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## Magnetisation in Two-Dimensions

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### Abstract

We discuss the magnetisation deviation in two-dimensions (2d) for an iron film for which the anisotropy is much smaller and hence the magnetisation is a stronger function of temperature. The magnetisation deviation can be adequately described by an RPA (random phase approximation) approach. Our results fit the experimental data at high fields and hence can be used to estimate the value of the effective exchange interaction ( $J_{2d}$ ) for monolayer (ML) films.

### Introduction

It is well known that in two dimensions (2d) a system possessing a continuous symmetry does not exhibit a conventional long-range order meaning that the expectation value of the order parameter vanishes at all finite temperatures, provided that the interaction between the spins is isotropic and that the interaction is short ranged [1].

In epitaxial structures, neither the magnetostatic dipole-dipole interaction nor magnetic anisotropies can be avoided, which break the continuous symmetry of the Heisenberg Hamiltonian. The direct result of this symmetry breaking is the occurrence of long-range order below a well-defined temperature  $T_c$ .

However for a film with a uniaxial anisotropy favouring a perpendicular easy magnetisation axis, long-range order can exist at finite temperatures since the anisotropy gives rise in this case to a significant spin-wave gap [2,3], but no such gap arises the case in which in-plane orientation of the magnetisation is favoured.

In our earlier work [4] we presented the results of a combined experimental and theoretical study of the finite temperature magnetisation of ultrathin epitaxial films and showed that the experimentally observed stabilisation of the magnetisation of ultrathin Co films with planar anisotropy is due to the in-plane magnetocrystalline anisotropy and Zeeman energy rather than the dipole interactions alone.

Lugert et al [6] have recently measured the sample moment  $M(T)$  of an iron film in a constant applied field as a function of temperature. They have grown ultrathin epitaxial

Fe (110) single films by MBE (molecular beam epitaxy) on Au (111). Ferromagnetic order was observed down to the thinnest films of 0.5 monolayer (ML) average thickness. In their experiments the effective magnetic anisotropy with respect to the film normal (surface anisotropy minus the demagnetizing effects) shows a strong thickness dependence: below 3ML the easy axis switches from in plane to perpendicular, this is the consequence of a strong magnetic interface anisotropy dominating the dipolar anisotropy at small thickness.

In this work, we investigate the magnetisation deviation which can be adequately described by the RPA approach to estimate the value of the effective exchange interaction in the monolayer film. We also calculate the numbers of monolayers, N.

### 1. Calculation of Magnetisation

We assume that the magnetisation deviation is determined by the excitation of spin waves and may be adequately described by an RPA (random phase approximation) approach which gives a set of bosons whose frequency is a function of T. This approach also becomes exact at low temperatures.

We consider an RPA calculation for a  $s = 1/2$  model (Tyablicov theory) [7]. The magnetisation  $\langle s^z \rangle$  is given in terms of averages of spin ladder operators.

$$\langle s^z \rangle = \frac{1}{2} - \langle s_n^- s_n^+ \rangle = \frac{1}{2} - \frac{1}{N} \sum_{\underline{k}} \langle s_{\underline{k}}^- s_{\underline{k}}^+ \rangle. \tag{1}$$

The correlation function is found from the two-time Green function which has been calculated using an RPA coupling,

$$\langle s_n^- s_n^+ \rangle \simeq \frac{2 \langle s^z \rangle}{e^{\beta \varepsilon_k} - 1}, \tag{2}$$

where  $\varepsilon_k$  is the spin-wave energy and  $\beta = 1/kT$ . Substituting Equation (2) in Equation (1), we get

$$\langle s^z \rangle = \frac{1}{2N \sum_{\underline{k}} \coth \frac{\beta \varepsilon_k}{2}}. \tag{3}$$

Here in the Tyablicov decoupling  $\varepsilon_k$  is a function of the magnetisation, i.e.,  $\varepsilon_k = \frac{D_o}{S} \langle s^z \rangle k^2$  and  $D(T) = D_o \langle s^z \rangle / s$  (a crude approximation over estimated renormalization of  $\varepsilon_k$ ). Hence,

$$\begin{aligned} \sum_{\underline{k}} \coth \frac{\beta \varepsilon_k}{2} &= \frac{A}{2\pi} \int_0^{\pi/a} dk k \coth \frac{\beta \varepsilon_k}{2} \\ &= \frac{Ak_B T}{2\pi D_o \langle s^z \rangle} \left[ \ln \sinh \frac{\beta \varepsilon_{max}}{2} - \ln \sinh \frac{\beta \varepsilon_o}{2} \right]. \end{aligned} \tag{4}$$

By considering our conditions in which the limits are the energy gap  $\varepsilon_o$  and the value of  $\varepsilon_k$  on the zone boundary  $\varepsilon_{max}$ ,  $\ln \sinh \frac{\beta\varepsilon_{max}}{2} \gg 1$  and  $\ln \sinh \frac{\beta\varepsilon_o}{2} \ll 1$  we can write

$$\sum_{\underline{k}} \coth \frac{\beta\varepsilon_k}{2} \simeq C - \frac{Ak_B T}{4\pi D_o \langle s^z \rangle} \ln \frac{\varepsilon_o}{k_B T}$$

where  $C \simeq 1$ . In two dimensions the integrals can be calculated as given in [4,5]

$$\langle s^z \rangle = \frac{1}{2} - \frac{\langle s^z \rangle a^2 k_B T}{4\pi N D_o \langle s^z \rangle} \ln \left( \frac{k_B T}{\varepsilon_g} \right), \quad (5)$$

where  $D_o = 2Jsa^2$ . The  $\langle s^z \rangle$  which appears in the denominator arises because the RPA approximation predicts that the spin wave stiffness is linearly proportional to  $\langle s^z \rangle$ :

$$\langle s^z \rangle = \frac{1}{2} \left( 1 - \frac{a^2}{2\pi D_o} k_B T \ln \left( \frac{k_B T}{\varepsilon_g} \right) \right). \quad (6)$$

We can write this in the form

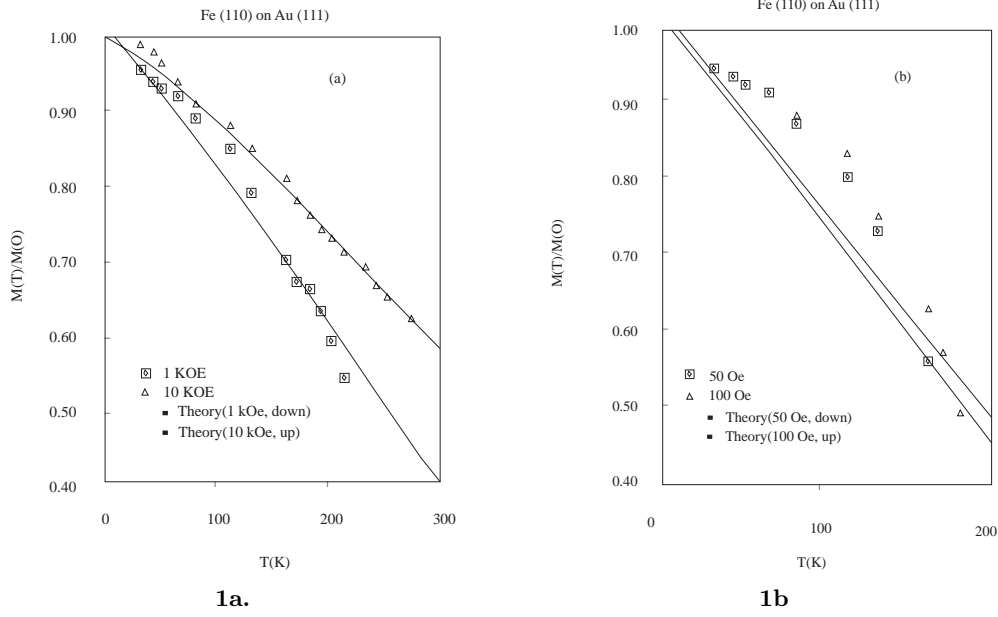
$$\frac{M(T)}{M_o} = 1 - \frac{T}{T_e} \ln \left( \frac{T}{T_g} \right), \quad (7)$$

where the exchange temperature  $T_e$  is determined by the atomic spin and the strength of the exchange interaction and is given by  $T_e = 8\pi S\mu_B H_{ex}/k_B$ , and  $T_g$  is the gap temperature [4].  $H_{ex}$  is the effective field and is given by  $H_{ex} = D/(g\mu_B a^2)$ , where  $D$  is the spin wave stiffness for the propagation of spin waves in the film and  $a$  is the lattice parameter [4]. Here, the RPA approximation is equivalent to the non-interacting spin wave theory, which is special to 2d. Equation (7) was obtained by cancelation of  $\langle s^z \rangle$  in the numerator and denominator of Equation (5). In a magnetic field

$$k_B T_g = \varepsilon_{anis} + g\mu_B H_{anis}, \quad (8)$$

where  $\varepsilon_{anis}$  and  $H_{anis}$  are the in-plane anisotropy energy and the anisotropic field, respectively. The anisotropy energy is expected to vary with the magnetisation.

We find that we can fit Lugert et al's [6] data very satisfactorily at high fields (for  $H=10$  kOe) to Equation (7) by neglecting the in-plane anisotropy,  $\varepsilon_{anis}$ . This is shown in Figure 1a. For low temperatures and large  $M$  the error should not be severe (the linear spin wave theory is exact as  $T \rightarrow 0$ ) so we fit the experimental data in the region  $\frac{1}{2} < \frac{M}{M_o} < 1$  only.



**Figure 1.** The temperature dependence of spontaneous magnetisation: (a) 1 kOe and 10 kOe; (b) 50 Oe and 100 Oe (in this case the theoretical and experimental points agree only if anisotropy is as shown in figure 2).

However, at lower fields such as 1 kOe, 100 Oe and 50 Oe, we have a poor fit without anisotropy as illustrated in Figures 1a and 1b. Obviously, the data are different at lower fields. By using Equation (7), in which a value of  $\sim 3800$  K is used for  $T_e$ , we have one parameter per point for fitting procedure to find the  $H_{anis}$  fields. This is shown in Figure 2. We have assumed an error of 0.01 in the experimental value of  $(M_o - M)/M_o$  and used this to deduce approximate error bars in this Figure. Where  $(M_o - M)/M_o$  is small the theory is very insensitive to the value of  $\varepsilon_{anis}$  (which appears in a logarithm) and hence the error bars are very large. We see that within the errors we find that our assumption that  $H_{anis}$  depends only on  $M$  and is independent of the external field is shown to be reasonable. As seen from this Figure, the anisotropy field is less than or on the order of 3 kOe. This confirms our earlier assumption that the results at 10 kOe can be fitted without including anisotropy.

We now consider only the 10 kOe data. From the satisfactory nature of this fit (see Figure 1a, for  $H=10$  kOe) we conclude that our theory is reasonable and therefore we can use the value  $(3800 \pm 200K)$  of  $T_e$  to deduce the exchange interaction in the monolayer film. This is a very interesting result because it is the first time that this quantity has been measured directly.

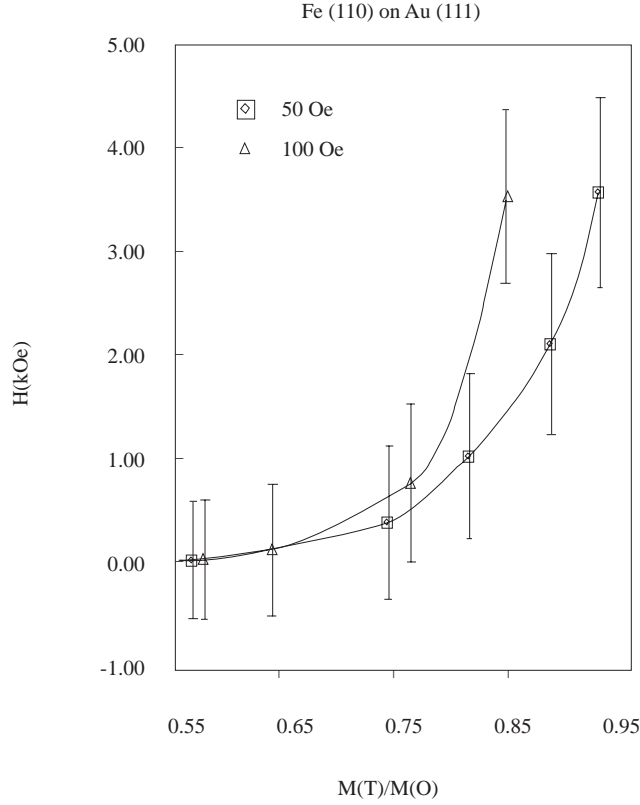
In three dimensions (3d) the magnetisation varies as

$$\frac{M(T)}{M(o)} = 1 - AT^{3/2}, \quad (9)$$

where the constant  $A$  has the experimental value  $(3.4 \pm 0.2) \times 10^{-6} \text{ deg}^{-3/2}$  for Fe in 3d [9]. Equation (9) can be written in the form:

$$\frac{\Delta M(T)}{M(0)} = \frac{0.0587}{SQ} \left( \frac{k_B T}{2JS} \right)^{3/2}, \quad (10)$$

where  $Q = 1, 2, 4$  for sc, bcc, fcc lattices, respectively [9].



**Figure 2.** The anisotropic field as a function of magnetisation deviation.

We compare the value of  $J_{2d}$  in plane with the value for 3d by evaluating both  $T_e$  and  $A$  in terms of  $J$ . By using Equation (10), we find the effective exchange interaction in three dimensions as  $(2JS)/k_B = J_{3d} \simeq 700K$  for Fe. In two dimensions, the magnetisation varies as

$$\frac{\Delta M(T)}{M(0)} = \frac{T}{T_e} \ln\left(\frac{T}{T_g}\right), \quad (11)$$

where  $T_e$  and  $T_g$  were determined in Reference [4]. From Equation (11) we find  $(2JS)/k_B = J_{2d} \simeq 300K$  and thus

$$\frac{J_{2d}}{J_{3d}} \simeq 0.42. \quad (12)$$

This was evaluated assuming that there is one cell of Fe (volume  $a^3$  containing 2 Fe atoms) in the layer. If in fact there is only a square array of area  $a^2$  containing one Fe atom then the result is doubled, i.e.  $\frac{J_{2d}}{J_{3d}} \simeq 0.84$ .

A few words are in order about the validity of the RPA result given by Equations (1) and (2) in 2d. It works relatively well in 3d where it predicts  $T_c$  which is much better agreement with experiment than mean field theory. However, in 2d the result is much more suspect. The first peculiarity is that, the magnetisation deviation depends only on the spin wave energy gap and is the same for easy axis and easy plane (with in plane anisotropy) with the same energy gap. This is suspect since we know that as the anisotropy goes to zero in the easy axis case, we go to a Heisenberg model with  $T_c = 0$  whereas as the in-plane anisotropy vanishes in the case of easy plane the system develops a Kosterlitz-Thouless phase [8]. Such subtleties are lost in the crude RPA approximation.

However there is an even worse consequence of Equation (5). Consider the situation in which there is an external field  $H$ , there is a finite energy gap given by,

$$k_B T_g = g\mu_B H$$

$$\frac{M(T)}{M(0)} = 1 - \frac{T}{T_e} \ln \frac{T}{T_g}. \quad (13)$$

Then the magnetism is zero for  $T > T_c$  where  $T_c$  is given by

$$\frac{T_c}{T_e} \ln \frac{T_c}{T_g} = 1. \quad (14)$$

This means that the theory predicts a zero susceptibility for  $T > T_c$  and demonstrates the inapplicability of the RPA in this temperature region in two-dimensions. The origin of this error is of course the RPA result which predicts that the spin wave stiffness  $D$  vanishes as  $\langle s^z \rangle \rightarrow 0$ .

The spin wave energies were calculated [4,5] for the cases of easy axis and easy plane in which the Hamiltonian contains exchange, cubic anisotropy, uniaxial anisotropy and Zeeman energies. The effect of an external magnetic field is to add a term  $g\mu_B H$  to the coefficient  $A$ . The spin waves are of two types: those in which the  $\underline{k}$  vector lies in the plane which may be of very small  $k$  and excitations across the film containing  $N$  monolayers which correspond to  $k_{\perp} = n\frac{\pi}{N}$ ;  $n=1, \dots, N$ . The film behaves 2-dimensionally

when the only modes which are thermally excited correspond to  $k_{\perp} = 0$ . We calculate the maximum thickness for the films to behave two dimensionally over the temperature range of interest.

We now consider how thin a film should be for it to be correctly described by the 2d theory. We evaluate the largest  $N$  for the 2d theory to be valid up to 300 K. The magnetisation deviation per site will be the sum over the modes for  $k_{\parallel}$  and  $k_{\perp}$ . No significant excitations of spin waves  $k_{\perp}$  film are assumed to occur over the temperature region of interest. From the magnetisation we can estimate the effective exchange for a thin film. We use the 3d value for  $(2JS)/k_B = J_{3d}$  (=700 K for Fe) [9] because it is known that the Curie temperature  $T_c$  increases rapidly with the sample thickness. For a film of  $N$  monolayers the smallest value of  $k_{\perp}$  is  $(\pi/aN)$ . This leads to an excitation energy,  $\varepsilon_{\perp}$ , of:

$$\varepsilon_{\perp} \simeq 2JSa^2 \left(\frac{\pi}{Na}\right)^2 = k_B T_{\perp}. \quad (15)$$

The excitation of this mode will be negligible for temperatures  $T$  such that

$$T \leq \left(\frac{1}{2}\right)T_{\perp}. \quad (16)$$

We demand that for  $T = 300K$  that  $k_B T \leq \frac{1}{2}\varepsilon_{\perp}$ , this gives us the required condition on  $N$  for this to hold for  $T \leq T_c$ . We find that the film is effectively 2d for  $N \leq 3$ .

## 2. Conclusions

We show that our results fit the data at high fields and hence can be used to estimate the value of the effective exchange for monolayer films. It is important to separate the statistical mechanics of a 2d array of spins from the determination of the in-plane exchange. It is only because we have a convincing fit of the theory as a function of both  $T$  and  $H$  (at high fields) that we are able to determine  $J_{2d}$  for the first time.

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