Temperature dependence of the magnetization in Fe islands on W(110): evidence for spin-wave quantization

V Senz\textsuperscript{1,3}, R Röhlsberger\textsuperscript{1}, J Bansmann\textsuperscript{1}, O Leupold\textsuperscript{2} and K-H Meiwes-Broer\textsuperscript{1}

\textsuperscript{1} Universität Rostock, Fachbereich Physik, Universitätsplatz 3, 18051 Rostock, Germany
\textsuperscript{2} European Synchrotron Radiation Facility, BP 220, 38043 Grenoble Cedex, France
E-mail: volkmar.senz@physik.uni-rostock.de

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\textbf{Abstract.} We have measured the temperature dependence of the magnetic hyperfine field in Ag-coated Fe(110) islands on W(110) between 4 and 300 K using nuclear resonant scattering of synchrotron radiation. The decay of the spontaneous magnetization of the islands with increasing temperature differs distinctly from the bulk characteristics and is not described by a simple Bloch’s $T^{3/2}$ law. The deviation is attributed to quantization of spin-waves as a result of geometric confinement in the islands. The data can be explained assuming an effective energy gap in the spin-wave spectrum of $\Delta E = 6.7 \pm 1$ meV.

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\textsuperscript{3} Author to whom any correspondence should be addressed.
1. Introduction

During the past two decades, Fe(110) films on W(110) have been used as a model system for magnetism of two-dimensional systems like monolayer films and islands. This can be attributed, on the one hand, to the formation of a stable pseudomorphic iron monolayer and, on the other, to a rich variety of magnetic phenomena due to the interplay of bulk, surface and shape anisotropies. Magnetic ordering in monolayer films of Fe(110) on W(110) has been studied by Mössbauer spectroscopy [1]–[4], spin-polarized low-energy electron diffraction [5]–[7] and magnetometry [8]. In addition, the iron on tungsten system has played a key role with regard to understanding the spin dynamics in thin films. For example, Hillebrands et al found an unexpected increase of the spin-wave frequencies for Fe film thicknesses decreasing below 60 Å. This could be related to strong magnetic anisotropies—a contribution that has not been observed in previous experiments [9, 10].

In this paper we investigate the temperature dependence of the spontaneous magnetization in 3D Fe(110) islands on W(110). The decay of the magnetization at low temperatures is determined by magnetic excitations of two types: one-electron excitations from the majority spin band to an empty state in the minority spin band are referred to as Stoner excitations. In contrast, spin-waves or magnons as collective excitations correspond to phase modulations of the precessing spin moments. Since at low temperatures the density of states of magnons is much larger than that of Stoner excitations, the temperature characteristics of the magnetization is, in any case, completely dominated by the spin-wave spectrum [11].

We have probed the magnetism of the sample by investigating the hyperfine splitting of the $^{57}$Fe nuclear levels. The experimental technique used here is nuclear resonant scattering of synchrotron radiation. The method is similar to classical Mössbauer spectroscopy in the sense that the recoilless excitation of nuclear levels is involved. However, there are significant differences of this method compared to the Mössbauer absorption technique, resulting in qualitatively new features. First, synchrotron radiation has properties complementary to a radioactive source because it is continuous in energy and exhibits a pulsed time structure. This suggests performing hyperfine spectroscopy on a timescale rather than on an energy scale: the simultaneous excitation of the hyperfine-split nuclear energy levels by an energetically broad pulse of synchrotron radiation leads to characteristic quantum beats in the temporal evolution of the subsequent nuclear decay. The analysis of this beat pattern allows a precise determination of the magnitude and the orientation of magnetic hyperfine fields in the sample [12]. Second, the measured signal results from coherent scattering events rather than from incoherent absorption processes. Therefore, relative phase factors between different nuclear resonances have to be taken into account. This allows for the identification of different spin arrangements that cannot be distinguished by classical Mössbauer spectroscopy. It should also be noted that this method is a probe of magnetism even in cases where the magnetic moment of the sample is zero, because it is sensitive to the magnetic field at the sites of the nuclei. Therefore, as a microscopic probe, it provides complementary information to methods like magneto-optical spectroscopy or x-ray dichroism. Thus, the use of synchrotron radiation offers significant advantages compared to the classical Mössbauer absorption technique. Due to the extremely high brilliance of the undulator synchrotron radiation at third generation facilities it provides unique possibilities to probe magnetic properties with sub-monolayer sensitivity [13, 14].

In the context of our study it is important to point out that the measured magnetic hyperfine field $B_{hf}$ is to a very good approximation proportional to the local magnetic moment, because $B_{hf}$
dominantly arises from the local core polarization of the 4s electrons and the orbital moment of
the 3d electrons [15]. While this relationship has been long established for bulk Fe [16], it was
also confirmed more recently in Fe thin films down to monolayer coverage [8, 17]. Therefore, the
temperature dependence of the magnetic moment can be directly inferred from the temperature
dependence of the magnetic hyperfine field.

2. Experimental details

The iron islands were prepared by thermal evaporation of a five monolayer (ML) $^{57}$Fe film (95% enriched) from a crucible under ultra-high vacuum conditions onto a clean single crystal of
W(110) and subsequent thermal treatment. Due to the large mismatch of the Fe and W lattices,
annealing at temperatures between about 500 and 1000 K leads to lattice relaxation and the
formation of separated 3D islands on top of one pseudomorphically ordered Fe monolayer on
W(110) [18, 19]. The transition from the closed film to the island structure can be directly
followed by means of low-energy electron diffraction (LEED): satellite spots characteristic for
lattice distortions in the closed film [20] vanish, while diffraction spots of the relaxed Fe(110)
diffuse close to the W spots.

The shape of these islands, depending crucially on the thermal treatment, was controlled by
scanning tunnelling microscopy (STM), as shown in figure 1. The Fe islands are of bcc (110)

Figure 1. STM image of Fe islands on W(110) created by annealing a 5 ML Fe
film at 700 K. The islands are preferentially aligned along the W[001] direction.
The lower part shows a cross section of a typical island along the distance indicated
by the white arrow.
type with the lattice constant of bulk iron as revealed from x-ray diffraction [21]. In our case the sample has been annealed at 700 K for 10 min, which results in the formation of elongated, rectangular shaped islands pointing with the long axis along the W[001] direction. The islands are about 200 nm wide and 200–1000 nm long. The mean island height of this particular sample has been determined to be 3.7 ± 0.8 nm. A typical cross section is shown in figure 1. After preparation the sample was coated with a few MLs of Ag in order to prevent contamination in the subsequent ex situ scattering experiment. Previously, the sample was magnetized along the in-plane [001] direction using a pulsed magnetic field of about 50 mT magnitude.

The experiment was carried out at the nuclear-resonance beamline ID18 of the ESRF (Grenoble, France) [22]. To facilitate time-resolved measurements, the storage ring was operated in 16-bunch mode, providing a time interval of 176 ns between subsequent bunches. The radiation was monochromatized by a high-resolution monochromator to an energy bandwidth of 6.5 meV around the 14.4 keV resonance of $^{57}$Fe. This ensured that the detector was not overloaded by the huge intensity of non-resonant photons. The sample was illuminated in grazing incidence geometry at an angle of 5 mrad, the critical angle of total reflection of the W substrate at 14.4 keV. It was mounted in a cryostat where the temperature could be adjusted between 4.5 and 300 K. The in-plane [001] direction, i.e. the bulk easy axis of magnetization, was aligned perpendicular to the incident wavevector. No external magnetic field was applied. In this geometry the maximum count rate from the sample is about 40 s$^{-1}$ at an average electron beam current of 70 mA in the storage ring. Under these conditions the typical data acquisition time for a time spectrum of nuclear resonant scattering from the sample is 0.5 h.

3. Results and discussion

A number of selected time spectra taken at various temperatures are shown in figure 2. The frequency of the temporal oscillations is a characteristic measure of the splitting of the nuclear levels in the ground and excited states that is induced by the magnetic hyperfine interaction. With increasing magnetic hyperfine field the temporal beat frequency increases linearly, leading to a shift in the beat maxima towards slightly earlier times. Due to the very good statistical quality of the data, the value of the hyperfine field can be determined with an accuracy of about 1%. The intensities of the oscillations are determined by the orientation of the magnetic fields at the nuclei relative to the wavevector of the radiation. This dependence is due to the M1 multipole character of the 14.4 keV transition of the $^{57}$Fe nucleus and the polarization of the synchrotron radiation. We have determined the orientational distribution of the magnetic fields in a separate experiment, resulting in two components oriented along the in-plane [001] and [1\bar{1}0] directions, with relative weights of 0.8 and 0.2 respectively [21]. The distribution of magnetization components was interpreted in that study to be the result of the interplay between the magnetic anisotropy contributions—magneto-crystalline, surface and shape—resulting in a thickness- and shape-dependent reorientation of the magnetization from the [1\bar{1}0] direction (the easy axis of thin Fe films on W(110)) to the [001] direction (the easy axis in bulk iron). It is worth noting, that although the capping layer is known to change the surface anisotropy [2], no difference was detectable between Ag- and C-coverage in that study. From this we are confident that the magnetic order really reflects the island geometry and is not the result of a particular capping layer. The solid curves presented in figure 2 are fits according to the dynamical theory of nuclear resonant scattering [23], extended to the case of grazing incidence reflection [24]. No indication was found that the orientation of the magnetic fields in the islands changed as
Figure 2. Selected set of time spectra of nuclear resonant scattering from Ag-capped $^{57}$Fe islands as a function of temperature. Solid curves show the result of simulations assuming two magnetic hyperfine fields of equal magnitude in the in-plane [001] and [110] directions with relative contributions of 0.8 and 0.2 respectively [21]. The change of the beat frequency, as emphasized by the dotted line, reveals the decreasing magnetic hyperfine field with increasing temperature. The values of the magnetic hyperfine field as derived from these spectra are shown in figure 3.

a function of temperature. Thus, the magnitude of the magnetic hyperfine field was the only parameter that was varied to fit the data.

The resulting temperature dependence of the magnetic hyperfine field is shown in figure 3. In contrast to the case of bulk iron the magnetization is constant within the experimental error for temperatures below 50 K. Whereas the bulk hyperfine field data, taken from [25], are well described by Bloch’s $T^{3/2}$ law, this is not the case for the iron islands. Here we most probably have to take into account the influence of the geometric confinement on the spin-wave spectrum, which will be elaborated on in the following.

Spin-waves in cubic crystals are most easily excited perpendicularly to the most dense lattice planes, i.e. the (111) planes in fcc lattices and the (110) planes in bcc lattices [26]. We assume that the spin-wave energy $E$ depends on the square of the wavenumber $q$, i.e.,

$$E = Dq^2,$$  (1)
Figure 3. Temperature dependence of the magnetic hyperfine field $B_{hf}$ in the Fe(110) islands as determined from the evaluation of the data in figure 2 (red dots). The solid red curve is a fit to the data according to equation (6), assuming a spin-wave stiffness of $D = 126$ meV Å$^2$ and an energy gap of $\Delta E = 6.7$ meV. The dotted red curve is the best fit to the data according to Bloch’s law disregarding the energy gap (equation (3)). For comparison the bcc Fe bulk data are shown (green diamonds), taken from [25] and fitted with the $T^{3/2}$ law without energy gap and $D = 272$ meV Å$^2$ (green curve), in agreement with experimental results [27, 28] and recent ab initio calculations [11].

where $D$ denotes the spin-wave stiffness parameter and amounts to about 280 meV Å$^2$ for bcc bulk iron [11, 27, 28]. The quadratic approximation is well justified for bulk iron, where no optical spin-wave branch evolves and the occupation factor for the high-energy spin-waves is relatively small, particularly at low temperatures [29]–[31]. As the excitation of one magnon lowers the total spin by about $\hbar$, the decrease in the magnetization is given by the number of excited magnons at a certain temperature $T$ [26]:

$$M(T) = M(0) - \int P(E) n(E) \, dE,$$

where $P(E) = (V_0/4\pi^2)D^{-3/2}\sqrt{E}$ is the density of spin-wave states, and $n(E)$ is the Bose occupation number. In the case of a continuous distribution of spin-wave states as in the bulk, this equation leads to the temperature dependence given by Bloch’s law [32], i.e.,

$$M(T) = M(0)(1 - BT^{3/2}) \quad \text{with} \quad B = 2.6149 \, V_0 \left(\frac{k_B}{4\pi D}\right)^{3/2},$$

where $V_0$ is the atomic volume and $B = 3.7 \times 10^{-6}$ K$^{-3/2}$. In contrast, in a confined space with linear dimension $d$ only discrete wavevectors are possible where the smallest value is $q = 2\pi/d$. The first excited level of the quantized spin-wave spectrum is then given by [26, 34]

$$\Delta E = D\left(\frac{\pi}{d}\right)^2.$$
In this case, essentially the Bose occupation number has to be rewritten as

\[ n(E) = \frac{1}{\exp \frac{E + \Delta E}{k_B T} - 1}. \] (5)

In order to solve the integral, the upper limit is extended to infinity:

\[
\int_0^\infty P(E) n(E) \, dE = \frac{V_0}{4\pi^2} \left( \frac{1}{D} \right)^{3/2} \int_0^\infty \frac{\sqrt{E}}{e^{(E + \Delta E)/k_B T} - 1} \, dE
= \frac{V_0}{4\pi^2} \left( \frac{k_B T}{D} \right)^{3/2} \sum_{k=1}^{\infty} e^{-k\Delta} \int_0^{\infty} e^{-kx} \sqrt{x} \, dx
= \frac{V_0}{4\pi} \left( \frac{k_B T}{D} \right)^{3/2} \sum_{k=1}^{\infty} \frac{e^{-k\Delta}}{k^{3/2}}, \] (6)

where \( x = E/k_B T, \Delta = \Delta E/k_B T \) and \( \int_0^{\infty} e^{-kx} \sqrt{x} \, dx = \sqrt{\pi}/(2k^{3/2}) \) (see e.g. [33]). The series sum has to be evaluated and ranges as a function of temperature from 1 \( \cdot e^{-\Delta} \) for \( \Delta E \gg k_B T \) (see [34]) to 2.6149 \( \cdot e^{-\Delta} \approx 2.6149 \) for the opposite limit with \( \Delta E \ll k_B T \), corresponding to Bloch’s law without an energy gap (equation (3)).

We have applied equation (6) to fit the temperature dependence of the magnetization, shown in figure 3, and obtained values of \( D = 126 \pm 10 \) meV Å\(^2\) and a gap in the spin-wave excitation spectrum of \( \Delta E = 6.7 \pm 1 \) meV. Inserting these values into equation (4) we find that a linear dimension of \( d = 1.4 \) nm reproduces the measured energy gap. Within our simple model this is in the range of the mean island height of 3.7 ± 0.8 nm, as shown in the STM line scan given in figure 1.

Obviously, the finite size of the islands has significant consequences for the temperature dependence of the magnetization. At low temperatures the magnetization of the islands decays slower with temperature than the magnetization in the bulk does. This is a consequence of the quantization of the spin-wave spectrum. At elevated temperatures the softening of the spin-wave eigenstates becomes dominant and the spontaneous magnetization of the islands decays faster with temperature than the bulk magnetization [26].

Because we attribute the spin-wave quantization to the vertical confinement in the islands it is worthwhile comparing these results with the spin dynamics of closed iron films on tungsten, which has been investigated in detail by means of Brillouin light scattering [9, 10, 35]. Although this scattering technique is restricted to low energy transfers and the main focus of these studies has been on the so-called Damon–Eshbach mode, a long-wavelength dipolar excitation, also the first standing spin-wave could be detected at 37.3 GHz (0.154 meV) for the case of a 170 Å film [35]. With decreasing film thickness the frequency of this mode rises, which is then in reasonable agreement with our values for the mean island height and the energy gap.

However, the quantization due to lateral confinement, as observed recently in nickel nanowires and permalloy wires and dots [36]–[38], would lead in our case to an energy gap of about 0.4 µeV (0.1 GHz) assuming an island width of 200 nm. This might be already discernible by means of Brillouin scattering, but would be far too small to influence the temperature dependence of the magnetization in a sizeable way.

We therefore conclude that the characteristic temperature dependence of the magnetization is the result of the spin-wave quantization induced by vertical confinement within the iron islands. We admit that it is difficult to derive details of the spin dynamics such as quantization effects measuring an integral quantity like the temperature dependence of the spontaneous
magnetization. Further theoretical studies, as carried out for the iron ML and bilayer [39], might be helpful in distinguishing the influence of the reduced coordination on the temperature characteristics via (i) the enhancement of the local magnetic moments, (ii) the energetically more favourable thermal disorder and hence reduced Curie temperature and (iii) the advent of quantization of spin-wave excitations. In addition, it will be of particular interest to extend these experiments to clusters on surfaces, where due to the confinement in three dimensions these effects are much more pronounced.

We would like to emphasize that the nuclear resonant method of scattering of synchrotron radiation bears the inherent advantage of isotopic sensitivity. The signal originates only from the Mössbauer nuclei in the sample and is free from contributions originating from surrounding magnetic species. It is thus possible to employ ultrathin isotopic probe layers to study magnetic properties of thin films with very high spatial resolution [40]. This can be applied, for example, to investigate the spin-wave density of states near interfaces, that plays an important role in mediating the exchange coupling between magnetic thin films. Such studies have been performed in the past using conversion electron Mössbauer spectroscopy [41]. These experiments, however, are time consuming even with the use of strong radioactive sources. On the other hand, the outstanding collimation and intensity of modern synchrotron radiation sources has reduced data acquisition times by orders of magnitude into the range of a few minutes. With the method used here it is now possible to study magnetic excitations in low-dimensional systems that were hardly possible before.

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