Néel temperature for quasi-two-dimensional dipolar antiferromagnets

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We calculate the Néel temperature T_N for two-dimensional isotropic dipolar Heisenberg antiferromagnets via linear spin-wave theory and a high-temperature expansion, by employing the method of Callen. The theoretical predictions for T_N for K_2MnF_4, Rb_2MnF_4, Rb_2MnCl_4, and $(CH_3NH_3)_2MnCl_4$ are in good agreement with the measured values.

Recently[1] it has been shown that long-range order is possible in two-dimensional isotropic Heisenberg antiferromagnets due to the anisotropy of the dipole-dipole interaction. The occurrence of a finite energy gap goes in hand with a nonvanishing order parameter and a finite Néel temperature. In the present paper a quantitative improvement of the theory is achieved by means of Callen's extension of the Tyablikov decoupling scheme.[2]

In Ref. 1 we have used linear spin-wave theory based on the Holstein-Primakoff transformation to evaluate the magnon dispersion relation. The evaluation of the Néel temperature by means of the temperature-independent dispersion relation leads to an overestimate of T_N. In reality the magnon frequency softens with increasing temperature and thus the actual transition temperature is lower. This feature is accounted for by an extension of the Tyablikov decoupling scheme due to Callen.[2] In essence the dependence on the magnitude of the spin S is replaced by σ, the temperature-dependent order parameter. Even at $T = 0$ the zero point fluctuations lead to a reduction of σ as compared to S. The resulting transition temperature is lowered in comparison to the estimate of Ref. 1 such that a satisfactory agreement between theory and experiment is achieved.

The Hamiltonian of a dipolar antiferromagnet reads

$$H = -\sum_{l \neq l'} \sum_{\alpha\beta} \left(J_{ll'} \delta_{\alpha\beta} + A_{ll'}^{\alpha\beta} \right) S_l^\alpha S_{l'}^\beta , \qquad (1)$$

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 usual dipole-dipole interaction. We consider a square lattice in the xy plane with lattice constant a and the spins orientated alternatingly along the z axis. The out-of-plane orientation is the classical ground state for the isotropic dipolar antiferromagnet with a nearest-neighbor exchange energy $|J|$ much larger than the dipole energy.[1]

Let us introduce the retarded double-time Green func-

tions according to Callen,[2]

$$G^1(\mathbf{R}_k - \mathbf{R}_0, t) = -i\Theta(t)\langle[S_k^+(t), e^{bS_0^z}S_0^-(0)]\rangle$$
$$\equiv \langle\langle S_k^+ | e^{bS_0^z}S_0^- \rangle\rangle, \qquad (2a)$$

$$G^2(\mathbf{R}_k - \mathbf{R}_0, t) = -i\Theta(t)\langle[S_k^-(t), e^{bS_0^z}S_0^-(0)]\rangle$$
$$\equiv \langle\langle S_k^- | e^{bS_0^z}S_0^- \rangle\rangle, \qquad (2b)$$

which obey the following Fourier transformed equations of motion:[3,4]

$$\omega\langle\langle S_k^+ | e^{bS_0^z}S_0^- \rangle\rangle_\omega = \Theta_b + \langle\langle[S_k^+, H]|e^{bS_0^z}S_0^- \rangle\rangle_\omega , \qquad (3a)$$

$$\omega\langle\langle S_k^- | e^{bS_0^z}S_0^- \rangle\rangle_\omega = \langle\langle[S_k^-, H]|e^{bS_0^z}S_0^- \rangle\rangle_\omega , \qquad (3b)$$

with the equal time commutator

$$\Theta_b = \langle[S^+, e^{bS^z}S^-]\rangle.$$

Here $\mathbf{R}_0$ is a lattice point of sublattice 1 and $\mathbf{R}_k$ is a general lattice vector. The higher order Green functions generated by the commutator in Eqs. (3a) and (3b) are approximated by the Tyablikov decoupling scheme

$$\langle\langle S_l^z S_m^+ | e^{bS_0^z}S_0^- \rangle\rangle \rightarrow \langle S_l^z \rangle \langle\langle S_m^+ | e^{bS_0^z}S_0^- \rangle\rangle. \qquad (4)$$

As a consequence of translational symmetry the mean spin value is independent of the lattice site for each sublattice: $\langle S_{l_1}^z \rangle = -\langle S_{l_2}^z \rangle = \sigma$ where $l_1 \in L_1$ ($l_2 \in L_2$) refers to sublattice 1 (2). Using this approximation we obtain from Eqs. (3a) and (3b) a set of four equations for the Green functions $G_\mathbf{q}^1$, $G_{\mathbf{q}+\mathbf{q}_0}^1$, $G_\mathbf{q}^2$, and $G_{\mathbf{q}+\mathbf{q}_0}^2$. The evaluation of the order parameter and the magnon dispersion relation requires only two of them:

$$\tilde{G}_\mathbf{q}^1(\omega) \equiv \frac{1}{2}[G_\mathbf{q}^1(\omega) + G_{\mathbf{q}+\mathbf{q}_0}^1(\omega)]$$
$$= \Theta_b \frac{A^{(1)} + B^{(1)}\omega + C^{(1)}\omega^2 + \omega^3}{(\omega^2 - \epsilon_1^2)(\omega^2 - \epsilon_2^2)}, \qquad (5)$$

with

$$-A^{(1)} = \frac{1}{2}A_\mathbf{q}(A_{\mathbf{q}+\mathbf{q}_0}^2 - B_{\mathbf{q}+\mathbf{q}_0}^2) + \frac{1}{2}A_{\mathbf{q}+\mathbf{q}_0}(A_\mathbf{q}^2 - B_\mathbf{q}^2) + (C_\mathbf{q}^2 + C_{\mathbf{q}+\mathbf{q}_0}^2)(A_\mathbf{q} + A_{\mathbf{q}+\mathbf{q}_0}) + (C_\mathbf{q}^2 - C_{\mathbf{q}+\mathbf{q}_0}^2)(B_{\mathbf{q}+\mathbf{q}_0} - B_\mathbf{q}),$$

$$-B^{(1)} = \frac{1}{2}(A_\mathbf{q}^2 - B_\mathbf{q}^2 + A_{\mathbf{q}+\mathbf{q}_0}^2 - B_{\mathbf{q}+\mathbf{q}_0}^2 + 8C_\mathbf{q}C_{\mathbf{q}+\mathbf{q}_0}),$$

$$C^{(1)} = \frac{1}{2}(A_\mathbf{q} + A_{\mathbf{q}+\mathbf{q}_0}).$$

Here $\mathbf{q}$ denotes the wave vector of the chemical Brillouin zone and $\mathbf{q}_0 = \frac{\pi}{a}(1,1,0)$. In the magnetic Brillouin zone which is half the chemical there exist two distinct spin-wave branches with frequencies ϵ_i $(i = 1,2)$ which read

$$\epsilon_i^2 = \frac{1}{2}(\Omega_1 \pm \Omega_2),$$

$$\Omega_1 = A_{\mathbf{q}}^2 - B_{\mathbf{q}}^2 + A_{\mathbf{q}+\mathbf{q}_0}^2 - B_{\mathbf{q}+\mathbf{q}_0}^2 + 8C_{\mathbf{q}}\,C_{\mathbf{q}+\mathbf{q}_0}\ , \qquad (6)$$

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

In Eq. (6) the coefficients

$$A_{\mathbf{q}} = \sigma(2J_{\mathbf{q}_0} - J_{\mathbf{q}} - J_{\mathbf{q}+\mathbf{q}_0}) + \sigma(2A_{\mathbf{q}_0}^{zz} - A_{\mathbf{q}}^{xx} - A_{\mathbf{q}+\mathbf{q}_0}^{yy}), \qquad (7a)$$

$$B_{\mathbf{q}} = \sigma(J_{\mathbf{q}+\mathbf{q}_0} - J_{\mathbf{q}}) + \sigma(A_{\mathbf{q}+\mathbf{q}_0}^{yy} - A_{\mathbf{q}}^{xx}), \qquad (7b)$$

$$C_{\mathbf{q}} = i\sigma A_{\mathbf{q}}^{xy} \qquad (7c)$$

have been introduced. This result, Eq. (6), coincides with the magnon frequencies derived by the Holstein-Primakoff transformation when σ is replaced by S and

the external magnetic field is set to zero.[1] But there is a difference even at absolute zero temperature, because the ground state of the antiferromagnet is not the Néel state of fully aligned spins; i.e. $\sigma(T = 0) < S$ as will be seen later. Note that the magnon frequency scales with the order parameter σ; i.e., the whole spectrum softens with increasing temperatures and vanishes at the phase transition.

Now we turn to the evaluation of σ. For arbitrary spin S the spin expectation value is given by the well-known relation

$$\langle S^z \rangle = S(S + 1) - \langle (S^z)^2 \rangle - \langle S^- S^+ \rangle. \qquad (8)$$

For $S = 1/2$ the order parameter can be calculated directly via the Green functions of Eqs. (2a) and (2b) with $b \equiv 0$ and $\Theta_0 = 2\sigma$. For higher spin quantum numbers the above formula is not so helpful. Then a convenient starting point is the generalized thermal average[2]

$$\psi(b) \equiv \langle e^{bS_0^z} S_0^- S_0^+ \rangle, \qquad (9)$$

by means of which a self-consistent system of equations can be derived by the method of Callen. The above thermal average can be represented by the spectral theorem[3,4]

$$\langle e^{bS_0^z} S_0^- S_0^+ \rangle = \lim_{\delta \to 0} -\frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega\ n(\omega) \frac{1}{N} \sum_{\mathbf{q}} \{\tilde{G}_{\mathbf{q}}^1(\omega + i\delta) - \tilde{G}_{\mathbf{q}}^1(\omega - i\delta)\}$$

$$= \Theta_b(n - 1/2), \qquad (10)$$

where Eq. (5) has been used in the last step and N denotes the total number of spins. Here we have introduced the Bose occupation number

$$n(\omega) = \left(e^{\frac{\omega}{k_B T}} - 1\right)^{-1}$$

and

$$n = \frac{1}{N} \sum_{\mathbf{q}} \frac{-A^{(1)} + C^{(1)}\epsilon_1\epsilon_2}{2\epsilon_1\epsilon_2(\epsilon_1 + \epsilon_2)}$$

$$+ \frac{A^{(1)} + C^{(1)}\epsilon_1^2}{\epsilon_1(\epsilon_1^2 - \epsilon_2^2)}n(\epsilon_1) + \frac{A^{(1)} + C^{(1)}\epsilon_2^2}{\epsilon_2(\epsilon_2^2 - \epsilon_1^2)}n(\epsilon_2)\ . \qquad (11)$$

The right-hand side of Eq. (11) depends on σ only via the occupation numbers $n(\epsilon_i)$. For spin $S = 1/2$ and vanishing parameter b the thermal average [Eq. (9)] represents the number of spin-wave excitations, which reduce the staggered magnetization from the totally ordered Néel state, not only for finite but also for zero temperature.

For arbitrary spin one has to express σ in terms of n which can be achieved by the method of Callen with the result[2,5]

$$\sigma = \left(S + \frac{1}{2}\right)\frac{(n + 1/2)^{2S+1} + (n - 1/2)^{2S+1}}{(n + 1/2)^{2S+1} - (n - 1/2)^{2S+1}} - n. \qquad (12)$$

Equations (11) and (12) constitute a self-consistent system of equations for n and the spin expectation value σ.

Let us now discuss the dispersion relation for $T = 0$. In this limit Eq. (11) reduces to

$$n_0 \equiv n(T = 0) = \frac{1}{N} \sum_{\mathbf{q}} \frac{-A^{(1)} + C^{(1)}\epsilon_1\epsilon_2}{2\epsilon_1\epsilon_2(\epsilon_1 + \epsilon_2)}, \qquad (13)$$

where the right-hand side is independent of σ. Let us denote the spin expectation value at zero temperature by $\sigma_0 = \sigma(T = 0)$ which is found by inserting Eq. (13) into (12). Knowing σ_0 one obtains, for the staggered magnetization $N(0)$ at $T = 0$,

$$N(0) = g\mu_B N\sigma_0. \qquad (14)$$

One can convince oneself that Eq. (14) for large S coincides with the expression derived by the Holstein-Primakoff transformation[1] (see also Ref. 5). This must be so because the latter is an expansion in $1/S$. In addition we derive an energy gap $(\mathbf{q} = 0)$ from Eq. (6)

$$E_0^\sigma \equiv \epsilon_1(T = 0) = \epsilon_2(T = 0) = 2\sigma_0\sqrt{A_{\mathbf{q}_0}^{zz} - A_0^{\rho\rho}}\sqrt{(J_{\mathbf{q}_0} - J_0) - (A_0^{\rho\rho} - A_{\mathbf{q}_0}^{zz})}\ , \qquad (15)$$

with

$$A_0^{\rho\rho} = A_0^{xx} = A_0^{yy}, \qquad A_{q_0}^{\rho\rho} = A_{q_0}^{xx} = A_{q_0}^{yy}.$$

This is of the same form as the result from the Holstein-Primakoff transformation E_0 Ref. 1 except for the prefactor which is smaller by the ratio σ_0/S.

Now we turn to the evaluation of the transition temperature T_N, i.e. consider the limit $\sigma \to 0$. Since the spin-wave energy [Eq. (6)] is proportional to σ the Bose occupation numbers can be replaced by their classical limit:

$$n(\epsilon_i) \to \frac{k_B T}{\epsilon_i} \ .$$

If this is inserted into Eq. (11) together with $\sigma \to 0$, one obtains

$$n = \frac{k_B T_N}{N\sigma} \sum_{\mathbf{q}} \frac{-\tilde{A}^{(1)}}{(\tilde{\epsilon}_1 \tilde{\epsilon}_2)^2} \ . \tag{16}$$

To keep track of the σ dependence we have introduced the σ-independent quantities $\tilde{A}^{(1)} = A^{(1)}/\sigma^3$ and $\tilde{\epsilon}_i = \epsilon_i/\sigma$. According to Eq. (16) n increases indefinitely with $\sigma \to 0$ and thus the second relation between σ and n, Eq. (12), becomes[2]

$$\sigma = \frac{S(S+1)}{3} \frac{1}{n}. \tag{17}$$

Combining Eqs. (16) and (17) we obtain an explicit expression for the Néel temperature:

$$T_N = \frac{S(S+1)}{3k_B} F^{-1} \ , \tag{18}$$

with

$$F = \frac{1}{N} \sum_{\mathbf{q}} \frac{-\tilde{A}^{(1)}}{(\tilde{\epsilon}_1 \tilde{\epsilon}_2)^2} \ .$$

For purely isotropic antiferromagnets the coefficient F diverges, excluding long-range order at finite temperature in two dimensions in accord with the Hohenberg-Mermin-Wagner theorem.[6,7]

In the presence of the dipolar interaction there is an energy gap and T_N becomes finite. If the dipolar interaction is weak in comparison with the exchange energy $(g\mu_B)^2/a^3 \ll |J|$ and if the argument of the summation is approximated by its small $\mathbf{q}$ limit, one obtains

$$T_N \sim \frac{|J|}{\ln\frac{|J|}{E_0}} \ , \tag{19}$$

which coincides with the analogous formula derived by the Holstein-Primakoff transformation [Eq. (18) in Ref. 1]. In the general case the above sum [Eq. (18)] is evaluated with the full dispersion relation Eq. (6) and by computing 100×100 points in the two-dimensional (2D) Brillouin zone and determining the other points by linear extrapolation. The dipole sums have been calculated via Ewald summation.[8]

Now we apply our theory to real quasi-two-dimensional antiferromagnets. Prominent examples of almost two-dimensional antiferromagnets are the tetragonal antiferromagnetic halides K_2MnF_4, Rb_2MnF_4, Rb_2MnCl_4, and $(CH_3NH_3)_2MnCl_4$. In these quadratic layer structures the out-of-plane exchange interaction is negligible in comparison to the in-plane exchange interaction (about 10^{-4}),[9,10] whereas the dipole energy is larger by an order of magnitude. This two-dimensional character has been shown experimentally by the absence of any dispersion along the z direction.[10] For these halides the measured exchange energy $|J|$, the lattice constant a, the energy gap E_0^{expt}, the spin-flop field H_{sf}^{expt}, and the transition temperature T_N^{expt} (Refs. 10–12) are listed in Table I. The spin-flop field H_{sf} is the critical magnetic field at which the antiferromagnetic Néel ground state changes to the spin-flop ground state. It can be calculated by adding to the Hamiltonian, Eq. (1), the Zeeman energy.[13] From the full dispersion-relation the spin-flop field H_{sf}^σ is defined by that field for which the magnon energy vanishes,

$$H_{sf}^\sigma = \frac{1}{g\mu_B} E_0^\sigma, \tag{20}$$

in close analogy to the formula obtained by the Holstein-Primakoff method [Eq. (11) in Ref. 1]. The table also contains the theoretical energy gaps E_0^σ and E_0 calculated via Eq. (15) and Eq. (9) of Ref. 1, the resulting spin-flop fields H_{sf}^σ and H_{sf} via Eq. (20) and Eq. (11) of Ref. 1, and the theoretical transition temperature T_N [Eq. (18)]. All these substances have spin $S = 5/2$, which yields from Eqs. (13) and (12) $\sigma_0 = 2.30$ for pure isotropic antiferromagnets. This value is increased only negligibly by the dipolar interaction as can be seen from Table I.

TABLE I. Exchange energy $|J|$, lattice constant a, energy gap E_0, spin-flop field H_{sf}, Néel temperature T_N, and zero temperature order parameter σ_0.

	$\begin{array}{c}|J| \\ (K)\end{array}$	$\begin{array}{c}a \\ (\text{Å})\end{array}$	$\begin{array}{c}E_0^{expt} \\ (K)\end{array}$	$\begin{array}{c}E_0^\sigma \\ (K)\end{array}$	$\begin{array}{c}E_0 \\ (K)\end{array}$	$\begin{array}{c}H_{sf}^{expt} \\ (T)\end{array}$	$\begin{array}{c}H_{sf}^\sigma \\ (T)\end{array}$	$\begin{array}{c}H_{sf} \\ (T)\end{array}$	$\begin{array}{c}T_N^{expt} \\ (K)\end{array}$	$\begin{array}{c}T_N^{th} \\ (K)\end{array}$	σ_0
K_2MnF_4	8.5[a]	4.17[a]	7.4[b]	7.1	7.6	5.4[d]	5.3	5.6	42[a]	41	2.33
Rb_2MnF_4	7.4[c]	4.20[g]	7.3[b]	6.5	7.0		4.8	5.1	38[g]	36	2.33
Rb_2MnCl_4	11.2[f]	5.05[e]	7.5[f]	6.1	6.6		4.5	4.9	56[f]	48	2.32
$(CH_3NH_3)_2MnCl_4$	9.0[f]	5.13[e]		5.3	5.7		3.9	4.3	45[f]	39	2.32

[a] Reference 10.
[b] Reference 15.
[c] Reference 9.
[d] Reference 11.

[e] Reference 16.
[f] Reference 12.
[g] Reference 17.

We find a good agreement with the measured Néel temperature although our theory accounts only for the dipolar interaction and no other anisotropy. Corrections due to dipolar interactions between different planes are negligible because of the large lattice constant in the z direction; e.g., the energy gap for K_2MnO_4 [Eq. (15)] is altered by

$$E_0^\sigma(3D) = E_0^\sigma(2D)[1 + \mathcal{O}(10^{-5})], \qquad (21)$$

which justifies the application of the two-dimensional model. Note that the nearest-neighbor exchange energy J is the only parameter entering in our theory. Experimentally this parameter has been derived by fitting the measured spin-wave spectrum with a dispersion relation which is different of ours [Eq. (6)].

For the halides listed in Table I, the energy gap obtained from Eq. (15) and the transition temperature are lower than the experimental values. The following reasons may be responsible for that: (i) In the Holstein-Primakoff approximation the softening of the magnons is neglected entirely. This leads to an overestimate of T_N. In the Callen method the magnons soften in the entire Brillouin zone; thus particularly near the phase transition the softening is overestimated and leads to a T_N which is somewhat too low. (ii) A small readjustment of J could be necessary if our dispersion relation, Eq. (6), is used to fit the data. (iii) A small additional anisotropy from the crystal field might be present as suggested by Ref. 14.

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