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To cite this version:
U. Gradmann, H. Elmers, M. Przybylski. MAGNETIC PROPERTIES OF ULTRA-THIN FILMS. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-1665-C8-1669. 10.1051/jphyscol:19888760. jpa-00229004

HAL Id: jpa-00229004
https://hal.archives-ouvertes.fr/jpa-00229004
Submitted on 1 Jan 1988

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MAGNETIC PROPERTIES OF ULTRA-THIN FILMS

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Abstract. — Magnetic properties of ultra-thin films are reported with main emphasis on recent analysis of Fe(110)-films on W(110) using Conversion Electron Mössbauer Spectroscopy (CEMS) and Torsion Oscillation Magnetometry (TOM), including the ferromagnetic pseudomorphic monolayer Fe(110) on W(110), which forms a thermodynamically stable system.

1. Magnetic and structural properties of ultrathin films

The rich variety of magnetic phenomena in thin films is based on their structural peculiarities. The main concern of the present article are intrinsic properties, which are inherently connected with the thin film geometry as follows.

Finite thickness and loss of translational symmetry along the surface normal result in magnetic size effects, like reduction of $J_s$ and $T_c$ [1, 2], and inhomogeneities of magnetic order; loss of point symmetry for the surface atoms results in magnetic surface anisotropies [3]; finally, electronic interaction with epitaxial substrate and coating material opens the broad field of surface and interface chemistry, which is in separably connected with magnetism of thin films, even for ideal epitaxial films.

In general, these intrinsic magnetic film properties are superimposed by defect-induced properties, caused by chemical reaction with the substrate or by complicated non-equilibrium structures (columns, grains etc.) connected with the preparation by quenched condensation. However, even in noninterdiffusing systems and for careful preparation at elevated temperatures, defects induced by mechanical interaction with the epitaxial substrate must be considered. For finite misfit, and interface dislocations pseudomorphic strain with resulting magnetic strain anisotropies are inevitable.

For the experimental analysis of intrinsic magnetic properties in ultrathin films, an important problem is how to avoid the formation of islands in the initial stages of growth. This is a wetting problem, which has been neglected frequently. In equilibrium, layer-by-layer growth can be expected only if the film wets the substrate [4], and this applies if the surface free energy $\gamma_{sub str}$ of the substrate is larger than that of the film, $\gamma_{film}$, increased by that of the interface, $\gamma_{intef}$. Accordingly, the condition for layer growth in equilibrium is given by

$$\gamma_{sub str} > \gamma_{film} + \gamma_{intef}.$$  \(1\)

For the possibilities to prepare metastable layer-by-layer structures, even for weak violation of equation (1), compare [5, 6]. In metallic systems, where $\gamma_{intef}$ is small in comparison with surface energies, the chances for monolayer stability can be estimated from surface energies only. In recent experiments on ultrathin magnetic films, Fe ($\gamma_{Fe}=2.0 J m^{-2}$ [7]) was prepared on Ag(001) [8-10] or Cu(001) [11], where layer growth can hardly be expected because of $\gamma_{Ag}=1.1 J m^{-2}$ and $\gamma_{Cu}=1.7 J m^{-2}$, respectively. Actually, clustering has been detected for Fe on Ag, [9] and Fe on Cu [12].

For basic research on ultrathin magnetic films down to the monolayer, wetting epitaxial systems like Ni(111) on Re(0001) ($\gamma_{Ni}=1.9 J m^{-2}$, $\gamma_{Re}=2.2 J m^{-2}$ [11-17] or Fe(110) on W(110) ($\gamma_{W}=2.9 J m^{-2}$) are preferred. The present review is restricted to recent experiments with Fe(110) on W(110), performed in our laboratory during the last years using in situ experiments with monolayer resolution Conversion Electron Mössbauer Spectroscopy (CEMS [18, 19]) and high resolution Torsion Oscillation Magnetometry (TOM [13]). For reviews on other wetting ultrathin magnetic film systems compare [2, 14].

The paper is organized as follows.

After some remarks on epitaxial growth of Fe(110) on W(110), given in section 2, magnetic size effects, magnetic surface anisotropies and interface magnetoc-chemistry are discussed in sections 3, 4 and 5, respectively. The concluding section 6 is concerned with ferromagnetism in the pseudomorphic monolayer and double layer.

2. Epitaxial growth of Fe(110) on W(110)

The wetting condition $\gamma_{W}>\gamma_{Fe}$ indicates layer by layer growth of Fe(110) on W(110), which has been established both by Auger Electron Spectroscopy (AES) [20, 21] and CEMS [21]. However, details of the film structure are determined by the lattice misfit $f_{W/F}=\left(\alpha_{F}-\alpha_{W}\right)/\alpha_{W}=-9.4\%$. It turns out that the first monolayer is pseudomorphic with the substrate, in-
dependently of the varying growth conditions. In the following, film thickness is given in bulk monolayers, \( D \). The pseudomorphic monolayer then is given by \( D = 0.82 \). It forms an extremely stable system; even in a Fe/W-interface, the first Fe(ll0) layer remains pseudomorphic with W(110), as has been shown by CEMS [22]. The pseudomorphic double layer, however, seems to be a metastable system, which can be prepared at room temperature only [21]; conversely, when prepared at 200 °C, the second monolayer has the density of bulk Fe; for \( 2 \leq D < 10 \), films are modulated by the strain field of periodic misfit dislocations, which fade out for \( D > 10 \). To get smooth surfaces for \( D > 5 \), enhanced preparation temperatures \( T_p > 200 \) °C must be used. Unfortunately, recrystallization into three-dimensional islands on a stable monolayer must be considered for annealing above 400 °C. As a result, Curie-temperatures cannot be reached, with the exception of the monolayer \( (T_c \approx 290 \) K, compare Sect. 6, below).

3. Magnetic size effects

The reduction of \( J_s (T; D) \) and \( T_c (D) \) with decreasing \( D \) has been analyzed in detail previously for Fe Ni- and NiFe-films using TOM [2, 13-17], resulting in good agreement with existing theories. For the present films Fe(110) on W(110), detailed analysis was done using CEMS [24]. For the temperature dependence of the mean hyperfine field \( B_{hf} (T) \), which represents the temperature dependence of the mean film magnetization to a very good approximation, for low temperatures, excellent agreement was found between the experiments [24] and Green's functions calculations [25]. The unique possibility of CEMS, however, is local analysis of magnetic order via the magnetic hyperfine field \( B_{hf} \). Both the local structure of \( B_{hf} \) near surfaces, which is different from that of \( J_s \), and its temperature dependence, which equals that of \( J_s \), at least at low temperatures, was measured in detail for Fe(110)-films on W(110) consisting of 21 atomic layers, both uncoated and coated by Ag [18, 19, 26, 27]. Near the Ag-coated and the free surface, Fe57 probe monolayers could be used for local analysis [18, 19, 27], near the W/Fe-interface, where enhanced interdiffusion prevented the use of monolayer probes, the strong changes of \( B_{hf} \) induced by the W-substrate itself could be used for local analysis [26]. For the uncoated surface, a Friedel-type oscillations was observed for \( B_{hf} (0) \), which is decreased in the first monolayer by 2.4 % and increased in the 2nd by 2.2 %, in comparison with bulk values. This has been confirmed qualitatively by band calculations [28], but with quantitative deviations (7 % reduction and 10 % enhancement in the 1st and 2nd layer, respectively). For Ag-coated films of type W(110) [21] Ag, consisting of \( D = 21 \) layers, a complete local analysis both of ground state field \( B_{hf} (0) \) and of its temperature dependence has been done [26]. In agreement with recent theories [29], \( B_{hf} (T) \) could be fitted by a \( T^{-3/2} \)-law \( B_{hf} (T) = B_{hf} (0) (1 - bT^{3/2}) \) for all atomic layers. Profiles of both \( B_{hf} (0) \) and spin-wave parameters \( b \) across the film are shown in figures 1a and b. Friedel-type oscillations of \( B_{hf} (0) \) are not observed near the Ag-coated surface, because they are connected with the spatial cutoff of the conduction electron gas, present in the free surface, missing in the Ag-coated one. The 3 % increase of \( B_{hf} (0) \) near Ag and its 30 % decrease near W remain to be explained. Spin-wave parameters in the interface are enhanced roughly by a factor 2 in comparison with the centre, in nice agreement with Rado's classical predictions from spin-wave theory [30] for ideal surfaces. Note that at the surface of a spin-glass Ni40Fe40B20 a surface enhancement of \( b \) by a factor 3 was observed [31], indicating weakened exchange near the surface [29]. Conversely, our result indicates homogeneous exchange up to the topmost monolayer of Fe(110), which is assumed in Rados work.
4. Magnetic surface anisotropies

A detailed review on magnetic surface anisotropies has been given recently [3]. In the present paper, this report is supplemented by a methodic remark as follows:

Magnetic surface anisotropies can be indicated or even estimated from the phenomenon of magnetization switching, with decreasing film thickness e.g. from the film plane, which is supported by bulk anisotropies, to the film normal, as supported apparently by out-of-plane surface anisotropies. This perpendicular magnetization has been observed recently for $\alpha$-Fe on Ag(100) [11] and for $\gamma$-Fe(100) on Cu(100) [8-10] and interpreted in terms of surface anisotropies. However, conclusive detection and quantitative determination of surface anisotropies definitely requires a linear dependence of film anisotropies on $1/D$, in order to separate surface type from volume type anisotropies, which can be very strong in ultrathin films, caused by pseudomorphic strain. There are even examples for perpendicular magnetization, caused by strain anisotropies, in the presence of easyplane surface anisotropies (Ni(111) on Cu(111) [32]). Strong anisotropies in ultrathin films are not necessarily of surface type.

The same remark applies to in-plane anisotropies, too. For the case of in-plane surface anisotropies of Fe(110), an estimate from magnetization switching ($K_{s,p}^{Fe/W}+K_{s,p}^{Fe/UHV} = 0.11 \times 10^{-3}$ J m$^{-2}$) [33] definitely deviates from a recent quantitative determination using the $1/D$-plot, $K_{s,p}^{Fe/W}+K_{s,p}^{Fe/UHV} = 0.54 \times 10^{-3}$ J m$^{-2}$ [34]. Again, the observation of switching gives a rough indication of surface anisotropies only, which must quantitatively be determined using the $1/D$-plot.

Out-of-plane surface anisotropies of Fe(110) on W(110) are positive [35], in our notation [3], that means they have the film plane as an easy plane, support shape anisotropy and therefore cannot be detected qualitatively by magnetization switching. The $1/D$-analysis results, roughly speaking, in $\mu_0 H_S^{Fe/W}+\mu_0 H_S^{Fe/Au} = (7 \pm 1)$ Tesla, for $x = UHV$, Cu, Ag, Au, respectively; for details compare [35].

5. Interface magnetochemistry

The dependence of magnetic film or surface properties on coating material has been analyzed, for the case of Ni(111), to some detail, for Cu, Pd and oxygen as coating materials [13, 15, 17]. For the case of Fe(110), the influence of noble metal coatings (Cu, Ag, Au) on magnetic moments and surface anisotropies has been measured recently [35]. Negligible changes in magnetic moments were observed, in contrast to previous experiments with polycrystalline Fe-films, for which strong changes of moment were reported [36, 37]. Considerable dependence on coating was observed, however, for in-plane surface anisotropies, resulting in magnetization switching as a result of coating only, as shown in figure 2. Note that $R/H$ (magnetic torque constant $R$; magnetic field $H$) approximately equals the magnetic moment (component along external field) according to

$$m = (R/H) / (1 + H/H_L)$$

![Fig. 2. – Magnetization curves of a $D = 30$ layers Fe(110)-film on W(110), magnetic field $H$ along [110]. The easy-axis loop of the as-prepared film (o) is changed to a hard-axis loop by Au-coating (●), caused by a change of in-plane surface anisotropies $\mu_0 H_{s,p}^{Fe} + \mu_0 H_{s,p}^{Fe/UHV} = 0.54 \times 10^{-3}$ J m$^{-2}$ for the uncoated film to $\mu_0 H_{s,p}^{Fe} + \mu_0 H_{s,p}^{Au} = -0.08 \times 10^{-3}$ J m$^{-2}$ for the Au-coated one. Annealing at 500 K for 130 min recovers the easy-axis loop.](image-url)
where $H_L$ is an out-of-plane anisotropy. The crucial role of interface anisotropies and their strong dependence on the chemical state of the interface for magnetic reversal properties of any magnetic ultrathin film devices is clearly demonstrated by the figure.

6. Monolayer magnetism

The thermodynamic stability of the pseudomorphic monolayer of Fe(110) on W(110) provides the unique possibility to study two-dimensional magnetism of one single monolayer, with common translational symmetry of substrate and film. Experimental analysis of this system has been done recently using both CEMS [21, 23, 38-40] and TOM [41]. At the present state, the most important results can be summarized as follows.

(1) The Fe(110)-monolayer on W(110), coated by Ag, is ferromagnetic, with a monolayer Curie-temperature $T_c = (290 \pm 10)$ K. The magnetic hyperfine field $B_{hf}(T)$, as a function of temperature $T$, resembles that for bulk material. Extrapolation to $T = 0$ results in $B_{hf}(0) = (11.9 \pm 0.3)$ Tesla, strongly reduced in comparison of 33.9 Tesla for bulk material [23, 38, 40].

(2) If the Fe(110)-monolayer on W(110) is prepared and coated by Ag at 300 K, its Curie-temperature $T_c(D)$, as a function of bulk number of layers $D$, as measured by CEMS, increases monotonously with $D$ near the pseudomorphic monolayer $D = 0.82$; the error in $T_c$ given above is caused by this increase [40]. If, however, the preparation is done at 500 K, $T_c = (282 \pm 5)$ K is measured independently of $D$, for $0.5 \leq D \leq 0.82$. It represents the intrinsic monolayer $T_c$ of large size monolayer patches [40]. The most reliable monolayer analysis therefore comes from films prepared at 500 K.

(3) For the uncoated monolayer, $B_{hf}(0)$ and $T_c$ are reduced to $B_{hf}(0) = (10 \pm 1)$ Tesla and $T_c = (210 \pm 10)$ K, respectively.

(4) Magnetometry of the Fe(110)-monolayer on W(110) prepared at 500 K, coated by Ag, shows clearly the phase transition near 290 K found by CEMS, compare figure 3. For $T \leq 220$ K, square loops are observed. The coercive field disappears at 282 K, just the $T_c$ measured by CEMS. Magnetostructural measurements of the magnetic moment for $T \geq 220$ K and extrapolation to $T = 0$ using $B_{hf}(T)$ from CEMS, compare figure 4, results in atomic moments per atom $\mu = 1.17 \mu_{bulk} = 2.60 \mu_B$, in surprisingly good agreement with the theoretical prediction [28] $\mu = 2.56 \mu_B$ for the uncoated monolayer. This is reasonable, because Ag-coating of a “thick” layer ($D \geq 5$) does not change its moment [35].
(5) The pseudomorphic double layer \((D = 1.64)\) can be prepared as a metastable structure at \(T = 300\) K \([21]\). When coated by Ag, it shows a (mean) magnetic moment per atom \(\mu = 2.46 \mu_B\), again in fair agreement with the theoretical prediction \(\mu = 2.60 \mu_B\).

(6) If films just above the pseudomorphic monolayer \((0.82 < D < 1.2)\) are prepared at 500 K, the magnetic moment \(m(T)\) versus temperature \(T\) can be explained by superposition of nearly independent monolayer and double layer patches; in particular, the monolayer \(T_c = 290\) K can clearly be seen.

In our opinion, the analysis of such thermodynamically stable monolayer systems like Fe(110) on W(110) provides the most promising and conclusive approach to monolayer ferromagnetism.

[34] Elmers, H. J. and Gradmann, U., to be published.