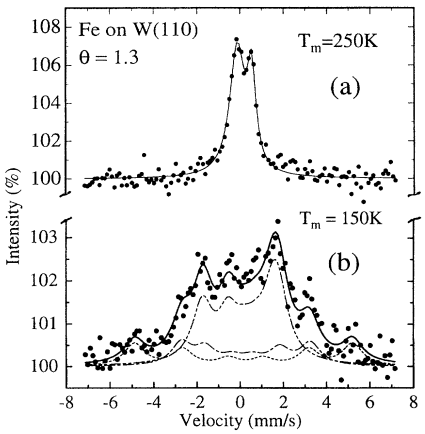

FIG. 4. CEMS spectra of a pure ^{57}Fe film on W(110), $\theta = 1.3$, prepared at 300 K, and measured in zero field (a) at 250 K and (b) at 150 K.

TOM experiments were done at 300 K. For their interpretation, it is crucial that the double layer islands of all films in the gap be superparamagnetic at RT if they are noninteracting. This follows conclusively from the superparamagnetic fluctuations of the film in the center on the 10^{-7} sec scale of CEMS [Fig. 4(a)]. Standard superparamagnetic theory tells that the blocking area for TOM at 300 K is 4.5 times larger than that for CEMS at 250 K. The island size increases roughly by this factor from the center to the upper limit of the gap. Hence, all islands in the gap would be superparamagnetic at RT in the absence of interactions. Straightforward calculation then shows that the samples should easily saturate in standard laboratory fields. In the maximum field available in TOM, $\mu_0 H = 0.4$ T, one would expect saturation (>95%) for all films in the gap at 300 K in the absence of noninteracting islands.

We tried to observe such a magnetic saturation in the gap at 300 K using TOM, with a definite negative result, as shown in Fig. 5. Note that we measure in TOM a magnetic torque constant R, and that for the case of a magnetically saturated sample, R is connected with the saturation moment m_0 by $R/H = m_0/(1 + H/H_L)$ with an out-of-plane anisotropy field H_L that is usually of the order of some tesla. This is just the signature of the thick films above the gap ($\theta = 2.7$, 2.2, and 1.9). The film in the center of the gap, $\theta = 1.35$, behaves completely differently, showing only negative values of R/H without any indication of saturation. This was reproduced for several samples. The data were taken with the field parallel to the film plane. In this geometry, negative values of R/H could be explained by perpendicular magnetization, which, however, could be excluded by additional measurements with the field along the surface normal. Straightforward analysis of TOM shows that negative values of R/H are also expected for a quasiantiferromagnetic system of uniaxial particles with equal or comparable abundance of moments parallel and antiparallel to H, respectively. The frustration of saturation for R/H, therefore, must be read as frustration of magnetic saturation. The rapid fluctuation, as shown by Fig. 4(a), excludes any hysteresis effects. Nevertheless, the expected magnetic saturation is not even indicated. *We conclude that an interaction between the double layer patches must be active even at 300 K which stabilizes the macroscopically nonmagnetic state even in this rapidly fluctuating system. This interaction induces a frustration of the long range order in the double layer island system which cannot be removed by external fields up to 0.4 T.* The magnitude of this field excludes the magnetostatic nature of the interaction. We guess that this frustrating, virtually antiferromagnetic interaction between the double layer patches is of electronic origin, mediated indirectly by the monolayer elements in common with the W substrate. The details of this interaction remain to be explained. Negative values of R/H are still observed for the films with $\theta = 1.6$ and 1.7 in Fig. 5. From Figs. 2 and 3, one would expect that these films would be out of the

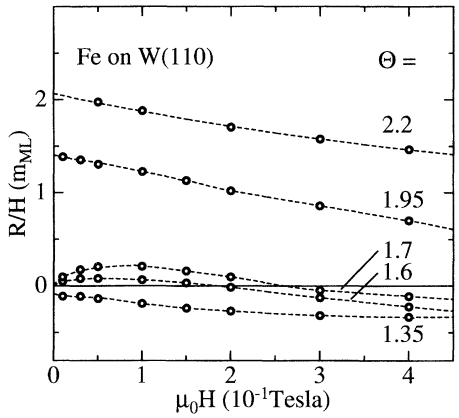

FIG. 5. Torsion oscillation magnetrometry of Fe(110) films on W(110) at 300 K. R/H (magnetic torque constant R per magnetic field H), in units of m_{ML} (the saturation moment of a pseudomorphic monolayer with $2.17\mu_B$ per atom), versus H which is applied along [1$\bar{1}$0].

gap. However, the residual gas exposure, after flashing the W substrate, was larger for the films in TOM than for those in SPLEED, and such exposures were observed to extend the upper bound of the gap. Accordingly, the samples $\theta = 1.6$ and 1.7 may have been in an extended gap, or just above its upper bound, where the frustrating interaction persists.

The frustrating interaction provides a natural explanation for the drop of A_{ex} for temperatures below T_C(ML) in the films $\theta = 1.48$, 1.49, and 1.50 in Fig. 2. Near the upper end of the gap, we expect a delicate balance between the frustrating interaction, transmitted by the monolayer patches, and the ferromagnetic interaction by the coalescence bridges. It is reasonable that the former is enhanced for $T < T_C$(ML), and therefore results in the observed drop of A_{ex} which indicates the incipient frustration. In general, the magnetic properties of the samples in the gap and near its upper bound can be conveniently discussed in terms of interacting magnetic particles. This suggests using an extension of the notion of superparamagnetism to interacting particles which has been proposed by Mørup *et al.* [9]. Superparamagnets are systems in which the atomic moments of paramagnets are replaced by the moments of magnetic particles. In the gap, where the frustrating interactions between the islands prevail, the films may then be addressed as super spin glasses [9], in which the magnetic ions of the spin glass are replaced by the interacting double layer islands. When the ferromagnetic bridge coupling overcomes the frustrating coupling at the upper end of the gap, $\theta = 1.48$, the system switches form a super spin glass to a superferromagnet, in which the atomic moments of the ferromagnet are replaced by the moments of the particles.

Superferromagnetism provides a natural explanation for the results of Back *et al.* [15], who investigated Fe films on W(110) just above the gap and observed giant susceptibilities $\chi = 3 \times 10^5$ near T_C, which they explained in terms of giant regions of correlated spins. To interpret these results in terms of superferromagnetism, note that the paramagnetic susceptibility of a uniaxial superferromagnet above T_C in a molecular field approximation is enhanced by N, the number of atoms in the particle. It is given by $\chi = C/(T - T_C)$, with a Curie constant $C \approx 2N$ K for Fe [9]. The mean value of N at the upper end of the gap is of the order of 10^4. Having in mind that correlations result in an enhancement of χ above the molecular field value, the giant susceptibilities observed by Back *et al.* can be easily explained as superferromagnetic. Moreover, the sensitive dependence of T_C on coverages by several metals, which was observed by Weber *et al.* [16] for films just above the gap, is easily understood from the balance between frustrating and bridge interactions, and the electronic nature of the former.

In conclusion, we have shown that clean Fe(110) films prepared on W(110) at 300 K show a gap of ferromagnetic long range magnetic order for coverages between 1.2 and 1.47 pseudomorphic monolayers. The frustration of magnetic order in this gap results apparently from a virtually antiferromagnetic interaction of electronic origin between double layer islands, mediated by the surrounding monolayer sea and its W substrate. The nature of this interaction remains to be explained in detail. The films in the gap can be considered as super spin glasses. At the upper end of the gap, they switch to superferromagnets when the ferromagnetic coupling by bridges overcomes the frustrating interaction. Much remains to be done for a complete understanding of this new type of interaction and its evidence in ultrathin films.

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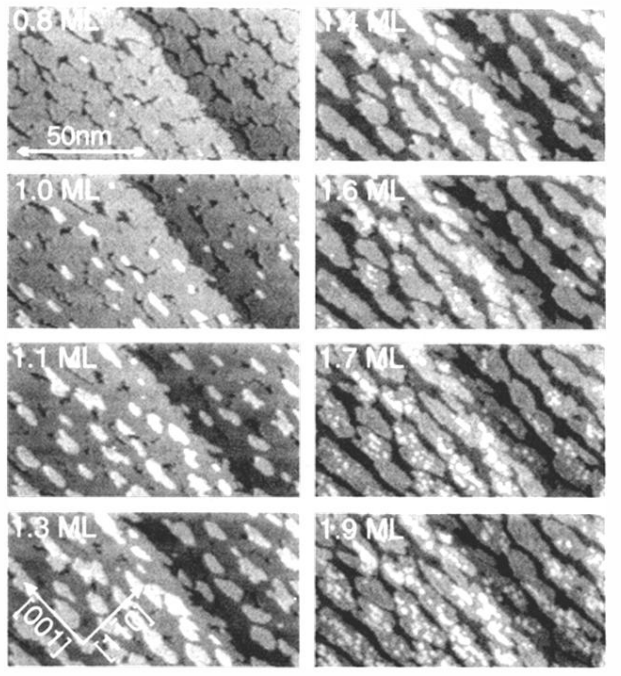

FIG. 1. STM images of Fe films prepared on W(110) at 300 K. Coverages θ in units of pseudomorphic monolayers are indicated.