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Magnetization of free Fe(110) surfaces from thin film magnetometry

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Abstract

Using torsion oscillation magnetometry of uncovered Fe(110) films on W(110) in situ in UHV, we have determined a surface excess moment in free Fe(110) surfaces of 0.39(16) monolayer equivalents. The results are compared with previous determinations using SPLEED, and with theoretical values.

Keywords: Surface magnetism; Thin film

1. Introduction

A fundamental phenomenon of surface magnetism is the enhancement of the magnetic moments in free ferromagnetic surfaces in comparison with their bulk values. In theory, the problem has been treated extensively by tight binding methods [1] and by first-principles band structure calculations (see Ref. [2] and references therein). The enhancement results from band narrowing and therefore increases with decreasing coordination. Accordingly, for the magnetic moment in the first monolayer, enhancements of 31% and 19% have been calculated for bcc Fe(100) surfaces [3] (4 neighbors) and Fe(110) surfaces [4] (6 neighbors), respectively. In experiments, the determination of magnetic moments in ferromagnetic surfaces has turned out to be difficult. The only experimental method applied so far is spin-polarized low-energy electron diffraction (SPLEED) at the ferromagnetic surface, in combination with multiple scattering calculations of the SPLEED spectra. First applications of the method to Ni surfaces resulted in non-significant values only for the net surface enhancement of magnetic moment, per surface Ni atom, given by 0.03(3) μ_B (5(5)% of a monolayer moment) for Ni(100) [5] and 0.06(12) μ_B (10(20)%) for Ni(111) [6], respectively, in rough agreement with theoretical values of 0.07 μ_B (12%) [7] and 0.11 μ_B (18%) [8], respectively.

The second ferromagnetic surface to which the SPLEED method has been applied is Fe(110). SPLEED measurements by Waller [9] have been evaluated by Tamura et al. [10]. It turned out that the experimental data could be best reproduced with the assumption of an enhancement of the top layer magnetization by about 35%, and a reduction in the second layer by about 15%, at room temperature, corresponding to roughly 38% or 21% in the ground

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state. However, Ormeci et al. [11], providing an alternative, careful theoretical discussion of Waller's data, had difficulty in reaching an unambiguous conclusion at all on the surface moment of Fe(110). From the experimental point of view, the question of the surface magnetization in Fe(110) therefore must be considered as open. An independent experimental determination is desired. The present paper presents such an independent experimental determination of the surface magnetization in Fe(110).

Our experimental approach is based on torsion oscillation magnetometry (TOM) of uncovered epitaxial Fe(110) films on W(110), performed in situ in UHV, for variable temperatures between 140 and 300 K. It will be shown that the appropriate extrapolation of measured magnetic moments to the ground state, and analysis of their dependence on film thickness results in a net enhancement of 32(15)% for both interfaces of the W(110)/Fe(110) samples together. In combination with a recent result of Pasyuk et al. [12], who determined a moment reduction of 7(4)% in the W(110)/Fe(110) interface, we finally find a ground state surface enhancement of 39(16)% for the free Fe(110) surface.

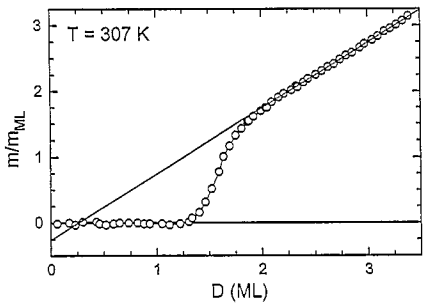

Fig. 1. Torsion oscillation magnetometry during growth of Fe(110) on W(110), at $T = 307$ K. Magnetic moment m in units of the monolayer moment m_{ML}, versus the number of (bulk) monolayers, D.

2. Experimental

The experiments were performed in two separate UHV systems, both equipped for preparing Fe films by epitaxy on atomically clean W(110) surfaces at pressures below 10^{-10} Torr and structural testing using LEED and AES. The thickness $t = Dd$ of a film consisting of D atomic layers with layer distance d was measured with an accuracy better than 0.1 ML using quartz oscillator monitors, which were calibrated using magnetometry of thick Fe films. The Fe films were prepared by an optimized growth mode described previously [13], starting the preparation with a preparation temperature $T_p = 300$ K which was held at up to $D = 10$ in order to avoid islanding, and raising T_p gradually to 500 K for $D > 10$, in order to obtain films with atomically smooth surfaces. The films showed sharp $p(1 \times 1)$ LEED patterns and had Auger-clean surfaces.

Both UHV systems were equipped with torsion oscillation magnetometers (TOMs) [14], capable of measuring the magnetic moments and out-of plane magnetic anisotropies, with a detection limit of the order of 0.01 ML Fe. In the first system (U-TOM I, [14]), magnetometry is possible during film growth, at temperatures above 290 K (see Fig. 1). In the second system (U-TOM II, [15]), magnetometry is possible for $130 < T < 700$ K. Preparation and magnetometry have to be done in different stages of U-TOM II; continuous magnetometry during film growth is therefore not possible.

3. Results

TOM was performed with external fields up to 0.4 T along $[1\bar{1}0]$. Due to the strong in-plane magnetic surface anisotropy (MSA) with easy axis $[1\bar{1}0]$ [16], all films showed square easy axis loops. In the following, we discuss the saturation magnetic moment $m(T;D)$ of the loops, which was measured in TOM for constant temperatures T as a function of D. Fig. 1 shows the evolution of $m(307\text{ K};D)$ as measured in U-TOM I during preparation. For $D < 1.2$, we observe $m = 0$, because the Curie temperature of the monolayer ($T_{C,mono} = 225$ K [17]) is below the temperature of the experiment. Near $D = 1.5$, we observe a steep increase in m, as a result of magnetic percolation in the double-layer island system [18]. For $D > 3$, we observe a linear increase in m, which can be described as

$$m(T;D) = m_{ML}(T)\,D + \Delta m(T), \qquad (1)$$

where $m_{ML}(T)$ is the moment of a bulk monolayer at a given T. Note the non-trivial result that the devia-

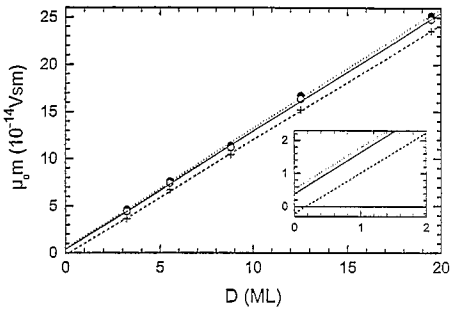

Fig. 2. Magnetic moment m versus number of atomic layers D, for a series of Fe(110) films on W(110). Crosses and broken line represent experimental data obtained at 285 K, full points (dotted lines) and open circles (full line) extrapoations to $T = 0$ using Eq. (4a) (Fig. 3a) and Eq. (4b) (Fig. 3b), respectively.

tion $\Delta m(T)$ is independent of D, for $D \geq 3$. However, $\Delta m(T)$ is not the ground state surface moment modification ('surface effect') which we ask for, as has been erroneously suggested by others [19]. Instead,

$$\Delta m(T) = \left[\Delta m_{\text{surf}}^{\text{W/Fe}} + \Delta m_{\text{surf}}^{\text{Fe/UHV}}\right] + \Delta m_{\text{size}}(T) \tag{2}$$

is composed of the true surface effect (terms in parentheses), which we assume to be independent of temperature, and the temperature-dependent 'size effect' $\Delta m_{\text{size}}(T)$, which describes the enhanced temperature dependence of magnetic order in thin films. Our task then is to measure $m(T;D)$ at finite temperatures, to determine $\Delta m(T)$, and finally to extrapolate to the ground state value,

$$\Delta m(0) = \Delta m_{\text{surf}} = \Delta m_{\text{surf}}^{\text{W/Fe}} + \Delta m_{\text{surf}}^{\text{Fe/UHV}}, \tag{3}$$

which in turn is composed of separate contributions $\Delta m_{\text{surf}}^{\text{W/Fe}}$ and $\Delta m_{\text{surf}}^{\text{Fe/UHV}}$ from the W/Fe interface and from the surface, respectively. Of course, we should use the range where $\Delta m(T)$ actually becomes independent of D, which is the case for $D > 2$ (see Fig. 1).

We measured $m(T;D)$ for a series with $D = 3.2$, 5.5, 8.8, 12.5 and 19.5 at temperatures between 140 and 285 K. The result for $T = 285$ K, $m(285\text{ K};D)$ versus D, is shown in Fig. 2 by the broken line (and the crosses). The slope of 1.221×10^{-14} V s m equals the monolayer moment $\mu_0 m_{\text{ML}}(285\text{ K})$. The problem is the extrapolation to $T = 0$. Because of the limited temperature range of our measurements, this extrapolation can be a good estimate only. We used two alternative extrapolation schemes. In scheme (a), we used a linear extrapolation, in the spirit of standard spin-wave theory statements for isotropic ultrathin films [20,21]. Scheme (a), written as

$$m(T;D) = m^{(a)}(0;D)(1 - a_D T), \tag{4a}$$

certainly overestimates the temperature dependence and therefore the value of $\Delta m(T) = -a_D T m^{(a)}(0;D)$.

For an alternative and more realistic fit (b), we took advantage of a previous observation in ultrathin Ag-covered Fe(110) films on W(110), which were analyzed by conversion electron Mössbauer spectroscopy (CEMS) [22]. It turned out that the temperature dependence of the magnetic hyperfine field, which is expected to equal the temperature dependence of the magnetization, can be described, down to the monolayer, by a Bloch $T^{3/2}$ law to a surprisingly good approximation. Our alternative and more realistic fit (b) is therefore chosen as

$$m(T;D) = m^{(b)}(0;D)\left(1 - b_D T^{3/2}\right). \tag{4b}$$

One further advantage of fit (b) is that it follows the thermodynamic requirement of zero slope at $T = 0$,

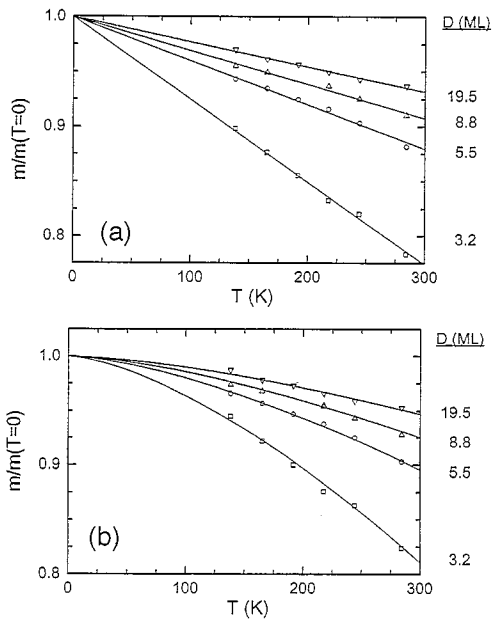

Fig. 3. Extrapolations of the magnetic moment m to $T = 0$, (a) by the linear scheme of Eq. (4a), and (b) by the 3/2 scheme of Eq. (4b).

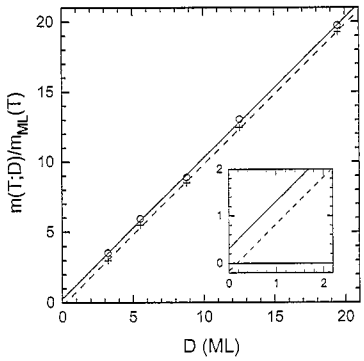

Fig. 4. Magnetic moment $m(T;D)$, normalized by the monolayer moment at given temperature, $m_{ML}(T)$, versus D. Data for $T = 285$ K (crosses and broken line), and for $T = 0$, from extrapolation (b) (open circles and full line), respectively.

which is not the case for fit (a). The two fits are presented in Figs. 2 and 3. The normalized temperature dependences, $m(T;D)/m^{(a/b)}(0;D)$ are shown in Fig. 3(a,b), respectively. In Fig. 2, we included ground state extrapolation values $m^{(a)}(0;D)$ (full points and dotted line) and $m^{(b)}(0;D)$ (open circles and full line), in addition to the experimental values $m(285\text{ K};D)$. For fit (b), which we prefer, the slope of $m^{(b)}(0;D)$ is determined as 1.254×10^{14} V s m. As expected, the ratios of the slopes for $T = 0$ and 285 K, $1.221/1.254 = 0.974$, respectively, nicely equal the ratio of the bulk magnetization values, 2.158 T/2.219 T = 0.969, within the error limits.

Introducing normalized values by dividing through the moment of a bulk monolayer $m_{ML}(T)$, Eq. (1) can be rewritten as

$$\{m(T;D/m_{ML}(T))\} = D + \{\Delta m(T)/m_{ML}(T)\}, \tag{5}$$

and by plotting the normalized magnetic moment $\{m(T;D/m_{ML}(T)\}$ versus D, which is done in Fig. 4. The figure clearly reflects our remarkable result that $\{\Delta m(T)/m_{ML}(T)\}$ is actually independent of D. This independence is quite natural for the (ground state) surface contribution, $\{\Delta m_{surf}/m_{ML}(0)\} = \{\Delta m(0)/m_{ML}(0)\}$, which is the main subject of our work. However, $\Delta m(T)$ is the sum of this temperature independent surface contribution Δm_{surf}, and a temperature-dependent size contribution $\Delta m_{size}(T;D)$ which reflects the enhanced thermal decrease in m in thin films and of course in principle could also depend on D. The constant value of $\{\Delta m(T)/m_{ML}(T)\}$ definitely means that this is not the case, and that

$$\{\Delta m_{size}(T)/m_{ML}(T)\} = \{\Delta m(T)/m_{ML}(T) - \{\Delta m_{surf}/m_{ML}(0)\} \tag{6}$$

also has a constant value. In other words, this means that the size effect reduction of the magnetic moment forms a common correction for all ultrathin films of our series, with a value which is independent of D. Note that $m_{ML}(T)$, the bulk monolayer moment, shows a minor variation of only a few percent for temperatures below 300 K. The independence of D therefore also applies to the absolute value of $\Delta m_{size}(T) = \Delta m(T) - \Delta m(0)$.

The interpretation of this independence of the size effect $\Delta m_{size}(T)$ on D is different for small and for large D, respectively. For small D (and low temperatures), it can be explained as being the result of the two-dimensional nature of thermally excitable magnetic modes, e.g. spin wave states. Stated in these terms, the spectrum of excitable spin wave states is then independent of D, and so is the total number of magnons, and consequently so is the moment reduction caused by them. For detailed discussions of this mechanism, see Refs. [20,21,23]. The experimental evidence discussed in Ref. [22] shows that this explanation holds, for $T \leq 300$ K, for $D \leq 5$, roughly. For thick films, on the other hand, that $\Delta m_{size}(T)$ is independent of D follows from a different reasoning. In thick films, $\Delta m_{size}(T)$ is independent of D because it is composed of contributions from both, virtually independent, surfaces. It remains to be explained why $\Delta m_{size}(T)$ shows the same values in the two regimes.

We note that the observation of a constant value for $\Delta m_{size}(T)$ is equivalent to the previous results on the spin-wave parameters b in Ag-covered Fe(110) films on W(110) [24]. It was observed that the mean values of b, equivalent to parameters b_D in Eq. (4b), depend on D according to

$$b_D = b_\infty + b_S/D, \tag{7}$$

where $b_\infty = 5.3 \times 10^{-6}$ K$^{-3/2}$ is the bulk spin-wave parameter of Fe, and $b_1 = b_\infty + b_S = 56 \times 10^{-6}$ K$^{-3/2}$ is that of the Ag-covered monolayer. It is easily shown that inserting Eq. (7) into Eq. (4b)

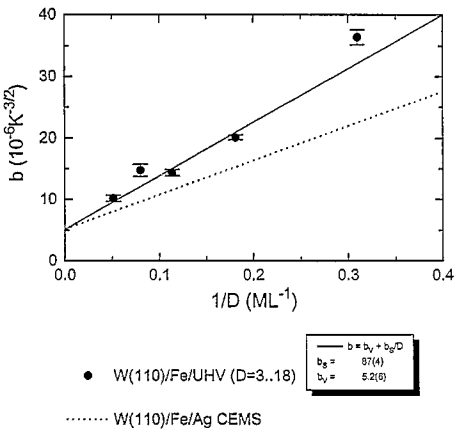

Fig. 5. Spin wave parameters b versus $1/D$. Experimental points and full line from the present work, for uncovered Fe(110) films on W(110); dotted line for Ag-covered films from Ref. [24], for comparison.

results in $\{\Delta m_{\mathrm{size}}(T)/m_{\mathrm{ML}}(T)\} = -b_S T^{3/2}$, which actually is independent of D. The b-parameters of our present series of uncovered films also follow Eq. (7), to a reasonable approximation, as shown in Fig. 5. Uncovered and Ag-covered films differ in the monolayer parameter b_1, given by 86×10^{-6} and 56×10^{-6} $\mathrm{K}^{-3/2}$, respectively. They differ also in their monolayer Curie temperatures $T_{\mathrm{C,mono}}$, which are 225 K [25] and 282 K [26], respectively. It would be reasonable if the thermal decrease in m were a function of T/T_{C}. From Eq. (3b) we argue that this is equivalent to a common value of $b_1(T_{\mathrm{C,mono}})^{3/2}$. We actually do obtain nearly equal values of $b_1(T_{\mathrm{C,mono}})^{3/2} = 0.29$ and 0.27 for uncovered and Ag-covered films, respectively. This is a nice confirmation of our picture.

We now come to the determination of the surface effect. We obtain from Fig. 2 different values $\{\Delta m^{(\mathrm{a})}(0)/m_{\mathrm{ML}}(0)\} = 0.38(17)$ and $\{\Delta m^{(\mathrm{b})}(0)/m_{\mathrm{ML}}(0)\} = 0.29(15))$ for the fits of Eqs. (4a) and (4b), respectively. With better confidence in the latter value, we obtain $\{\Delta m(0)/m_{\mathrm{ML}}(0)\} = 0.32(15)$ as a confidence value for the size effect of both surfaces in common. Finally, we use the result of Pasyuk [12], $\{\Delta m_{\mathrm{surf}}^{\mathrm{W/Fe}}/m_{\mathrm{ML}}(0)\} = -0.07(4)$, to conclude using Eq. (2), to obtain our final result for the surface enhancement of the magnetic moment in the free Fe(110) surface, $\{\Delta m_{\mathrm{surf}}^{\mathrm{W/Fe}}/m_{\mathrm{ML}}(0)\} = 0.39(16)$.

4. Conclusions

We have shown that magnetometry in situ in UHV of ultrathin Fe(110) films on W(110) with variable temperatures is able to determine the surface enhancement of magnetic moment in a free Fe(110) surface. Even with measurements using a moderate temperature range of only 140 K $\leq T \leq$ 285 K, it is possible to obtain a good estimate of the combined enhancement in both interfaces of the films. Combining values from Pasyuk et al. for the moment reduction in the W/Fe interface, we obtain for the free Fe(110) surface a net moment enhancement of 39(16)% of a bulk monolayer moment, or 0.86 μ_{B} per surface atom. This is in agreement with the previous determination of 38(5)% from SPLEED and a theoretically calculated value of 30%.

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