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Spin Dynamics in Low-dimensional Magnetic Structures^{*}

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Abstract

Maxwell's equations applied to low-dimensional magnetic structures result in a number of interesting features. For example, the magnetic fields generated by two-dimensional arrays of Heisenberg spins can stabilise long range magnetic order and determine critical temperatures. Aspects of this problem are discussed, and considerations for the dynamic response of weakly coupled arrays of fine magnetic particles are presented. Finally, a form of effective medium theory designed to overcome difficulties in treating magnetostatic interactions in magnetic nanostructures is described.

1. Introduction

The important interactions responsible for ordering and critical phenomena in magnetic materials are usually considered to be due to strong, but short ranged correlations between electrons. Weak, long range interactions caused by the magnetic fields generated by magnetisation are of course also present, but the corresponding energies are usually negligible in comparison to the short range interaction energies. Critical temperatures and magnetic excitation energies are therefore mainly determined by the short range interactions, and the weak long range dipolar interactions are significant only for long wavelength dynamic behaviour and phenomena related to domain formation.

The situation is quite different in the case of low-dimensional magnetic structures. A well-known example is the two-dimensional Heisenberg magnet, where nearest neighbour interactions are believed to be insufficient to stabilise long range order (Mermin and Wagner 1966), whereas long range order may be possible if dipolar interactions are taken into account (Yafet *et al.* 1986). There are numerous other questions of interest for two-and one-dimensional magnetic structures, and because experimental models of low-dimensional magnetic systems are readily available, there has been extensive work over the years on the theory and measurement of magnetic phenomena in low dimensions. The literature of the field is extensive and no attempt to reference it is made here. Some sources particularly relevant to the present paper include a review of theoretical work on thin ferromagnetic films given by Mills (1991), and an overview of theoretical and experimental studies of critical phenomena in low-dimensional magnetic systems is presented in the book by Collins (1989).

One approach towards understanding the importance of weak, long range dipolar interactions in magnetic systems is through the study of the dynamics associated with the magnetic system. The fundamental excitations of the magnetic system are spin waves. The

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purpose of this paper is to explore the impact of dipolar interactions on spin wave excitations in three different contexts: (1) low and high temperature behaviour of twodimensional ferromagnets; (2) microwave response and switching of arrays of nanometre sized magnetic dots; and (3) infrared response of antiferromagnetic multilayers.

2. Spin Waves in Two Dimensions

The Heisenberg Hamiltonian is a useful model for describing properties of localised moment magnetic structures. The simplest form contains an exchange energy term representing a short range interaction between neighbouring spins:

$$H = \frac{1}{2} \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j .$$
(1)

In this expression, J_{ij} is the exchange interaction between spins located at sites *i* and *j*. The angle brackets on the sum indicate that the sum includes nearest neighbours only.

Linearised spin waves calculated from this Hamiltonian have energies that go as the square of the momentum. If the lattice spacing is *a*, and the spin wave has a plane wave form, $\exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$, where the frequency of the spin wave is ω and the wavevector is \mathbf{k} , then the dispersion relation is $\omega = 2 S J a^2 k^2$ for ferromagnetic coupling (J > 0). The low temperature thermal reduction of the total magnetisation ΔM can be calculated by summing the thermally occupied spin wave states according to

$$\Delta M = \sum_{\mathbf{k}} \left[\exp(\hbar\omega / k_B T) - 1 \right]^{-1} .$$
⁽²⁾

In three dimensions, this gives the well-known Bloch $T^{3/2}$ law. In two dimensions, the k^2 dependence of the energy causes a divergence at k = 0, leading to instability of the ferromagnetic ground state at finite temperatures. Dipolar interactions remove this divergence by modifying the long wavelength dependence of the frequency on k (Yafet *et al.* 1986; Pescia and Pokrovsky 1990; Erickson and Mills 1991). Written as a discrete sum over magnetic dipoles, the dipolar Hamiltonian is

$$H_d = \frac{1}{2} (g \mu_B)^2 \sum_{i,j} \left[\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{r_{ij}^3} - 3 \frac{(\mathbf{r}_{ij} \cdot \mathbf{S}_i)(\mathbf{r}_{ij} \cdot \mathbf{S}_j)}{r_{ij}^5} \right].$$
(3)

The frequency of long wavelength spin waves in an ultra-thin film of thickness d can be shown to be (Stamps and Hillebrands 1991)

$$(\omega/\gamma)^2 = [Dk^2 + 4\pi M f (1 - k d/2)][Dk^2 + 2\pi M f k d \sin^2 \phi] .$$
(4)

Here *M* is the saturation magnetisation and ϕ is an angle specifying the direction of spin wave propagation in the film plane relative to the direction of *M*. The parameter *D* contains the exchange integral *J* between spins at neighbouring lattice sites in addition to geometrical factors. The gyromagnetic ratio is γ , and *f* is a numerical factor obtained by summing the contributions to the magnetic field at a lattice point due to the other point magnetic dipoles at the lattice sites throughout the film. This factor is unity in the limit of a continuous distribution of dipoles, and is less than one if the dipoles are arranged on a

lattice. The terms linear in k are sufficient to remove the divergence in the sum over states of equation (2).

Experimental evidence exists for significantly reduced critical temperatures in twodimensional ferromagnets (see e.g. Gradmann 1984), and an interesting theoretical question is how the dipolar interaction affects high temperature behaviour. Whereas existing work has been largely formulated in terms of scaling theory (Pokrovskii and Feigelman 1977), little has been done using spin wave theory.

An interesting approach is to examine spin wave interactions mediated by the dipole field. This has been attempted for the exchange part of the Hamiltonian (Wang and Mills 1993), but has not been rigorously carried out for the dipolar terms of equation (3). One approach is to expand the spin operators $S^{\pm} = S_x \pm iS_y$ in magnon variables in a perturbation treatment. For S^+ , the lowest order interaction terms are

$$S^{+} = \sqrt{\frac{2S}{N}} \sum_{\mathbf{k}} e^{i\,\mathbf{k}\cdot\mathbf{r}} \mathbf{a}_{\mathbf{k}} - \frac{1}{N\sqrt{8NS}} \sum_{\mathbf{k},\mathbf{k}',\mathbf{k}''} e^{i\,(\mathbf{k}-\mathbf{k}'-\mathbf{k}'')\cdot\mathbf{r}} \mathbf{a}_{\mathbf{k}}^{+} \mathbf{a}_{\mathbf{k}'} \mathbf{a}_{\mathbf{k}''}, \qquad (5)$$

with a corresponding expression for S^- . Here S is the total spin at a lattice site, N is the number of spins in the array, and the $\mathbf{a_k}$ are boson operators for the magnon excitations. The full calculation is rather involved, but the end result is a spin wave frequency that is a function of the total number of spinwaves in the system, $\sum_{\mathbf{k}} n_{\mathbf{k}}$, where $n_{\mathbf{k}}$ is the number of spin waves with momentum \mathbf{k} . The thermal reduction in the magnetisation given by equation (2) must then be solved self-consistently:

$$\sum_{\mathbf{k}} n_{\mathbf{k}} = \sum_{\mathbf{k}} \left\{ \exp[\hbar\omega \left(\sum_{\mathbf{k}} n_{\mathbf{k}}\right) / k_B T] - 1 \right\}.$$
 (6)

This equation has been solved numerically for a two-dimensional array of spins using parameters for M and D appropriate to bulk Fe. Results are shown in Fig. 1 where $M - \Delta M$ is shown as a function of temperature. The magnetisation decreases steadily until about 250 K where the magnetisation suddenly drops toward zero. Physically what happens it that the spin waves create fluctuations in the dipole field. When the magnitude of these fluctuations becomes large, the dipole field acting at a lattice site averages to zero, and the two-dimensional array of ferromagnetically coupled spins is no longer stable to thermal



Fig. 1. Magnetisation of a two-dimensional ferromagnet calculated as a function of temperature. The reduction in the magnetisation is calculated using spin wave theory. Note the critical point at 250 K, due to fluctuations destroying the ability of demagnetising fields to stabilise long range order.

fluctuations. It is interesting to note that the spin wave eigenfunctions are highly elliptical at low temperatures in the sense that $|S_x| \neq |S_y|$. This is due to the demagnetising fields created by the dipolar interactions. As the temperature is increased, fluctuations decrease the magnitude of the demagnetising fields, causing the eigenfunctions to become more circular and $|S_x| = |S_y|$ when the magnetisation becomes unstable.

While the result is a critical temperature significantly lower than that for bulk Fe, in agreement with experiment (Gradmann 1984), one must interpret the above argument with great care. Iron is only approximately represented by a local spin Hamiltonian such as that of equation (1), and there are several other contributions to local effective fields, such as anisotropy (Krams *et al.* 1992), that may dominate the behaviour of the system. Furthermore, for any actual comparison to experiment, it would be necessary to use values for exchange and magnetisation appropriate for a thin film.

3. Weakly Interacting Dot Arrays

The technology to fabricate high quality magnetic wire and dot structures with physical dimensions in the nanometre range has advanced rapidly in the past few years. One potential application of great current interest is for construction of magnetic random access memory devices. As a consequence, relevant magnetic processes involve dynamics of the magnetisation under application of moderately large magnetic fields.

Except in the special case of small amplitude spin wave excitations, magnetisation dynamics are governed by highly nonlinear equations of motion. Some classes of insulating magnetic compounds, such as yttrium iron garnet, have in fact served as model systems for studying nonlinear effects via absorption of high power microwaves (Rezende *et al.* 1986). In such processes, spin wave interactions determine the response, and magnetostatic interactions can be very important.

To date, investigations of these phenomena have only been performed for large, continuous magnetic films, ellipsoids and spheres. An interesting question is how high power absorption occurs in patterned arrays of sub-micron sized magnetic dots. A model system can be examined as follows. Suppose a nanoscale single domain magnetic element is approximated as a giant magnetic moment **m**. The classical equation of motion for a single **m** in a static field h, and an applied microwave field $\mathbf{h}_{\omega} = \mathbf{x} h_{\omega} e^{i\omega} d\mathbf{r}^{t}$, is

$$\frac{d\mathbf{m}}{dt} = \gamma \,\mathbf{m} \times (\mathbf{z} \,h + \mathbf{x} \,k \,m_x + \mathbf{h}_{\omega}) - \alpha \,\mathbf{m} \times \frac{d\,\mathbf{m}}{dt} \,. \tag{7}$$

Here α describes the rate at which the magnetisation dissipates energy. The anisotropy constant *k* represents an effective anisotropy field that describes preferential orientation of **m** with respect to a symmetry axis of the underlying crystal lattice. In equation (7), **x** is normal to the film plane, and the sign of *k* means that **m** has its lowest energy static configuration for orientation in the film plane. The driving field is applied normal to the film plane and oscillates with frequency ω_{dr} .

Dynamics of an array of interacting dots can be described by writing equations of motion of the form in equation (7) for each **m** in the array. The interaction is specified by a dipolar field \mathbf{h}_d added to the other fields in the equations motion. The field \mathbf{h}_d is produced by the other **m** in the array and can be constructed from equation (3) by replacing the spin operators with vector equivalents **m**, and calculating $\mathbf{h}_d = -(\partial/\partial \mathbf{m})H_d$.



Fig. 2. (*a*) Microwave power absorption calculated for different microwave field intensities. The upper panel shows the linear response for low intensities and the lower panel shows the nonlinear response at higher intensities. (*b*) Sketch of the relative amplitudes of the magnetisation in the dot array.

The ferromagnetic resonance mode (FMR) is typically measured in power absorption experiments. This mode corresponds to the situation where all of the **m** precess in phase. The power absorption was calculated by numerically integrating equation (7). An example of the power spectrum is shown in Fig. 2*a* for a 3×3 array of magnetic dots. The upper panel shows the linear response for a small h_{ω} . The large peak contains the FMR mode, but there are contributions from many nearly degenerate modes of the array in both peaks.

As the intensity of the driving field is increased, the modes of the array interact, and the power absorption spectrum acquires more structure. This behaviour is shown in the lower panel where a number of sharp peaks appear. The precession amplitudes associated with the FMR peak frequency are shown in Fig. 2b. The linear response shown in the upper panel is characterised by precession amplitudes that are the same for each \mathbf{m} . The high power response in the lower panel shows localisation to the centre dots of the array. Increasing the intensity of the driving field leads to greater localisation, and can even result in complete reversal of the central \mathbf{m} . A more complete analysis is given in Stamps and Camley (1999).

The process of switching, in which the direction of magnetisation is reversed by applying a magnetic field, involves the dissipation of energy from the spins to the lattice rather than the absorption of energy from a large amplitude field. Even so, the resulting dynamics are highly nonlinear even for isolated, single domain magnetic particles. For switching of a single magnetic moment \mathbf{m} by a constant field \mathbf{h} , the equation of motion is

$$\frac{d\mathbf{m}}{dt} = \gamma \mathbf{m} \times [\mathbf{z} (k m_z - h_z) + \mathbf{x} h_x] - \alpha \mathbf{m} \times \frac{d\mathbf{m}}{dt}.$$
(8)

In equation (8), the choice of $\mathbf{z}k$ for the anisotropy means the system can be thought of as a two-state system with **m** preferentially lying either along the +z or -z directions at equilibrium.

If the reversal process is assumed long compared to the precession time, equation (7) can be averaged over time scales long compared to the precession and short compared to the reversal. The resulting equation can be solved exactly in the case k = 0 with a characteristic reversal time τ_s given by (Stamps and Hillebrands 1999)

$$\tau_s = \frac{1 - h_x / h_z}{\alpha \gamma h_z} \tag{9}$$

in the case $h_x \ll h_z$.

The switching dynamics of an array of dots is strongly affected by dipolar interactions represented by the field \mathbf{h}_d . This was studied by Stamps and Camley (1999), and it was found that the switching time is a sharply peaked function of the magnitude of \mathbf{h}_d . In terms of packing density of the array, this means that fast switching occurs for densely packed arrays, where \mathbf{h}_d is largest, and for large array spacings, where \mathbf{h}_d is small. In between these two limits there is a range of array spacings where the switching rate is slowed significantly due to excitation of dipolar modes in the array.



Fig. 3. Sketch of the easy plane antiferromagnet with out-of-plane fluctuations. Demagnetising fields are due to spins within a two-dimensional plane, so that even the net magnetisation of a unit cell vanishes, the local effective fields acting on each sublattice contain demagnetising effects.

4. Effective Medium Theory

As a final example of how dipolar interactions between magnetic moments are important in determining the dynamics of nanostructured materials, mention is made of a very different class of systems from those considered so far: antiferromagnets. Because the dipole field in Maxwell's equations is generated by a net magnetic moment, it is not too surprising that it can be important in ferromagnetic systems. This is usually not the case in antiferromagnets where there is no net macroscopic magnetic moment, and dipole interactions are often ignorable.

It turns out that dipolar fields are important for understanding the optical response of easy plane antiferromagnets. These materials, such as NiO and EuTe, typically have responses in the far infrared that are governed by the dynamics of the antiferromagnetic sublattices. The easy plane geometry of these systems means that spins of a given sublattice are aligned parallel in two-dimensional sheets. As a result, fluctuations out of plane involve a demagnetising field due to other spins in the *same* two-dimensional sheet, even though the magnetisation of the unit cell remains zero. This idea is shown schematically in Fig. 3 where a unit cell of an easy plane antiferromagnet is depicted. The net out of plane moment associated with an individual sublattice results in a demagnetising field acting on the same sublattice.

The consequences of this idea were explored by calculating response functions for the easy plane structure by including explicit sums over the dipole moments produced by each sublattice spin in a manner similar to the calculation of spin wave frequencies, as described in the Introduction (Stamps *et al.* 1993). The result was compared to an effective medium calculation in which demagnetising effects are included by applying electromagnetic boundary conditions during the calculation of electromagnetic susceptibilities.

The effective medium susceptibilities are defined by the relation

$$\langle \mathbf{m} \rangle = \chi \langle \mathbf{h} \rangle, \tag{10}$$

where

$$\langle \mathbf{m} \rangle = \frac{\mathbf{a} + \mathbf{b}}{2} , \qquad (11)$$

$$\langle \mathbf{h} \rangle = \frac{\mathbf{h}_a + \mathbf{h}_b}{2} \,. \tag{12}$$

The magnetisation of the two sublattices are **a** and **b**, and the corresponding dipole fields acting on each sublattice are \mathbf{h}_a and \mathbf{h}_b . The effective medium approximation is used to relate \mathbf{h}_a and \mathbf{h}_b subject to the requirement that components tangential to the sublattice planes are continuous, while components normal to the planes are discontinuous by an amount proportional to the sublattice magnetisation. This means imposing the relations

$$(\mathbf{h}_a)_{\parallel} = (\mathbf{h}_b)_{\parallel} \quad , \tag{13}$$

$$(\mathbf{h}_{a})_{\perp} + 4 \pi a_{\perp} = (\mathbf{h}_{b})_{\perp} + 4 \pi b_{\perp} , \qquad (14)$$

on the calculation of χ .

The results for χ calculated in this way agree well with the equation of motion approach using dipole sums (Stamps *et al.* 1993). The primary effect is to introduce a term in χ_{zz} that shifts the pole frequency by an amount $4\pi M$, where *M* is the sublattice magnetisation. This represents the effects of local demagnetisation on the individual sublattices. The effective medium method has turned out to be quite useful, and has since been extended to describe accurately the response of thin films (Stamps and Camley 1996).

5. Summary

Three examples have been presented in this paper illustrating some consequences of the macroscopic Maxwell equations for electromagnetism on the dynamics of sub-microscopic magnetic structures. Magnetic properties and the dynamic response of magnetic systems are strongly dependent on dimensionality, and weak, long ranged interactions between magnetic dipoles can be surprisingly important for one- and two-dimensional magnetic structures.

By modifying the long wavelength spin wave energies of two-dimensional ferromagnets, long range magnetic order can exist at finite temperatures. Spin wave interactions mediated by the dipole coupling can destabilise the order and lead to a critical temperature significantly reduced from that of a three-dimensional system.

High power microwave absorption, which is known to produce a variety of interesting nonlinear effects in continuous ferromagnetic films, can also strongly modify the response of arrays of nanoscale magnetic particles. In particular, mode localisations can occur, lifting degeneracies in the magnetostatic mode spectrum and causing large amplitude fluctuations on selected particles.

Finally, the necessity of accounting for demagnetising effects in layered antiferromagnets was discussed. This is an interesting example of how demagnetising fields are formed, in that it is shown how demagnetising effects can appear even when the net magnetisation of the material is zero.

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