

Order of two-dimensional isotropic dipolar antiferromagnets

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(Received 24 September 1992)

The question of the existence of order in two-dimensional isotropic dipolar Heisenberg antiferromagnets is studied. It is shown that the dipolar interaction leads to a gap in the spin-wave energy and a nonvanishing order parameter. The resulting finite Néel temperature is calculated for a square lattice by means of linear spin-wave theory.

The question of order in low-dimensional systems has attracted the interest of theoretical and experimental physics for a long time. It has been pointed out by Bloch¹ and proven exactly² that long-range order is absent in isotropic two-dimensional Heisenberg ferromagnets with short-range interaction. The same is true for one- and two-dimensional ⁴He (Refs. 3 and 4) and antiferromagnets.

In real systems one unavoidably has a dipolar interaction in addition to the short-range exchange interaction, which breaks the rotational symmetry. It has been shown by Maleev⁵ that the q^2 dispersion law of the isotropic ferromagnet is modified such that a finite order parameter exists in two dimensions. The finite-temperature behavior and in particular the transition temperature have been calculated by Pokrovsky and Feigelman.⁶

At first glance one might believe that the dipolar interaction is unimportant in antiferromagnets due to cancellations because of the alternating order and it may come as a surprise that this expectation is incorrect. A first hint that the dipolar interaction can influence antiferromagnetic behavior comes from the critical region. There the nonlinear coupling of fluctuations of the staggered magnetization and the magnetization, which is no longer conserved, leads to a change of the critical dynamic exponent and of the scaling functions.⁷ In the low-temperature phase, antiferromagnetic spin waves involve the coupled precessional motion of magnetization and staggered magnetization. Since the conservation law for the magnetization is broken by the dipolar forces also, the magnon frequency becomes finite at a wave vector $\mathbf{q} = \mathbf{0}$. Thus we will show below that (i) two-dimensional (2D) Heisenberg antiferromagnetic order exists on simple square lattices due to the dipolar interaction, with the spin orientation perpendicular to the plane; (ii) the magnon frequency has a gap, the magnitude of which depends on the dipolar interaction and the exchange interaction; (iii) the critical field for which the spins rearrange is finite; and (iv) there is a finite Néel temperature which is evaluated.

The Hamiltonian of a dipolar antiferromagnet reads

$$H = - \sum_{l \neq l'} \sum_{\alpha\beta} \left(J_{ll'} \delta_{\alpha\beta} + A_{ll'}^{\alpha\beta} \right) S_l^\alpha S_{l'}^\beta - g\mu_B H_0 \sum_l S_l^z, \quad (1)$$

with spins \mathbf{S}_l at lattice sites \mathbf{x}_l . The first term in brackets is the exchange interaction $J_{ll'}$ and the second the dipole-dipole interaction with

$$A_{ll'}^{\alpha\beta} = -\frac{1}{2}(g\mu_B)^2 \left(\frac{\delta_{\alpha\beta}}{|\mathbf{x}_l - \mathbf{x}_{l'}|^3} - \frac{3(\mathbf{x}_l - \mathbf{x}_{l'})_\alpha(\mathbf{x}_l - \mathbf{x}_{l'})_\beta}{|\mathbf{x}_l - \mathbf{x}_{l'}|^5} \right). \quad (2)$$

Although we are mainly interested in these two terms, we have also included a homogeneous external field H_0 along the z axis (g denotes the Landé factor and μ_B the Bohr magneton). In a two-dimensional system with additional dipole-dipole interaction, the rotational symmetry is broken; thus the Hohenberg-Mermin-Wagner theorem^{2,4} does not apply.

We consider a square lattice in the xy plane with lattice constant a and the spins orientated alternately along the z axis. By means of the Holstein-Primakoff transformation,⁸ the Hamilton operator can be expressed in terms of the Bose operators (a_l^\dagger, a_l) [neglecting terms higher than bilinear in Eqs. (3) and (4)]

$$S_l^x = \sqrt{\frac{S}{2}}(a_l + a_l^\dagger), \quad S_l^y = \mp i\sqrt{\frac{S}{2}}(a_l - a_l^\dagger), \quad (3)$$

$$S_l^z = \pm(S - a_l^\dagger a_l),$$

where the upper (lower) sign is for the first (second) sublattice. This transformation and a Fourier transformation yields

$$H = \sum_{\mathbf{q}} \left\{ A_{\mathbf{q}} a_{\mathbf{q}}^\dagger a_{\mathbf{q}} + \frac{1}{2} B_{\mathbf{q}} (a_{\mathbf{q}} a_{-\mathbf{q}} + a_{\mathbf{q}}^\dagger a_{-\mathbf{q}}^\dagger) + C_{\mathbf{q}} a_{\mathbf{q}} a_{-\mathbf{q}-\mathbf{q}_0} + C_{\mathbf{q}}^* a_{\mathbf{q}}^\dagger a_{-\mathbf{q}-\mathbf{q}_0}^\dagger + D_{\mathbf{q}} a_{\mathbf{q}}^\dagger a_{\mathbf{q}+\mathbf{q}_0} + D_{\mathbf{q}}^* a_{\mathbf{q}+\mathbf{q}_0}^\dagger a_{\mathbf{q}} \right\}, \quad (4)$$

with the coefficients

$$A_{\mathbf{q}} = S(2J_{\mathbf{q}_0} - J_{\mathbf{q}} - J_{\mathbf{q}+\mathbf{q}_0}) + S(2A_{\mathbf{q}_0}^{zz} - A_{\mathbf{q}}^{xx} - A_{\mathbf{q}+\mathbf{q}_0}^{yy}), \quad (5a)$$

$$B_{\mathbf{q}} = S(J_{\mathbf{q}+\mathbf{q}_0} - J_{\mathbf{q}}) + S(A_{\mathbf{q}+\mathbf{q}_0}^{yy} - A_{\mathbf{q}}^{xx}), \quad (5b)$$

$$C_{\mathbf{q}} = iS A_{\mathbf{q}}^{xy}, \quad (5c)$$

$$D_{\mathbf{q}} = iS A_{\mathbf{q}}^{xy} + \frac{1}{2} g\mu_B H_0. \quad (5d)$$

In this description [Eq. (3)] the primitive cell is the chemical,⁹ which is half the magnetic. The wave vector $\mathbf{q}_0 = \frac{\pi}{a}(1, 1)$ represents the antiferromagnetic, staggered modulation via $e^{i\mathbf{q}_0 \cdot \mathbf{x}_i}$. The $A_{\mathbf{q}}^{\alpha\beta}$ are the Fourier transform of the dipole tensor [Eq. (2)] and can be calculated by the method of Ewald summation.¹⁰

The Hamiltonian (Eq. 4) is diagonalized by a generalized Bogoliubov transformation with two kinds of creation and annihilation operators $c_{\mathbf{q}}^1, c_{\mathbf{q}}^{1\dagger}, c_{\mathbf{q}}^2, c_{\mathbf{q}}^{2\dagger}$:

$$H = E(0) + \sum_{\mathbf{q}} \sum_{i=1}^2 E_{\mathbf{q}}^i c_{\mathbf{q}}^{i\dagger} c_{\mathbf{q}}^i, \quad (6)$$

$$a_{\mathbf{q}} = \sum_{i=1}^2 u_{\mathbf{q}}^i c_{\mathbf{q}}^i + v_{\mathbf{q}}^{i*} c_{-\mathbf{q}}^{i\dagger} + s_{\mathbf{q}+\mathbf{q}_0}^i c_{\mathbf{q}+\mathbf{q}_0}^i + t_{\mathbf{q}+\mathbf{q}_0}^{i*} c_{-\mathbf{q}-\mathbf{q}_0}^{i\dagger}, \quad (7)$$

$$[c_{\mathbf{p}}^i, c_{\mathbf{q}}^{j\dagger}] = \delta_{\mathbf{p}\mathbf{q}} \delta_{ij}, \quad [c_{\mathbf{p}}^i, c_{\mathbf{q}}^j] = [c_{\mathbf{p}}^{i\dagger}, c_{\mathbf{q}}^{j\dagger}] = 0,$$

with wave vectors restricted to the magnetic Brillouin zone. Here $E(0)$ is the ground-state energy. The spin-wave energies then assume the form

$$(E_{\mathbf{q}}^i)^2 = \frac{1}{2}(\Omega_1 \pm \Omega_2), \quad (8)$$

with

$$\Omega_1 = A_{\mathbf{q}}^2 - B_{\mathbf{q}}^2 + A_{\mathbf{q}+\mathbf{q}_0}^2 - B_{\mathbf{q}+\mathbf{q}_0}^2 + 8C_{\mathbf{q}} C_{\mathbf{q}+\mathbf{q}_0} + 2(g\mu_B H_0)^2$$

and

$$\Omega_2^2 = (A_{\mathbf{q}}^2 - B_{\mathbf{q}}^2 - A_{\mathbf{q}+\mathbf{q}_0}^2 + B_{\mathbf{q}+\mathbf{q}_0}^2)^2 + 16[C_{\mathbf{q}+\mathbf{q}_0}(A_{\mathbf{q}+\mathbf{q}_0} - B_{\mathbf{q}+\mathbf{q}_0}) - C_{\mathbf{q}}(A_{\mathbf{q}} - B_{\mathbf{q}})] \times [C_{\mathbf{q}}(A_{\mathbf{q}+\mathbf{q}_0} + B_{\mathbf{q}+\mathbf{q}_0}) - C_{\mathbf{q}+\mathbf{q}_0}(A_{\mathbf{q}} + B_{\mathbf{q}})] + 4(g\mu_B H_0)^2[(A_{\mathbf{q}} + A_{\mathbf{q}+\mathbf{q}_0})^2 - (B_{\mathbf{q}} - B_{\mathbf{q}+\mathbf{q}_0})^2].$$

Let us now discuss Eq. (8) in the case of primary interest, namely, vanishing external field ($H_0 = 0$). The dipolar interaction has two effects: First, in contrast to the isotropic case, the excitation spectrum is no more degenerate; i.e., two different branches appear. Second, it produces an energy gap for $\mathbf{q} \rightarrow 0$,

$$E_0 = 2S \sqrt{A_{\mathbf{q}_0}^{zz} - A_{\mathbf{q}_0}^{\rho\rho}} \sqrt{(J_{\mathbf{q}_0} - J_0) - (A_0^{\rho\rho} - A_{\mathbf{q}_0}^{zz})}, \quad (9)$$

with

$$A_{\mathbf{k}}^{\rho\rho} = \frac{1}{2}(A_{\mathbf{k}}^{xx} + A_{\mathbf{k}}^{yy}).$$

In Fig. 1 the dispersion relation is shown for three values for the ratio of dipolar and exchange energy $\kappa = \frac{(g\mu_B)^2}{4|J|a^3}$ with isotropic nearest-neighbor exchange interaction ($J < 0$). The two branches can be resolved only for large values of κ . For more realistic ratios (10^{-3}) the two magnon branches practically coincide, but a significant deviation from the pure exchange case still remains

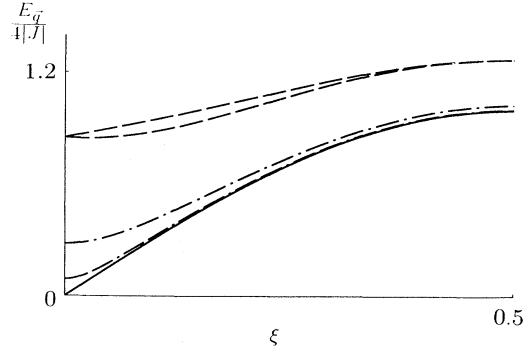


FIG. 1. Spin-wave dispersion relation [Eq. (8)] of pure exchange antiferromagnets with nearest-neighbor interaction (solid line) and with additional dipolar interaction ($S = 1/2$), for the ratios of dipolar energy to exchange energy $\kappa = \frac{(g\mu_B)^2}{4|J|a^3}$ along the $\frac{\pi}{a}[\xi, \xi, 0]$ direction: $\kappa = 0.1$ (dashed line), $\kappa = 0.01$ (dot-dashed line) and $\kappa = 0.001$ (dot-dashed-dashed line). The splitting of the two magnon branches is visible only for $\kappa = 0.1$.

in the immediate vicinity of the zone center. The argument of the first square root in Eq. (9) for the gap equals the difference of dipolar energy for out-of- and in-plane staggered orientation and is positive. Thus stability of the ground state requires

$$J_{\mathbf{q}_0} - J_0 > A_0^{\rho\rho} - A_{\mathbf{q}_0}^{zz} > 0.$$

For dipole interaction alone the system would order with the magnetization in the plane. Thus the dipolar energy difference between in-plane and out-of plane orientation must be exceeded by the exchange energy in order to favor the assumed configuration. In particular, the gap is proportional to the square root of the difference of the static energy between the configurations of in-plane and out-of-plane magnetization. In a three-dimensional simple cubic lattice the first root in Eq. (9) vanishes because of the symmetry, but in two-dimensional systems there is a finite gap for perpendicular antiferromagnetic order. We note that for sufficiently large exchange energy the gap is the geometric mean of dipole and exchange energy, which in turn implies that the gap is much larger than the dipolar energy for $\kappa \ll 1$.

Let us add some comments on the interplay of exchange and dipolar interaction. The former imposes the antiferromagnetic order, while the latter leads to the orientation perpendicular to the plane and prevents thermal fluctuations from its destruction. To exhibit more clearly the physical origin of the energy gap and its principal dependence on dipolar and exchange interaction, we exhibit the equations of motion for the spin components. Approximating the longitudinal part (z component) by $S_i^z \approx S e^{i\mathbf{q}_0 \cdot \mathbf{x}_i}$ and specializing to $\mathbf{q} = 0$, the equations for the transverse components become

$$\dot{S}_0^x = (A_0 - B_0)S_{\mathbf{q}_0}^y, \quad (10a)$$

$$\dot{S}_{\mathbf{q}_0}^y = -(A_0 + B_0)S_0^x, \quad (10b)$$

with an analogous set for the S_0^y component. The co-

efficient on the right-hand side of Eq. (10a) assumes a finite value in contrast to pure exchange antiferromagnets where $S_{\mathbf{q}}^x$ is conserved. Thus the coupled motion of $S_{\mathbf{q}_0}^x$ and $S_{\mathbf{q}_0}^y$ leads precisely to the finite-energy gap E_0 [Eq. (9)].¹¹

From the spin-wave energy [Eq. (8)] we can calculate the critical field for which the antiferromagnetic Néel state gets destabilized by a magnetic field. It is given by the field H_0^c for which the energy ($\mathbf{q} = 0$) vanishes:

$$H_0^c = \frac{1}{g\mu_B} E_0. \quad (11)$$

Hence the critical field is proportional to the energy gap. In 2D the anisotropic dipolar interaction stabilizes the antiferromagnetic configuration in an external field up to the above value.

Now we turn to the evaluation of T_N , the transition temperature for a vanishing external field, i.e., the temperature at which the staggered magnetization vanishes. We use linear spin-wave theory; i.e., interactions between magnons and temperature renormalization of the magnon energy are neglected. This approximation is justified at low temperature and should lead to an order-of-magnitude estimate of the main dependence on exchange and dipolar interaction. The staggered magnetization then reads

$$N(T) = g\mu_B \left(NS - \sum_l \langle a_l^\dagger a_l \rangle \right) = m_0 - N_0 - N_{\text{th}}(T). \quad (12)$$

Here, N denotes the number of lattice sites. This sum is calculated by means of the transformation [Eq. (7)] and requires the evaluation of the coefficients $u_{\mathbf{q}}, v_{\mathbf{q}}, s_{\mathbf{q}}, t_{\mathbf{q}}$, which are complicated functions of the the coefficients in Eqs. (5a)–(5d). The number of thermally excited magnons $N_{\text{th}}(T)$ can be expressed in terms of the mean number of excitations,

$$n_{\mathbf{q}}^i = \langle c_{\mathbf{q}}^i \dagger c_{\mathbf{q}}^i \rangle = \left(e^{E_{\mathbf{q}}^i/k_B T} - 1 \right)^{-1}. \quad (13)$$

We now consider isotropic nearest-neighbor exchange ($J < 0$) and focus only on the limit of small wave vectors and small dipolar energies ($\kappa \ll 1$). The deviation of the ground-state magnetization due to thermal excitations then takes the form

$$N_{\text{th}}(T) = g\mu_B \sum_{i,\mathbf{q}} \frac{D}{E_{\mathbf{q}}^i} n_{\mathbf{q}}^i \quad (14)$$

and the deviation, originating from the zero-point oscillations,

$$N_0 = \frac{1}{2} g\mu_B \sum_{i,\mathbf{q}} \left(\frac{D}{E_{\mathbf{q}}^i} - 1 \right), \quad (15)$$

with $D = 8S|J|$ and $E_{\mathbf{q}}^i \approx \sqrt{D^2 a^2 q^2/2 + E_0^2}$. The zero-point deviation N_0 has a finite value¹² for the pure exchange interaction already, and is effected only negligibly by the dipole interaction. The dipole interaction favors the antiferromagnetic order and leads to a reduction of N_0 of order κ .

Now we turn to $N_{\text{th}}(T)$, which is divergent for pure exchange antiferromagnets, implying the absence of antiferromagnetic order in this case. The small wave-vector approximation in Eqs. (13)–(15) is not accurate for temperatures near the phase transition, but it is sufficient for our crude estimate. From Fig. 1 it becomes clear that the regime of small wave vectors is essential for the calculation of the sum. The existence of a nonvanishing gap makes the sum convergent and allows a phase transition [Eq. (12)] at a finite temperature T_N :

$$N_{\text{th}}(T_N) = m_0 - N_0 = b'. \quad (16)$$

After replacing the sum in Eq. (14) by an integral and the Brillouin zone by a circle of the same area, the evaluation leads to the following implicit equation (the upper bound of the integral is set to infinity):

$$e^{\frac{E_0}{k_B T_N}} - e^{\frac{E_0 - b}{k_B T_N}} = 1, \quad (17)$$

for T_N with $b = \frac{\pi D}{2g\mu_B N} b'$. In the limit of vanishing gap ($E_0 \rightarrow 0$) we recover again the impossibility of a phase transition. From Eq. (17) an asymptotic solution for small dipole energies can be derived:

$$T_N = -\frac{b}{k_B \ln \frac{E_0}{b}} \sim \frac{D}{\ln \frac{D}{E_0}}. \quad (18)$$

We now compare our results with experiments on K_2MnF_4 for which the spin-wave dispersion has been measured.¹³ K_2MnF_4 is a quasi-two-dimensional antiferromagnet with the spin orientation perpendicular to the ab plane, a transition temperature $T_N = 42$ K, and an exchange energy $|J_1| = 8.7$ K ($S = 5/2$). An energy gap of $E_0 = 7.5$ K is observed. Evaluation of the energy gap via Eq. (9) yields $E_0 = 7.6$ K, which is in remarkable agreement with the experimental result. Solving Eq. (17) with the zero point deviation $N_0 = 0.2Ng\mu_B$,¹² our estimate for the transition temperature becomes $T_N = 112$ K, which is too large by a factor of 3. It will be lowered if instead of the small wave-vector expansion the correct dispersion relation [Eq. (8)] shown in Fig. 1 is used in the evaluation of N_{th} [Eq. (14)]. Furthermore we have neglected the interaction of magnons, which will be important at higher temperatures and will lower T_N . This could be treated by more elaborate theories, e.g., Ref. 14, but goes beyond the scope of this paper.

In summary, we have shown that two-dimensional antiferromagnetic order is possible due to the dipolar interaction.

ACKNOWLEDGMENTS

This work has been supported by the German Federal Ministry for Research and Technology (BMFT) under Contract No. 03-SC2TUM. We thank E. Frey and W. Gasser for useful comments.

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