

Temperature dependence of the hyperfine field and magnetization in ultrathin epitaxial Fe films

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Conversion-electron Mössbauer spectroscopy and superconducting quantum interference device magnetometry have been used to measure the magnetic hyperfine field and the spontaneous magnetization of ultrathin Fe(110) films grown epitaxially on Ag(111) substrate films as a function of temperature. For the first time, hyperfine-field data with monolayer resolution and magnetization data are obtained from the same sample consisting of four Fe(110) atomic layers. A comparison of the present results to previous experiments and theories leads to the following conclusions: (1) the ground-state hyperfine field at the Fe(110)/Ag(111) interface is enhanced relative to bulk in agreement with other experiments, but in disagreement with recent band calculations; this discrepancy may be explained by considering dipolar field contributions at the interface; (2) the relative spin deviation from saturation ($T=0$) measured by the hyperfine field is the same as measured by the spontaneous magnetization for the interface atoms as well as for the center of the film; (3) the spin deviation, $\Delta M(T)$, is well described by a $T^{3/2}$ law in all layers of an ultrathin Fe film; hence, a linear M - T relation is not a characteristic feature of a two-dimensional ferromagnet, as often quoted in the literature; (4) ultrathin Fe films show pronounced surface and size effects of the thermally excited spin deviation; no theory exists at present which includes layer-dependent ground-state moments, surface and bulk spin waves; and (5) the spin-wave parameter, B , differs in different experiments for similar film systems. It is suggested that the structure and roughness of the interface in real films play an important role.

I. INTRODUCTION

Considerable progress has been made recently in the evaluation of magnetic moments in thin layers of ferromagnetic $3d$ metals by self-consistent band calculations from first principles (see, for example, Refs. 1–4). They provide local spin densities, magnetic moments per atom, and magnetic hyperfine fields^{1,2} for individual atomic layers in a thin slab. This is of great interest for the question of dimensionality effects in itinerant ferromagnets which have been discussed for about three decades. The most unexpected result of such calculations is an enhanced ground-state magnetization at the surface of metallic ferromagnets like Ni and Fe.

Several experiments have been carried out in order to test these predictions; however, a part of them were performed at room temperature. The comparison of such experimental results to ground-state theories therefore requires an extrapolation of the magnetization to $T=0$. This is only possible if the temperature dependence of the magnetization $M_S(T)$ is known for a thin film geometry.

Various attempts have been made to calculate the magnetization of a film layer by layer as a function of temperature, most of them based on a model of localized magnetic moments. Only recently finite-temperature calculations for magnetic surfaces in an itinerant electron picture have become available.⁵

The most efficient way to experimentally test the predictions of existing theories is to measure the magnetization for individual atomic layers close to a surface or interface. Up to now Mössbauer spectroscopy is the only method which provides information of this kind by

means of the magnetic hyperfine field using a depth profiling technique with monoatomic probe layers or ultrathin films. In this paper we discuss Mössbauer data of ultrathin epitaxial Fe(110) films and compare them to earlier magnetization measurements on the same samples. Thickness and structure of the films have been chosen so as to closely match the model film assumed in recent ground-state band calculations.¹

II. EXPERIMENT

For the present study a system of epitaxially grown Fe(110) films on Ag(111) was chosen because of several attractive properties: (i) The small misfit ($<1\%$) guarantees good epitaxial growth and the absence of a high density of misfit dislocations; (ii) the relatively close-packed (110) surface of Fe does not show surface reconstruction which might complicate the interpretation of observed interface properties; (iii) solubility and diffusion of Fe in Ag and vice versa are extremely small at temperatures below 450 K. Thus atomically sharp interfaces can be expected if the appropriate growth conditions are chosen.

The films were prepared in UHV ($p < 5 \times 10^{-9}$ mbar) by thermal evaporation (Fe enriched to >92 at. % in ⁵⁷Fe). Details of the procedure and the various methods used to characterize the films were given in a previous paper.⁶ Magnetization measurements were done by a vibrating-sample magnetometer and a superconducting quantum interference device (SQUID) magnetometer ($2 \leq T \leq 400$ K; $H \leq 5$ T). Mössbauer spectra of the films were measured by conversion electron Mössbauer spectroscopy (CEMS) with a variable temperature cryogenic

proportional counter in the temperature range between 80 and 350 K. The detection limit is about $\frac{1}{10}$ of a monolayer of ^{57}Fe with 20 h of data acquisition. Details of the design and operation of this counter are reported elsewhere.⁷

III. RESULTS

In an earlier paper⁶ the magnetization of ultrathin Fe(110) films sandwiched between Ag(111) films was measured as a function of applied field and temperature. The data combined with Mössbauer spectra of the films clearly showed that films thicker than three atomic layers are essentially flat and continuous over large distances ($> 1000 \text{ \AA}$) while films of two monolayers average thickness or less consist of islands smaller than 20 \AA diameter which are superparamagnetic at room temperature. The temperature dependence of the spontaneous magnetization $M_S(T)$ is nonlinear for all continuous films and can be fitted by Bloch's spin-wave expression

$$\frac{\Delta M(T)}{M_0} = \frac{M_0 - M_S(T)}{M_0} = BT^{3/2} + \dots \quad (1)$$

Higher terms can be neglected except for high-accuracy measurements which are not easily accomplished on ultrathin films. Values of B obtained from these fits are discussed in Sec. IV B.

A four-atomic-layer Fe film was then chosen for a more detailed study, first because it is expected to show pronounced dimensional effects while still being continuous, second because in principle there are only two non-equivalent lattice sites while the structure is still very close to the five-monolayer-model film calculated by Ohnishi *et al.*¹

Figure 1 shows Mössbauer spectra of the four-layer film at room temperature and at 80 K. The asymmetric line broadening indicates the presence of more than one Zeeman sextet. The spectra are analyzed by assuming a structure model as sketched in Fig. 2: a 4 ML (= monolayer) Fe(110) film covered by 3 ML of Ag(111) at either

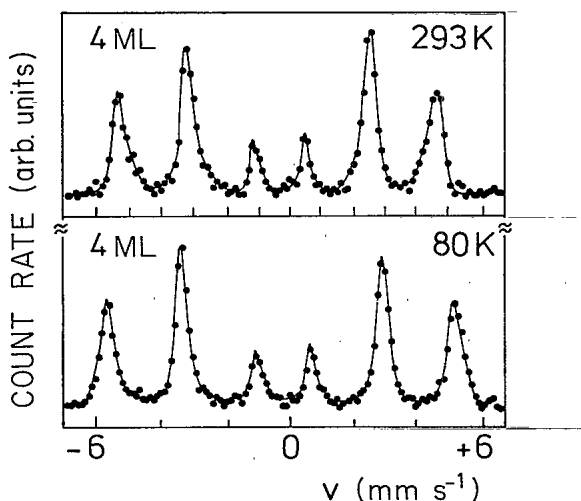


FIG. 1. Conversion electron Mössbauer spectra of a 4-ML (8 \AA) Fe(110) film on Ag(111); solid lines represent fits based on a structure model according to Fig. 2 (see text).

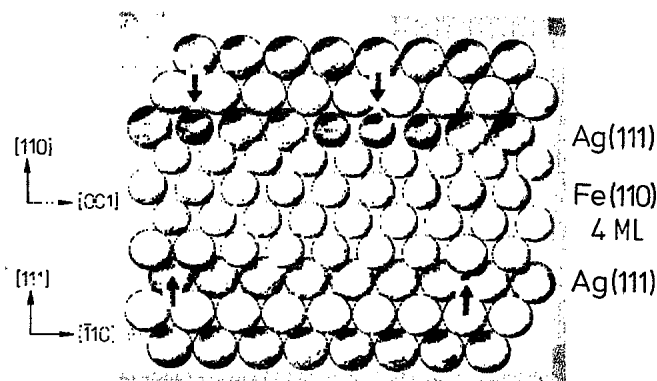


FIG. 2. Structure model of 4-ML Fe(110) film between Ag(111); cross section along the film normal parallel to a $(\bar{1}10)$ plane of the Fe film and a (211) plane of the adjacent Ag layers; we distinguish two central Fe layers and two interface layers with finite roughness. Small circles represent Fe atoms, larger circles Ag atoms. The arrows point to Fe atoms in a Ag layer (\downarrow) or vice versa (\uparrow).

side is shown in a cross section perpendicular to the film plane. This section is parallel to a $\text{Fe}(\bar{1}10)$ plane and a (211) plane of the Ag fcc lattice. Fe and Ag atoms are represented by circles with diameters equal to the nearest-neighbor distance in the respective bulk lattices. The shaded atoms refer to positions in a layer behind the Fe layer shown in the cross section. The arrows indicate an imperfect layer growth: The arrows at the top point to Fe atoms incorporated in the first Ag layer; the arrows at the bottom refer to Ag atoms in the Fe interface layer.

Based on this structure model the spectra were fitted by two sextets corresponding to the interface and the central layers, respectively. This unfolding procedure is justified by the following observations.

(a) The intensity of the interface spectrum is equivalent to 2 ML of Fe and is independent of film thickness; the spectral intensity of the inner layer component decreases linearly with total film thickness t and vanishes at $t=2 \text{ ML}$. This has been shown in a previous paper.⁸

(b) For the 4-ML Fe film discussed in the structure model, we obtain equal intensities of both subspectra at all temperatures covered by the experiment.

(c) The linewidth of the interface component is always larger than for the inner layers. This reflects the larger variety of local atomic configurations within the interface layer as shown in the structure model of Fig. 2.

As a consequence, the fitting procedure provides us the hyperfine parameters of the interface and the central portion of the 4-ML Fe film. In Fig. 3 the magnetic hyperfine fields of the interface layers and the central layers $H_{\text{eff}}^{\text{in}}$ and $H_{\text{eff}}^{\text{c}}$, respectively, are plotted as a function of temperature. For comparison, $H_{\text{eff}}(T)$ for bulk $\alpha\text{-Fe}$ is shown as well. The solid lines represent fits to the experimental data points using an expression analogous to Eq. (1). It yields the spin wave parameter B as well as the hyperfine field at $T=0$, $H_{\text{eff}}(0)$.

In addition, the intensity ratio of the Mössbauer lines 1 and 2 is 3:4 for all partial spectra indicating that the magnetization is oriented parallel to the film plane for all individual layers. This observation together with the value of the saturation field perpendicular to the film plane,

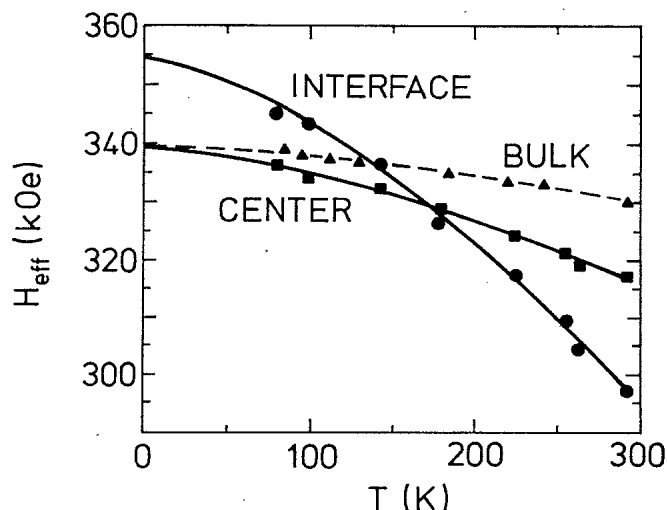


FIG. 3. Magnetic hyperfine field $H_{\text{eff}}(T)$ as a function of temperature for interface (●) and center layers (■) of a 4-ML Fe(110) film between Ag(111); $H_{\text{eff}}(T)$ for bulk α -Fe is shown for comparison; line fits according to $T^{3/2}$ law.

$H_{\text{sat}} = 2.1 \text{ T} \approx 4\pi M_S$, for the four-layer film⁶ shows that no surface or interface anisotropy is present in our films.

IV. DISCUSSION

From Fig. 3 we conclude that the hyperfine field within the experimental error can be described by a $T^{3/2}$ spin-wave law in the interface layer as well as in the interior of the film. It is known⁹ that the hyperfine field $H_{\text{eff}}(T)$ is not exactly proportional to the spontaneous magnetization $M_S(T)$ even in bulk Fe due to lattice vibrations and the thermal lattice expansion. However, the deviation from proportionality does not exceed 0.5% between $T=0$ and 300 K.⁹ This is within the uncertainty of our monolayer hyperfine-field values ($\pm 1\%$) and may therefore be neglected in the present discussion.

The fit according to

$$H_{\text{eff}}^i(T) = H_{\text{eff}}^i(0)(1 - B^i T^{3/2}) \quad (2)$$

yields the ground-state hyperfine field $H_{\text{eff}}^i(0)$ for the i th layer and the spin-wave parameter B^i ($i=1,2$). They will be discussed in the following sections.

A. Ground-state hyperfine field

The magnetic hyperfine field at the Fe nucleus is not simply proportional to the magnetic moment of the Fe atom. This is evident from the hyperfine field in Fe alloys¹⁰ and also from *ab initio* band calculations for thin

slabs either with free surfaces or covered with Ag.¹ Although the calculations were made for a (100) orientation it is nevertheless interesting to compare them to the present experiment and two previous studies of the same Fe(110)/Ag interface. Table I shows experimental and calculated data.

All the experiments agree that $H_{\text{eff}}(0)$ in the Fe layer next to the interface with Ag is clearly enhanced compared to the bulk value [$H_{\text{eff}}(0) = 33.9 \text{ T}$] in contrast to the calculation which yields a slight reduction of the hyperfine field. Possible reasons for this discrepancy have been discussed by Ohnishi *et al.*¹

(i) *Different orientation.* The calculation refers to a (100) orientation of the film while in the experiments (110) films were investigated. Recently, Fu and Freeman¹³ have calculated a nine-layer Fe slab in (110) orientation with free surfaces. As a result, the surface effects at the more close-packed (110) surface are less pronounced than at the open (100) surface, i.e., the enhancement of the magnetic surface moment is weaker [$\mu_S(110) = 2.65\mu_B$ compared to $\mu_S(100) = 2.98\mu_B$] as is the reduction of the surface hyperfine field [$H_{\text{eff}}(110) = 32.4 \text{ T}$ compared to $H_{\text{eff}}(100) = 25.2 \text{ T}$ for a seven-layer slab]. In analogy, we might expect the magnetic moments and the contact hyperfine field at the Fe(110)/Ag interface to lie between the value for the Fe(100)/Ag interface and bulk (33.9 T). Even this would mean a slight reduction of $H_{\text{eff}}(0)$ in the outermost Fe layer in contrast to the experimental findings.

(ii) *Residual finite-size effect.* In the calculation this effect was suspected by Ohnishi *et al.* to hamper the comparison of their results to the experiments with surfaces of "quasibulk" samples (21 to 50 atomic layers^{11,12}). This argument, however, would not be valid for our experiment where the chosen structure is in relatively close match to the one used in the calculation (four and five atomic layers of Fe, respectively, between Ag). Both arguments given by Ohnishi *et al.* to explain the discrepancy between theory and experiment therefore are not supported by the present study.

In order to find an alternative explanation we recall an aspect already discussed in a previous paper.⁸ The effective magnetic field at the ^{57}Fe nucleus (commonly called the hyperfine field) may be decomposed into different contributions according to

$$H_{\text{eff}} = H_{\text{cp}} + H_{\text{ce}} + H_{\text{dip}} + H_a \quad (3)$$

In the following we will always assume zero applied field ($H_a = 0$). The contact hyperfine field calculated by Ohnishi *et al.*¹ contains the core polarization term H_{cp} and the contribution of conduction electrons H_{ce} , while

TABLE I. Ground-state hyperfine field $H_{\text{eff}}(0)$ at the Fe(110)/Ag interface.

	Calc. ^a	Expt. ^b	Expt. ^c	This work
Sample	5 Fe(100)/Ag	W/21 Fe/Ag	Ag/50 Fe/Ag	Ag/4 Fe/Ag
$H_{\text{eff}}(0)$	33.5 T	(34.9 \pm 0.14) T	(34.94 \pm 0.06) T	(35.4 \pm 0.2) T

^aReference 1.

^bReference 11.

^cReference 12.

dipolar fields H_{dip} are not considered. The total dipolar field, in turn, at a given lattice site results from the superposition of the dipolar fields of all the magnetic moments of the sample. In a localized moment picture this field is calculated using the fictitious Lorentz sphere leading to

$$H_{\text{dip}} = H_d + H_L + H_S, \quad (4)$$

where H_d is the demagnetizing field, $H_L = (4\pi/3)M_S$ is the Lorentz field, and H_S is the dipole sum within the Lorentz sphere. In a cubic material we find $H_S = 0$. Inside a cubic crystal with spherical shape we expect the dipole field to vanish because of $H_L = -H_d = (4\pi/3)M_S$.

Inside a thin film with the magnetization parallel to the plane we find $H_d = 0$ and therefore

$$H_{\text{dip}} = H_L = \frac{4\pi}{3}M_S \quad (5)$$

which amounts to about 0.7 T in a Fe film at low temperature. H_L is directed parallel to the magnetization and hence opposite to the net hyperfine field.

At the surface of a ferromagnet, however, the situation is different: Here H_L is not defined and H_S will not vanish. Therefore the lattice sum has to be carried out explicitly. H_{dip} will necessarily depend on the topology of the surface or interface on an atomic scale and on the orientation of the magnetization relative to the surface.

Christensen and Morup¹⁴ have calculated the dipolar fields at the surface of small α -Fe particles and of a Fe(100) film. If the magnetization is parallel to the plane the value of $H_{\text{dip}} = 0.7$ T in the interior of the film is reduced to 0.53 T in the surface layer. However, for Fe atoms sitting on top of the surface layer H_{dip} is further reduced to about 0.15 T. As a consequence, the magnitude of the observed surface hyperfine field should be enhanced by an amount between 0.17 and 0.55 T compared to the bulk value of 33.9 T depending on the amplitude and wavelength of the surface roughness.

This means that the difference between theoretical and experimental values of ≥ 1.4 T cannot be explained by the presence of dipolar fields alone. It is possible, however, that the roughness of the interface also leads to an enhancement of the magnetic moment and, hence, of the hyperfine field. This can be expected from the general result of dimensionality studies for itinerant ferromagnets: The magnetic ground-state moment per Fe atom increases with decreasing coordination from $2.2\mu_B$ in bulk to $2.95\mu_B$ at the free surface, further to $3.2\mu_B$ for a free monolayer of Fe, to $3.3\mu_B$ in a linear chain, and to $4\mu_B$ in the free Fe atom. In practice, a certain interface roughening on a monolayer scale cannot be avoided; it

will create Fe atoms with a reduced number of Fe nearest neighbors and enhanced magnetic moments. A band calculation for Fe interfaces taking into account the roughness of the interface would be very useful in order to understand the difference in calculated and measured ground-state hyperfine fields.

Finally, we have to discuss some differences between our results and two previous experimental studies.^{11,12} (i) The surface effect, i.e., enhanced H_{eff} at the surface, is more pronounced in our measurement relative to the other authors (see Table I). (ii) In our experiment H_{eff} of the second layer is equal to the bulk value, while in the other studies a quasilinear transition from the surface value to the bulk hyperfine field over a distance of four atomic layers is observed.

Both observations could in principle be explained by assuming a certain interdiffusion of ⁵⁷Fe and ⁵⁶Fe in the probe-layer experiments of Droste *et al.*¹² and Korecki *et al.*¹¹ which naturally cannot occur in our samples. That interdiffusion of this kind may take place on the scale of a few atomic layers under specific experimental conditions has been shown by Przybylski *et al.*¹⁵ Furthermore, the ⁵⁷Fe probe layers were two- to three-atomic-layers thick in the work of Droste *et al.*,¹² and for this reason, even atomically sharp transitions should be smeared out over such a distance.

The second important difference between the present work and the previous ones is the total thickness of the ferromagnetic films under investigation (4 compared to 21 or 50 ML). A comparison of different thin slabs calculated by Ohnishi *et al.*¹ shows that size and interference effects are present in slabs of one to seven layers which show up in pronounced H_{eff} oscillations from layer to layer. Therefore more detailed experiments with three to seven Fe layers between Ag and comparable calculations are required in order to fully understand the behavior of the magnetic hyperfine field at the Fe/Ag interface.

B. Temperature dependence of the magnetic hyperfine field $H_{\text{eff}}(T)$ and of magnetization $M_S(T)$

The magnetic hyperfine field $H_{\text{eff}}(T)$ was fitted with a $T^{3/2}$ law according to Eq. (1). The spin-wave parameters B obtained for the interface and the inner layer of a 4-ML Fe(110) film between Ag(111) and for bulk α -Fe are listed in Table II together with B values from magnetization data $M_S(T)$.

Layer-specific values of the spin deviation can only be obtained by decomposing the Mössbauer spectra into difference subspectra. The magnetization data, on the other hand, yield a value averaged over all the layers.

TABLE II. Spin-wave parameter B from hyperfine-field and magnetization data for a four-layer Fe(110) film on Ag(111) and bulk α -Fe. (Data between $T=0$ and $T=300$ K.)

	Bulk α -Fe	Interface layer	Center layer	Average of interface and center layer
$B(K^{-3/2})$ from H_{eff}	$(0.53 \pm 0.02) \times 10^{-5}$	$(3.2 \pm 0.2) \times 10^{-5}$	$(1.3 \pm 0.1) \times 10^{-5}$	$(2.25 \pm 0.2) \times 10^{-5}$
$B(K^{-3/2})$ from M_S	$(0.50 \pm 0.01) \times 10^{-5}$			$(2.0 \pm 0.15) \times 10^{-5}$

Next we compare the B value for our 4-ML film from magnetization data to the value from hyperfine-field data averaged over both layer positions (last column of Table II). We find a good agreement considering the limited accuracy of the film-thickness determination ($t = 8 \pm 0.8 \text{ \AA}$) upon which the model structure of Fig. 2 is based. This means that the normalized effective field at the ^{57}Fe nucleus $H_{\text{eff}}(T)/H_{\text{eff}}(0)$ is to a good approximation proportional to the relative spontaneous magnetization $M_S(T)/M_S(0)$ at the surface as well as in the center of an α -Fe film. This result (which should be checked in future experiments by a variation and precise determination of film thickness) is very important because it has been used on an intuitive basis in the interpretation of hyperfine-field data by various authors.^{16,17}

In order to compare the B values of the four-layer-Fe film with theoretical predictions and with experimental data reported by different authors we have to distinguish between two effects: (i) a size effect due to the reduced extension of the system in one direction of space; this means that the average spin-wave parameter B of a thin film is expected to differ from the bulk value; and (ii) a surface effect due to the reduced magnetic coordination close to the Fe/Ag interface; this means that B may be different for nonequivalent atomic layers or, at a given temperature, the magnetization should be different in different atomic layers.

First we want to compare our results to previous experiments. Magnetization measurements on ultrathin Ni-Fe films by Gradmann¹⁸ gave a stronger increase of the spin deviation with increasing temperature than those in the present study (both normalized to the bulk Curie temperature of the material), e.g., for a four-layer film the spin deviation is approximately twice as large as in our Fe film at $T = 0.3 T_C$. This may be due to the different material (spin-quantum number, coordination, exchange coupling, T_C) or different film structure.

Looking for quantitative experiments on thermal excitations of magnetic interfaces we only find the Mössbauer studies on epitaxial Fe films by Walker and coworkers^{16,19} and by Korecki *et al.*¹¹ In both cases $H_{\text{eff}}(T)$ at the Fe/Ag interface was studied for a relatively thick Fe film (52 and 21 atomic layers of Fe, respectively). The values of the spin-wave parameter B at the Fe(110)/Ag interface are found to be 3.5 (Refs. 16 and 19) or 2 to 2.3 times¹¹ the bulk value.

Besides Mössbauer spectroscopy several techniques have provided information about surface magnetic order. Most of them are based on spin polarization of electrons emitted from the surface via the photoelectric effect, electron capture by fast ions,²⁰ or spin-polarized low-energy electron diffraction (SPLEED).²¹ Results of relevance for the actual discussion have been obtained by Pierce *et al.*²² in a SPLEED experiment. At the surface of a $\text{Ni}_{0.4}\text{Fe}_{0.4}\text{B}_{0.2}$ glass they found an enhanced spin deviation with a lower limit of $B^{\text{surf}}/B^{\text{bulk}} \approx 3$. Other investigations do not include the low temperature (i.e., spin-wave) range ($T \leq 0.5 T_C$) (Ref. 20) or due to limited accuracy can only give qualitative evidence of an enhanced spin deviation at the surface.²³ A general limitation of these methods with respect to layer-specific spin-wave pa-

rameters is the finite escape depth of the electrons which in general will produce average values over several atomic layers (perhaps with the exception of electron-capture spectroscopy).

Before we attempt an interpretation of the different experimental results in comparison to the present study we will summarize some theoretical predictions concerning the question of magnetic excitations at low temperature in thin films and surfaces. Size effects in ferromagnetic systems as well as surface effects have been treated in various theories. Spin-wave theory has been used to calculate magnetic size effects, i.e., the magnetization $M_S(T)$ as a function of film thickness using a Heisenberg model.²⁴⁻²⁸ Surface effects of magnetic excitations have been treated for a semi-infinite Heisenberg type²⁹⁻³³ or Ising³⁴ ferromagnet. Only recently surface spin waves have been calculated in an itinerant electron model.^{35,36,5}

The simultaneous presence of size and surface effects has been considered in the Heisenberg spin-wave calculation by Levy and Motchane^{28,37} and in the molecular field calculation by Valenta.^{38,39} However, both calculations assume a homogeneous ground state which is not realistic in the light of modern band calculations.¹⁻⁴ In this respect the approach of Hasegawa⁵ using a functional-integral method in an itinerant-electron theory is more appropriate because it uses a ground state which is correct according to present knowledge. Unfortunately, Hasegawa's calculation was done for a semi-infinite Fe crystal³⁶ and for Ni films (1-10 ML) sandwiched between Cu⁵ and, therefore, is not easily compared to our Fe films because of the opposite interface effect on the ground-state magnetic moment in the Ni/Cu system. Also, this theory is not likely to give very accurate low-temperature data of $M_S(T)$ because it neglects spin-wave excitations and anisotropies. Nevertheless, improved itinerant electron theories for the actual problem should give valuable data to be compared to our experiment.

To summarize, no calculation is available currently which gives the temperature dependence of the magnetization in an ultrathin Fe film for each individual layer including a realistic ground state as well as spin-wave excitations with bulk and surface modes. Hence, a comparison of our experimental data to theory can only be qualitative.

Our data of Table II show that the total spin deviation, $\Delta M(T) = M_0 B T^{3/2}$, in a four-layer Fe film is by a factor of 4 larger than in bulk α -Fe:

$$\Delta M_4 = 4 \Delta M_\infty .$$

In comparison, many thin-film spin-wave calculations overestimate the size effect in ferromagnetic films: For a four-layer film Döring²⁶ found $\Delta M \approx 6 \Delta M_\infty$, Corciovei²⁷ gave a 14-fold, Glass and Klein²⁵ an 18-fold increase of the spin deviation compared to the bulk value.

It has been shown recently by Yafet *et al.*⁴⁰ that the inclusion of the dipolar interaction in the calculation of spin-wave excitations in very thin films leads to a considerable reduction of the spin deviation and to a stabilization of the ferromagnetic order in the monolayer. The numerical results, however, apply to a hexagonal lattice and, therefore, may not directly be compared to our ex-

periment. Nevertheless, it will be important to consider dipolar interactions in improved theories of magnetic excitations in thin films together with a realistic interface structure.

In a second group of theoretical papers the surface magnetization $M_S(T)$ of a semi-infinite ferromagnet has been calculated by various methods. As first pointed out by Rado²⁹ and later confirmed by Mills and Maradudin³⁰ a more extensive calculation the surface magnetization of a Heisenberg system according to spin-wave theory should have the same $T^{3/2}$ temperature dependence as the bulk but with a spin deviation twice as large:

$$\frac{\Delta M^{\text{surf}}}{\Delta M^{\text{bulk}}} = \frac{B^{\text{surf}}}{B^{\text{bulk}}} = 2.$$

Hasegawa^{5,36} obtained a similar result for an itinerant electron ferromagnet; $\Delta M^{\text{surf}}/\Delta M^{\text{bulk}}$ varies with temperature from 2 to 3 between $T=0.2$ and $0.6 T_C$. Due to the pronounced size effect in our four-layer film its surface behavior may not directly be compared to these surface calculations.

V. SUMMARY

The temperature dependence of the spontaneous magnetization $M_S(T)$ and the magnetic hyperfine field $H_{\text{eff}}(T)$ have been studied in ultrathin epitaxial Fe films. The high sensitivity of the CEMS technique using a cryogenic electron proportional counter allows to determine $H_{\text{eff}}(T)$ for each individual atomic layer of a four-layer-Fe (110) film.

In addition, the magnetization and the layer hyperfine fields were determined on the same sample. In this way a source of error due to limited reproducibility in film preparation was avoided and the relation between magnetization and hyperfine field could be directly checked. The main results may be summarized as follows.

In contrast to a common opinion widely accepted until recently, the spin deviation in two-dimensional ferromagnets at low temperatures $\Delta M(T) = M_0 - M_S(T)$ is not necessarily a linear function of temperature but to a good approximation follows a $T^{3/2}$ law at the bulk surface and in the different layers of ultrathin Fe films. This conclusion which was already deduced from an earlier study⁸ has recently been confirmed by a Mössbauer study on monolayer Fe films.¹⁷ The question of which conditions can produce a quasilinear T dependence of the magnetization at surfaces and in thin films has been treated by several theoretical approaches. Levy *et al.*³⁷ have demonstrated that only the presence of a surface anisotropy can change the nature of the M_S-T relation while an altered surface exchange only changes the coefficient of the $T^{3/2}$ term. Mathon and Ahmad,⁴¹ however, found that an extreme softening of the surface exchange might also lead to a quasilinear $M_S(T)$.

Recently, Rado^{42,43} pointed out that assuming particular values of the surface anisotropy constants K_S and K_{SS} a quasilinear contribution to the spin deviation at the surface can be expected due to surface spin waves which might be dominant under certain experimental conditions. It remains to be explained how such surface anisotropies might be connected with the microscopic structure of a real-film system, the orientation and roughness of the interface, and the topology of the film (e.g., island formation).

The relative spin deviation measured by the hyperfine field $\Delta H_{\text{eff}}(T)/H_{\text{eff}}(0)$ is within the experimental error of $\pm 1\%$ equal to $\Delta M_S(T)/M_0$ at the surface as well as in the central part of an Fe film. This had been assumed intuitively in previous publications^{16,17} but had not been experimentally confirmed before. The spin-wave parameter B averaged over the entire film volume increases with decreasing film thickness in qualitative but not in quantitative agreement with earlier thin-film spin-wave calculations.

At the Fe(110)/Ag(111) interface we observe an enhancement of the ⁵⁷Fe ground-state hyperfine field of about 4% compared to bulk α -Fe. This is in clear disagreement with recent calculations of the contact hyperfine field.¹ To elucidate the origin of this discrepancy the roughness of the interface and the associated magnetic moment changes and dipolar fields have to be included in future calculations.

The spin deviation shows a pronounced surface effect which is superimposed on the size effect of our four-layer film. Actually, no satisfactory theory is available to treat combined surface and size effects in ultrathin ferromagnetic films. It is hoped that further experimental work on films consisting of several atomic layers may stimulate corresponding theoretical studies.

Observed surface spin deviations are markedly different in different experiments. This may be understood when taking into account a result of theoretical investigations³³ which predict a strong influence of the structure, the orientation, and structural perturbations of the surface on the density of states of surface magnons. In a recent theoretical work Mathon and Ahmad⁴¹ using an effective Heisenberg Hamiltonian for a metallic ferromagnet found that a softening of the exchange interaction between adjacent layers at the surface produces a crossover from a low-temperature B value of $2B_{\text{bulk}}$ according to Mills and Maradudin³⁰ to a second larger region with an effective $T^{3/2}$ law and a surface B value $B_{\text{surf}} > 2B_{\text{bulk}}$ which now depends strongly on the particular surface exchange. Hence, further experiments with well-defined interface structures and theoretical investigations will have to elucidate the connection between the microstructure of real interfaces, the exchange and anisotropy at the interface, and the resulting thermal spin excitations.

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