Dipole-dipole interactions and two-dimensional magnetism

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The effect of the dipole-dipole interaction on the magnetism of a two-dimensional lattice of spins coupled by isotropic short-range exchange interactions is investigated. In the noninteracting spin-wave approximation it is shown that for positive J the system becomes ferromagnetic.

The temperature dependence of the magnetization of thin magnetic films has recently been of considerable interest. A review of the experimental situation is given in a paper by Bayreuther.[1] Recent experiments[2] on Gd-Y superlattices show a very gradual decrease of the magnetization $M(T)$ with temperature T for Gd arrays only three atomic layers thick but in a magnetic field H of 12.8 kG. In such a field, the behavior of $M(T)$ can be understood to follow simply from the gap introduced by H into the spin-wave spectrum at $k=0$. More generally, however, the understanding of the temperature dependence of the magnetization in nearly two-dimensional systems is difficult because a detailed understanding of the anisotropy is not available and the measurements are made in a magnetic field of the order of kG. In particular, the role of dipole-dipole interactions on two-dimensional magnetism has, to our knowledge, not really been clarified. As is well known, an infinite two-dimensional lattice of spins coupled by isotropic exchange cannot sustain long-range order.[3] To ensure the existence of a magnetization it has therefore been necessary to assume a finite-sized specimen,[4] or to introduce an external field[5] or an anisotropy field.[6] Yet because of its anisotropy and long-range character, the dipole-dipole interaction may itself stabilize long-range order and the effect may be particularly important in Gd which has a large spin $S=\frac{7}{2}$.

Surprisingly, little seems to have been done on this subject: Brodkorb[7] included the dipole-dipole interaction in a Green's-function treatment and concluded that its effect was negligible. In a later paper on thin film magnetism, Navarro and de Jongh[8] replaced the dipole-dipole interaction by an effective anisotropy field. However, it can be seen from the work of Holstein and Primakoff[9] that this is not a valid approximation for calculating the spin-wave spectrum near $k=0$, and this spectrum is what determines whether long-range order can exist.

In this paper we calculate, using simple spin-wave theory, the low-temperature magnetization of a two-dimensional lattice of ferromagnetically coupled spins in the presence of dipole-dipole interactions. We show that the latter remove the divergence of $\Delta M(T)$, the deviation of the magnetization from saturation that results from thermal excitation of spin waves, and hence lead to ferromagnetic ordering. For large S, the calculated $\Delta M(T)$ is smaller than that obtained in Refs. 4 and 5 and is comparable with the experimental result.[2]

An estimate of the relative strength of the dipolar to the exchange energy can be obtained by comparing a typical dipolar energy $E_D=4\pi\beta M_B$ where β is the Bohr magneton and M_B the saturation magnetization, with the stiffness constant α of the spin waves. Excluding the polarization of the conduction band (a 10% effect), the value of (E_D/k_B) is found to be 1.66 K. For nearest-neighbor interactions the exchange integral J is related to the Curie temperature T_C by the relation[10]

$$\frac{k_B T_C}{J}=\tfrac{5}{96}(z-1)[11S(S+1)-1]\,, \tag{1}$$

where z is the number of nearest neighbors. Gd has $T_C=292$ K which gives $(J/k_B)\cong 3$ K, a value less than twice that of E_D. Actually the physically meaningful parameter with which to compare E_D is $\alpha=4SJ$ in terms of which the spin-wave energy for small wave vector k is αK^2. Here the dimensionless quantity K is ka, a being the lattice constant of the hexagonal plane. The ratio (E_D/α) is then $\simeq\frac{1}{25}$, which is not very small. For a spin of $\frac{1}{2}$ and the same T_C, this ratio would have the much smaller value of $\frac{1}{600}$. It is clear then that the effect of the dipolar interactions can be important when S is large.

The formalism for including H_D in the calculation of $m(T)$ at low temperatures has been given in Ref. 9 and all that has to be done is to carry out the same calculation in two dimensions. We follow the notation of Ref. 9 and refer to that paper for the meaning of the symbols. M_0 being the saturation magnetization (magnetic moment per *unit area),* Eq. (23) of Ref. 9 gives

$$\Delta m(T)\equiv\frac{\Delta M(T)}{M_0}$$

$$=\frac{1}{(2\pi)^2 S}\int d\mathbf{K}\,\frac{A(\mathbf{K})}{[A(\mathbf{K})^2-|B(\mathbf{K})|^2]^{1/2}}\times\frac{1}{\exp\left[\dfrac{[A(\mathbf{K})^2-|B(\mathbf{K})|^2]^{1/2}}{k_B T}\right]-1}\,. \tag{2}$$

(As in three dimensions, the deviation of the ground-state magnetization from saturation is negligible.)

The quantities $A(\mathbf{K})$ and $B(\mathbf{K})$ are

$$A(\mathbf{K})=2SJ\sum_{l}[1-\exp(i\mathbf{K}\cdot\mathbf{l})]+2\beta H$$
$$-\frac{4\beta^2S}{a^3}\sum_{l}\frac{1}{l^3}\left[1-\frac{3l_z^2}{l^2}\right]$$
$$-\frac{2\beta^2S}{a^3}\sum_{l}\frac{1}{l^3}\left[1-\frac{3l_z^2}{l^2}\right]\exp(i\mathbf{K}\cdot\mathbf{l}) , \quad (3a)$$

$$B(\mathbf{K})=\frac{6\beta^2S}{a^3}\sum_{l}\frac{l_x^2}{l^5}\exp(i\mathbf{K}\cdot\mathbf{l}) . \quad (3b)$$

Here xz is the plane of the layer, which for simplicity has been taken to be a square net of mesh a, z is the direction of the magnetization and l is a vector whose components are integer l_x,l_y,l_z, the point at the origin being excluded. The external field H will be taken to be zero until later. Note that replacing the dipolar interaction by an effective field is equivalent to leaving out the terms in $\exp(i\mathbf{K}\cdot\mathbf{l})$, which would result in $B(\mathbf{K})=0$ and hence in a gap $2\beta H_{\rm dip}$ in the spin-wave spectrum. Retaining the dynamical character of the interaction gets rid of this gap since, from Eq. (3), for lattices with sufficient symmetry, $A(\mathbf{K})=B(\mathbf{K})$ in the limit $\mathbf{K}=0$. Further examination is therefore needed to determine whether the divergence of $\Delta M(T)$ is removed.

The contribution to $\Delta m(T)$ from small K values, $\Delta m(T)_l$ is obtained by expanding the Bose factor:

$$\Delta m(T)_l=\frac{k_BT}{(2\pi)^2S}\int_0^{2\pi}d\phi\int_0^{q}\frac{A(\mathbf{K})KdK}{A(\mathbf{K})^2-|B(\mathbf{K})|^2} . \quad (4)$$

The choice of the upper limit q will be discussed later. The dependence of the denominator on $\mathbf{K}$ will determine whether $m(T)$ converges. To find this dependence we must calculate the lattice sums in Eq. (3). We do this in two steps. First, we calculate the sums for $\mathbf{K}=0$. Then, by symmetry,

$$\sum_{l}(l_x^2/l^5)=\sum_{l}(l_z^2/l^5)=\tfrac{1}{2}\sum_{l}(1/l^3) .$$

The sum $\sum(1/l^3)$ is just a measure of the local field at a lattice site when the spins point along the normal to the layer. For a thin film (three dimensional) this sum would be equal to the difference between the demagnetizing coefficient 4π and the coefficient of the Lorentz field, $4\pi/3$. for the square net the calculated sum is equal to $(8\pi/3)e$ where the numerical factor $e=1.078$ is close to unity. For the triangular lattice $e=1.318$, the larger value reflecting the close packing of this lattice. Second, for $\mathbf{K}\neq0$, we replace the sum over lattice points by an integral over l which can be done analytically. A cutoff r_0 at the lower limit of l is chosen such as to reproduce the calculated value of the sum at $\mathbf{K}=0$. The validity of this approximation for $\mathbf{K}\neq0$ has been checked numerically and found to be excellent. For the square lattice the sum in $B(\mathbf{K})$ becomes

$$F(\mathbf{K})=\sum\frac{l_x^2}{l^5}\exp(i\mathbf{K}\cdot\mathbf{l})$$
$$\simeq\int_{r_0}^{\infty}\frac{dl}{l^2}\int_0^{2\pi}\sin^2(\theta+\phi)\exp(iKl\cos\phi)d\phi , \quad (5)$$

where ϕ is the angle between l and $\mathbf{K}$ and θ is the angle between $\mathbf{K}$ and the magnetization. Making use of the relation[11]

$$\exp(ix\cos\phi)=J_0(x)+2\sum_{n=1}^{\infty}i^nJ_n(x)\cos(n\phi) ,$$

we obtain

$$F(\mathbf{K})\simeq\pi\int_{r_0}^{\infty}\frac{dl}{l^2}[J_0(Kl)+\cos(2\theta)J_2(Kl)] . \quad (6)$$

Furthermore, noting that[11] $[dJ_0(x)/dx]=-J_1(x)$ and $\int_0^{\infty}[J_1(x)/x]dx=1$, we finally obtain to first order in K,

$$A(\mathbf{K})=\alpha K^2+2\beta H+4\pi\beta M_Bf\left[1-\frac{K}{4f}[1+\cos(2\theta)]\right] , \quad (7a)$$

$$B(\mathbf{K})=4\pi\beta M_Bf\left[1-\frac{K}{4f}[3-\cos(2\theta)]\right] . \quad (7b)$$

For the simple-cubic lattice, the factor $f=e=1.078$, while for the hcp lattice $f=(e/\sqrt{2})=0.932$, and in addition the terms linear in $\mathbf{K}$ acquire a factor $\sqrt{2/3}$. Letting $E_M=4\pi\beta M_B$, we obtain for $H=0$,

$$A(\mathbf{K})^2-|B(\mathbf{K})|^2=\alpha K^2+2E_M$$
$$\times\{1-(K/4f)[1+\cos(2\theta)]\}$$
$$+E_M^2(K/f)[1-\cos(2\theta)] . \quad (8)$$

This expression has a term linear in K which is surprising at first glance since each l term in Eqs. (3) depends on $\mathbf{K}$ through $\cos(\mathbf{K}\cdot\mathbf{l})$ which contains only even powers of K. [The terms in $\sin(\mathbf{K}\cdot\mathbf{l})$ drop out by inversion symmetry.] This linear dependence is due to the long-range character of the dipolar interaction: Eq. (5) was integrated for finite $\mathbf{K}$ but an infinite lattice. If the lattice were taken to be finite but very large, say $L\times L$, then the linear dependence of Eqs. (3) on K would hold for $KL\gg1$; in the opposite limit $KL\ll1$, the dependence on K would be $\sim K^2L$. Even though $KL\simeq\pi$ for the smallest $\mathbf{K}$ in the physical case, it is easy to see that the correct way of calculating $\Delta m(T)$ is to assume an infinite lattice as we have done. Thus, the long-range character of the dipolar interaction is what is ultimately responsible for the modification of the spin-wave spectrum near $k=0$.

In the limit of small $\mathbf{K}$, the integrand reduces to $\{2\alpha K+(E_M/f)K[1-\cos(2\theta)]\}^{-1}$, and doing the angular integration first we find

$$\Delta m(T)_l=\frac{k_BT}{2\pi S\alpha}\ln[(1+\alpha qf/E_M)^{1/2}+(\alpha qf/E_M)^{1/2}] \quad (9)$$

which for $\alpha q/E_M\ll1$ reduces to

$$\Delta m(T)_l\simeq\frac{1}{2\pi S}\frac{k_BT}{(4\pi\beta M_B\alpha)^{1/2}}\sqrt{q} . \quad (10)$$

The convergence of the integral shows that, at least in the

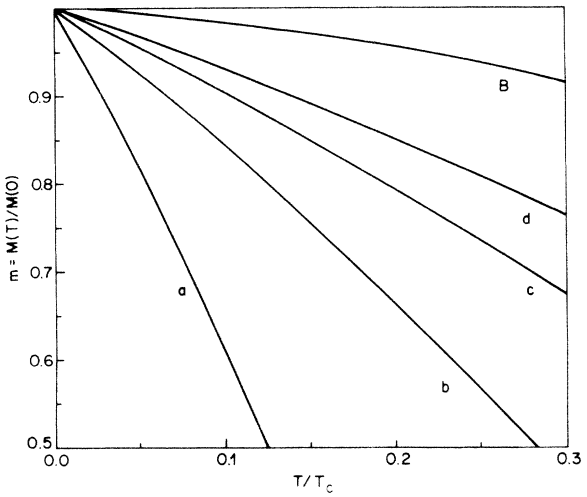

FIG. 1. Normalized magnetization versus reduced temperature. Curves *a*, *b*, *c*, and *d* are for one, two, three, and four hexagonal layers, respectively. *B* is the bulk $T^{3/2}$ law. $S=\frac{1}{2}$. All calculations, for $S=\frac{1}{2}$ as well as $S=\frac{7}{2}$ are done for the hcp lattice, with the coefficient $\sqrt{2/3}$ multiplying the linear K term in Eq. (8).

linearized spin-wave approximation, the dipole-dipole interaction leads to ferromagnetism in a two-dimensional lattice of spins with short-range Heisenberg interactions. For $\mathbf{K}$ values larger than q the evaluation of Eq. (2) must be done numerically. It is possible to choose q small enough that the expansion of the Bose factor is valid, and at the same time large enough for the variation of $A(\mathbf{K})^2-|B(\mathbf{K})|^2$ with the angle θ to be negligible. For such a q the angular integration gives a factor of 2π and Eq. (2) for $|\mathbf{K}|>q$ reduces to a simple integral over K which has been done numerically. The result is shown in Figs. 1 and 2, but before discussing it we shall outline the extension of the calculation to the case of a film of several layers.

Döring[5] has shown that the boundary conditions at the surface of the film require that the spin-wave excitations with wave vector $\mathbf{K}_n$ normal to the film have standing wave rather than running wave character. For a film consisting of D atomic layers the values of $\mathbf{K}_n$ are quantized according to $K_n=(\pi/D)n$ where $n=0,1,\ldots,(D-1)$. The deviation from saturation, $\Delta m(T)$, is now the sum of terms with $n=0,1,\ldots,(D-1)$.

The contribution of the $n=0$ mode is similar to that of Eq. (2) but differs as follows: (a) M_0 and $\Delta M(T)$ are now magnetic moments per unit volume; (b) a factor $1/D$, which is just a normalization factor, enters the right-hand side of Eq. (2); (c) in Eqs. (7a) and (7b) the term linear in $\mathbf{K}$ acquires a factor D. This is because each layer contributes to sums of the type in Eq. (7), but with l^5 replaced by $[l^2+(\mu-t)^2]^{5/2}$, where μ and t refer to the source and field layers, respectively. As discussed above, the contributions to the linear $\mathbf{K}$ term in the sums of Eqs. (3) come primarily from distant points (large l) so that $(t-u)^2$ can be neglected. hence each layer gives the same result, leading to the factor D.

The spin-wave modes with $n\neq 0$ have a gap due to the finite exchange energy, and as a result the influence of the dipolar interaction can be neglected. Summing the contributions from all n values, $\Delta m(T)$ has been calculated for two, three, and four layers, for the extreme values of the spin $S=\frac{1}{2}$ and $S=\frac{7}{2}$. Together with the single-layer case, these results are shown in Figs. 1 and 2. Also shown are the bulk values, corrected in the $S=\frac{7}{2}$ case for the effect of the dipolar interaction.[9] This correction is not negligible and even at $(T/T_C)=0.3$ it decreases the $\Delta m(T)$ of the Bloch $T^{3/2}$ law by 15%.

From Figs. 1 and 2 it is seen that the effect of the dipolar interactions is much more pronounced in the case $S=\frac{7}{2}$. For a single layer and $S=\frac{1}{2}$, the value of $m(T)$ has dropped to 0.6 already by $T=0.1T_C$ while for $S=\frac{7}{2}$

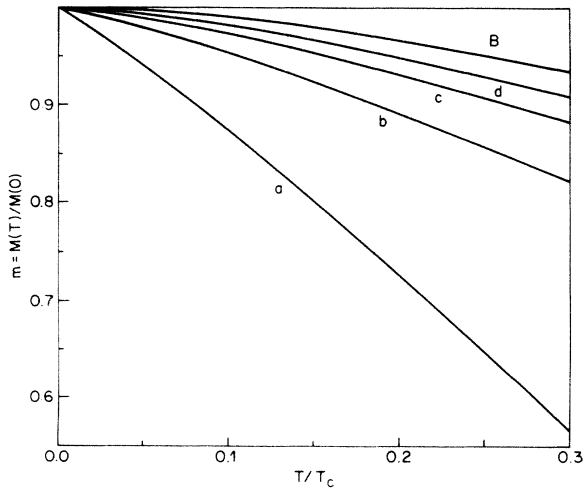

FIG. 2. Normalized magnetization versus reduced temperature. Curves *a*, *b*, *c*, and *d* are for one, two, three, and four hexagonal layers, respectively. *B* is the bulk $T^{3/2}$ law, corrected for the influence of the dipole-dipole interaction. $S=\frac{7}{2}$.

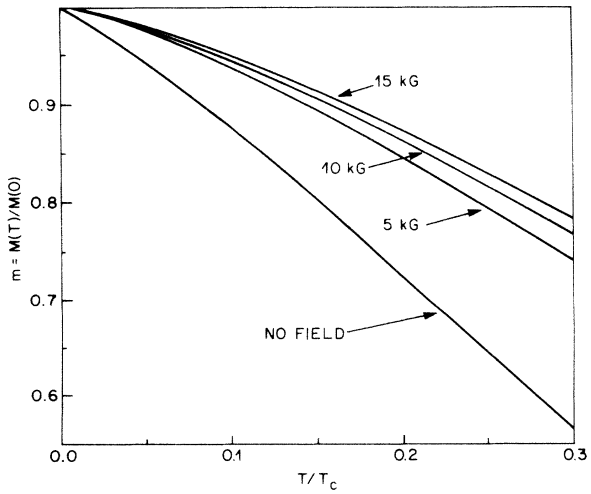

FIG. 3. Single layer magnetization versus reduced temperature in an applied magnetic field: $H=5$, 10, 15 kG. $S=\frac{7}{2}$.

the corresponding value is 0.88. For a three-layer film and $S=\frac{7}{2}$ the curve of $m(T)$ is very close to that of the bulk and bears little resemblance to a linear T law. Calculations for $S=\frac{7}{2}$ have been done with an applied magnetic field and the results are shown in Figs. 3 and 4, respectively, for one- and three-layer films. In Ref. 2, measurements of $m(T)$ in a superlattice having three atomic layers of Gd alternating with ten layers of Y gave the value $m(T)=0.91$ at $T=0.3T_C$ in a field of 12.8 kG. For comparison, the calculated value of $m(T)$ at these values of T and H can be read in Fig. 4; it is $m(T)=0.925$. The agreement is very good but it should not be taken literally because Gd and Y interdiffuse during sample preparation. Furthermore, in the presence of a field as strong as 12 kG the dipolar interaction is nearly negligible (see below for the simulation of the dipolar interaction by an external field) so that the agreement with experiment cannot be viewed as a test of the present theory. Such a test would require measurements of $m(T)$ in low magnetic fields.

Finally it seemed of interest to perform a calculation omitting the dipolar interaction H_D but including an external field, with its magnitude adjusted such as to give the same temperature dependence of the magnetization as that calculated with the dipolar interaction alone. The result, for a single layer and $S=\frac{7}{2}$ is $H\approx 740$ G which can be compared with $4\pi M_B=2.6\times 10^4$ G. The large difference between these values makes it clear that H_D cannot be treated as an effective field.

The simulation of H_D by an external field has also been done for $S=\frac{1}{2}$, in which case a field of only 23 G reproduces the magnetization curve *a* of Fig. 1. So, in substances with small values of S (as in Ni films) and especially if T_C is high (large J) the effect of the dipolar interaction can be simulated by a very small magnetic field which in practice can always be rationalized as an anisotropy field.

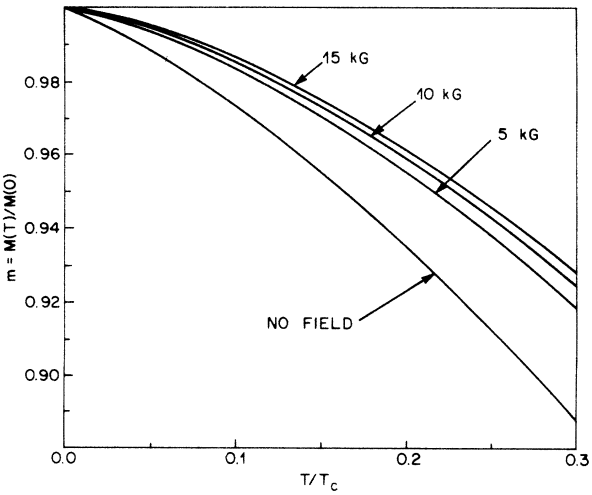

FIG. 4. Magnetization versus reduced temperature of a three-layer array in an applied field: $H=5$, 10, 15 kG. $S=\frac{7}{2}$.

To conclude, we have shown that for localized spins coupled by short-range interactions, the dipole-dipole interaction leads to ferromagnetism in two-dimensional systems, and that this effect is especially important in substances with a large value of the spin and not too high Curie temperatures.

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