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Softened spin-wave dispersion and sublattice magnetization at finite temperature for a three-dimensional anisotropic Heisenberg antiferromagnet

Jin An[1,2], **Chang-De Gong**[3,1] **and Hai-Qing Lin**[2]

[1] National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, China
[2] Department of Physics, The Chinese University of Hong Kong, Hong Kong, China
[3] Chinese Centre of Advanced Science and Technology (World Laboratory), PO Box 8730, Beijing 100080, China

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Abstract

We study a three-dimensional anisotropic quantum antiferromagnetic Heisenberg model at finite temperature in which the interplanar coupling can be varied gradually. With the magnon interactions considered, we have calculated the spin-wave excitation and sublattice magnetization. We find that when the interplanar coupling increases, the Néel ordered state is stabilized gradually. Moreover, we find power laws at low temperatures: a T^4-law for the spin-wave excitation, and a T^2-law for the sublattice magnetization, which is consistent with the nuclear magnetic resonance experiments on antiferromagnets. The interplanar coupling dependences of the finite-temperature spin-wave excitation and magnetization are also given.

1. Introduction

Since the discovery of high-temperature superconductors, a great deal of theoretical interest in the physics of quantum antiferromagnets has arisen since the undoped mother materials like La_2CuO_4 were found to be well described by a square-lattice spin-1/2 antiferromagnetic (AF) Heisenberg model. Now it is widely accepted that the ground state of the spin-1/2 square-lattice Heisenberg antiferromagnet exhibits the Néel long-range order [1–3], with the sublattice magnetization reduced by quantum fluctuation to about 0.303. Until now, most theoretical work has concentrated on the properties of the ground state of the model. Many methods have been developed to investigate them, including $1/S$ expansion (the Holstein–Primakoff (HP) or Dyson–Maleev method) [4–6], Monte Carlo calculations [7–10], exact diagonalizations [11], extended Jordon–Wigner transformation [12] and others [13–17]. However, finite-temperature properties have been studied very little. This is possibly due to the Mermin–Wagner theorem [18] which states that at finite temperatures no true long-range AFM or FM orders can exist in a 2D quantum Heisenberg model.

Nevertheless, on one hand, from experiments based on susceptibility measurements [19] and neutron scattering [20], it has been shown that there exists a Néel ordered temperature T_N below which materials like La_2CuO_4 exhibit a 3D long-range AF order even at rather high temperatures. On the other hand, Cu NMR measurements on antiferromagnets have been reported in recent years for the layered materials La_2CuO_4 [21], $YBa_2Cu_3O_6$ [22] and $Ca(Sr)CuO_2$ [23] which strongly suggest a T^2-law for the low-temperature sublattice magnetization up to approximately $T_N/2$, where T_N is the Néel temperature. These materials can all be seen as 2D AF layers, which are coupled to their nearest neighbours by the effective interplanar couplings. Thus it seems more appropriate to interpret this and other finite-temperature features of La_2CuO_4 in the framework of an anisotropic 3D AF Heisenberg model [24].

The purpose of this paper is to investigate how the interplanar coupling affects a 2D antiferromagnet by studying a 3D anisotropic AF Heisenberg model. Actually, we study a more general problem, where the interplanar coupling can vary from zero to a finite value. When the interplanar coupling is absent, we turn to the 2D isotropic AF Heisenberg model. In this situation, we find that the Néel ordered state is unstable at finite temperature to the quantum fluctuations, which is in agreement with the Mermin–Wagner theorem. When the coupling increases, the Néel ordered state is stabilized gradually. In another limit, where the interplanar coupling is identical to the intraplanar coupling, we have a 3D isotropic AF Heisenberg model, which can be quite well described by the linear spin-wave theory, together with the zero-point contributions from magnon interactions.

At finite temperature, magnons will be excited from the ground state and they will interact with each other and then cause nontrivial corrections to the physical properties of the system. In this paper, we consider these interactions between magnons and investigate what contribution is given to the temperature-dependent part of the spin-wave excitation and sublattice magnetization and then compare the results with experiments.

2. The model

The Hamiltonian of a 3D anisotropic quantum Heisenberg antiferromagnet is defined by

$$H = J \sum_{\langle i,j\rangle_{\parallel}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J_{\perp} \sum_{\langle i,j\rangle_{\perp}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{1}$$

where a cubic crystal structure is assumed and $\langle i, j\rangle_{\parallel}$, $\langle i, j\rangle_{\perp}$ indicate sums over pairs of nearest neighbours in the same horizontal planes or along the axes perpendicular to the planes, respectively. What should be noted is that although the interplanar coupling $J_{\perp}$ may be rather small, compared to the exchange constant J, it is required for the appearance of a 3D long-range AF order at finite temperatures [25].

The presence of the Néel long-range order suggests that the spin-wave expansion ($1/S$ expansion) makes sense. Actually, it has been reported that the linear spin-wave theory—the leading order of the $1/S$ expansion—gives good results, although the whole expansion would be divergent [4]. Therefore, here in this paper we employ an improved linear spin-wave approach to investigate this problem.

Dividing the system into two sublattices (A and B) and performing the HP transformation, we can express the spin operators in terms of sublattice boson operators a_i, b_i and their conjugates:

$$\begin{aligned} S_{ai}^{+} &= (S_{ai}^{-})^{+} = \sqrt{2S} f_i(S) a_i \\ S_{ai}^{Z} &= S - a_i^{+} a_i \end{aligned} \tag{2a}$$

and

$$S_{bi}^{+} = (S_{bi}^{-})^{+} = \sqrt{2S} b_i^{+} f_i(S)$$
$$S_{bi}^{Z} = -S + b_i^{+} b_i \tag{2b}$$

with

$$f_i(S) = \sqrt{1 - \frac{n_i}{2S}} = 1 - \frac{1}{2}\frac{n_i}{2S} - \frac{1}{8}\left(\frac{n_i}{2S}\right)^2 + \cdots \tag{3}$$

where $S = 1/2$, and $n_i = a_i^{+} a_i$ or $b_i^{+} b_i$ is the particle number operator for site i. Substituting equations (2) and (3) into equation (1), and performing Fourier transformation, we have the following Hamiltonian in $\boldsymbol{k}$-space:

$$H = -\frac{NJ}{2}\left(1 + \frac{\lambda}{2}\right) + H_0 + H_I \tag{4}$$

where N is the number of sites and $\lambda = J_{\perp}/J$ is defined as a coupling parameter. H_0, H_I read

$$H_0 = 2J \sum_{\boldsymbol{k}} \left\{ \left(1 + \frac{\lambda}{2}\right)(a_{\boldsymbol{k}}^{+} a_{\boldsymbol{k}} + b_{\boldsymbol{k}}^{+} b_{\boldsymbol{k}}) + \gamma_{\boldsymbol{k}}(a_{\boldsymbol{k}} b_{-\boldsymbol{k}} + \text{h.c.}) \right\} \tag{5}$$

$$H_I = -\frac{2J}{N} \sum_{\boldsymbol{k}_1, \boldsymbol{k}_2, \boldsymbol{q}} \{ \gamma_{-\boldsymbol{k}_1} a_{\boldsymbol{k}_1+\boldsymbol{q}}^{+} a_{\boldsymbol{q}-\boldsymbol{k}_2} a_{\boldsymbol{k}_2} b_{\boldsymbol{k}_1} + \gamma_{-\boldsymbol{k}_1+\boldsymbol{q}} a_{-\boldsymbol{k}_1+\boldsymbol{q}} b_{\boldsymbol{k}_2+\boldsymbol{q}}^{+} b_{\boldsymbol{k}_2} b_{\boldsymbol{k}_1}$$
$$+ 2\gamma_{\boldsymbol{q}} a_{\boldsymbol{k}_1-\boldsymbol{q}}^{+} b_{\boldsymbol{k}_2+\boldsymbol{q}}^{+} b_{\boldsymbol{k}_2} a_{\boldsymbol{k}_1} + \text{h.c.} \} \tag{6}$$

where the sums are over the whole magnetic Brillouin zone and $\gamma_{\boldsymbol{k}}$ is defined by

$$\gamma_{\boldsymbol{k}} = \gamma_{\boldsymbol{k}}^{(1)} + \frac{\lambda}{2}\gamma_{\boldsymbol{k}}^{(2)}$$

with $\gamma_{\boldsymbol{k}}^{(1)} = \frac{1}{2}(\cos k_x + \cos k_y)$, $\gamma_{\boldsymbol{k}}^{(2)} = \cos k_z$. The part H_0 refers to the freely propagating magnons and the part H_I refers to the various processes of interaction between the magnons.

3. Results and discussion

Because of the interactions, magnons would be created or annihilated, but the total momenta are still conserved. The lifetime of the magnons would be finite due to this damping effect. Thus we would like to find another kind of quasi-particle to account for the low-temperature excitation. In order to do this, we introduce another two quasi-particle operators by performing Bogoliubov transformation:

$$\boldsymbol{\alpha}_{\boldsymbol{k}} = u_{\boldsymbol{k}} a_{\boldsymbol{k}} + v_{\boldsymbol{k}} b_{-\boldsymbol{k}}^{+}$$
$$\boldsymbol{\beta}_{\boldsymbol{k}}^{+} = v_{\boldsymbol{k}} a_{\boldsymbol{k}} + u_{\boldsymbol{k}} b_{-\boldsymbol{k}}^{+} \tag{7}$$

where the coefficients $u_{\boldsymbol{k}}$, $v_{\boldsymbol{k}}$ are related by $u_{\boldsymbol{k}}^2 - v_{\boldsymbol{k}}^2 = 1$, and will be determined later by using the condition that $\boldsymbol{\alpha}_{\boldsymbol{k}}$ is a quasi-particle operator and its equation of motion should be diagonalized.

Now we construct the equation of motion for the operator $\boldsymbol{\alpha}_{\boldsymbol{k}}$ and then diagonalize it. However, in solving the equation of motion

$$\mathrm{i}\hbar \frac{\partial}{\partial t}\boldsymbol{\alpha}_{\boldsymbol{k}} = [\boldsymbol{\alpha}_{\boldsymbol{k}}, H]$$

one encounters commutators like $[\boldsymbol{\alpha}_{\boldsymbol{k}}, a_{\boldsymbol{k}1}^{+} b_{\boldsymbol{k}2}^{+} a_{\boldsymbol{k}3} a_{\boldsymbol{k}4}]$; they have to be treated approximately. Here, we adopt a mean-field approach. We calculate all kinds of commutators like

$[\alpha_k, a^+_{k1} b^+_{k2} a_{k3} a_{k4}]$ and then combine every possible pair of operators into their averages to decouple the three-operator terms in the final result. This method is quite different from that employed by other authors in deriving physical properties of the model at zero temperature, where zero-temperature Green's functions are used to perform perturbations of the $1/S$ expansion [5, 6]. Moreover, in the analysis there, quasi-particle representation has been used and thus very complicated vertex functions introduced in the interactions part of the Hamiltonian; this causes the analysis there to be much more complicated. Here, we do not substitute the transformation of equation (21) into the Hamiltonian and just retain the magnon representation in it.

Because the number and momentum of magnons are always conserved (note that annihilating a magnon in an A sublattice corresponds to a process of creating a magnon in a B sublattice), in the decoupling processes here, only the pair averages like $\langle a_q b_{-q}\rangle$, $\langle a^+_q a_q\rangle$, $\langle b^+_q b_q\rangle$ and their conjugates do not vanish, and they can be given by

$$\begin{aligned} \langle a^+_q a_q\rangle &= u_q^2 \langle \alpha^+_q \alpha_q\rangle + v_q^2 \langle \beta_q \beta^+_q\rangle = \langle b^+_q b_q\rangle \\ \langle a_q b_{-q}\rangle &= -u_q v_q (\langle \alpha^+_q \alpha_q\rangle + \langle \beta_q \beta^+_q\rangle). \end{aligned} \tag{8}$$

Thus all of the possible types of four-operator commutator can be calculated and finally we obtain the commutators of α_k and the interaction part of the Hamiltonian as follows:

$$\begin{aligned} [\alpha_k, H_I] = 2J \Bigg\{ &\frac{4}{N} \sum_q \left(\gamma_q \langle a_q b_{-q}\rangle + \left(1 + \frac{\lambda}{2}\right) \langle a^+_q a_q\rangle \right) (-\alpha_k (u_k^2 + v_k^2) + \beta^+_k 2 u_k v_k) \\ &+ \frac{4}{N} \sum_q (\gamma_{k+q} \langle a_q b_{-q}\rangle + \gamma_k \langle a^+_q a_q\rangle)(\alpha_k 2 u_k v_k - \beta^+_k (u_k^2 + v_k^2)) \Bigg\} \end{aligned} \tag{9}$$

where the equations $\langle a^+_q a_q\rangle = \langle b^+_q b_q\rangle$ have been used in order to derive the above equation. It can be seen that in order to make α_k a true quasi-particle operator, one has to arrange for the cross-terms to be eliminated. Note that

$$\sum_q \gamma^{(i)}_{k+q} f(q) = \gamma^{(i)}_k \sum_q \gamma^{(i)}_q f(q) \qquad (i = 1, 2) \tag{10}$$

where $f(q)$ is an any function of q with the symmetry of the crystal. Thus the two coefficients can be determined to give

$$\begin{aligned} u_k &= \left\{ \frac{1}{2} \left(\frac{1}{\sqrt{1 - (\Gamma_k/\Gamma_0)^2}} + 1 \right) \right\}^{1/2} \\ v_k &= \mathrm{sgn}(\Gamma_k/\Gamma_0) \left\{ \frac{1}{2} \left(\frac{1}{\sqrt{1 - (\Gamma_k/\Gamma_0)^2}} - 1 \right) \right\}^{1/2} \end{aligned} \tag{11}$$

where

$$\Gamma_k = \gamma_k \left(1 - \frac{4}{N} \sum_q \langle a^+_q a_q\rangle \right) - \frac{4}{N} \sum_q \gamma_{k+q} \langle a_q b_{-q}\rangle. \tag{12}$$

The spin-wave dispersion of the antiferromagnet can then be evaluated to give

$$E_k = 2J \sqrt{\Gamma_0^2 - \Gamma_k^2}. \tag{13}$$

This gapless excitation is quite different from that calculated from the linear spin-wave theory for it has included the effect of the magnon interactions. Introducing two gravity parameters f and g, which are constants but dependent on the interplanar coupling, we find that Γ_k can be written generally in the following form:

$$\Gamma_k = f \gamma^{(1)}_k + \frac{\lambda}{2} g \gamma^{(2)}_k \tag{14}$$

where f and g can be shown to be determined by the following two self-consistent equations:

$$f = 2 - \frac{1}{(f + \lambda g/2)} \frac{2}{N} \sum_q (1 + 2n_B(E_q)) E_q / 2J$$
$$\frac{g}{2} = 1 - \frac{1}{N} \sum_q \frac{2J}{E_q} (1 + 2n_B(E_q)) \left\{ f[1 - \gamma_q^{(1)} \gamma_q^{(2)}] + \frac{\lambda}{2} g[1 - (\gamma_q^{(2)})^2] \right\} \tag{15}$$

with E_q given by equation (13). In the long-wavelength limit, we have a photon-like energy dispersion:

$$E_k = \sqrt{c_1^2(k_x^2 + k_y^2) + c_2^2 k_z^2}$$

where

$$c_1 = J\sqrt{f(2f + \lambda g)}$$
$$c_2 = J\sqrt{\lambda g(2f + \lambda g)} \tag{16}$$

are the two spin-wave velocities within or perpendicular to the planes, respectively. These two velocities at zero temperature, c_{10}, c_{20}, as functions of the interplanar coupling are also shown in figure 1 (triangles). It can be seen from the figure that, although, as a matter of fact, the spin-wave excitations will be softened at finite temperature, there are still positive corrections to the two spin-wave velocities due to the zero-point contribution from the magnon interactions, compared with that calculated from the linear spin-wave theory. When the interplanar coupling is not very small, i.e. $J_\perp$ is more than about $0.1J$, the two spin-wave velocities increase almost linearly with the increasing interplanar coupling. Finally, they reach the same value for the 3D isotropic AF Heisenberg model: $c_1 = c_2 = 1.900J$. The spectrum in this limit can also be written in the form of a renormalized antiferromagnetic spin-wave dispersion with the renormalized factor given by $\eta = 1.097$, which is relatively small, compared to the 2D case where the renormalized factor $\eta = 1.158$.

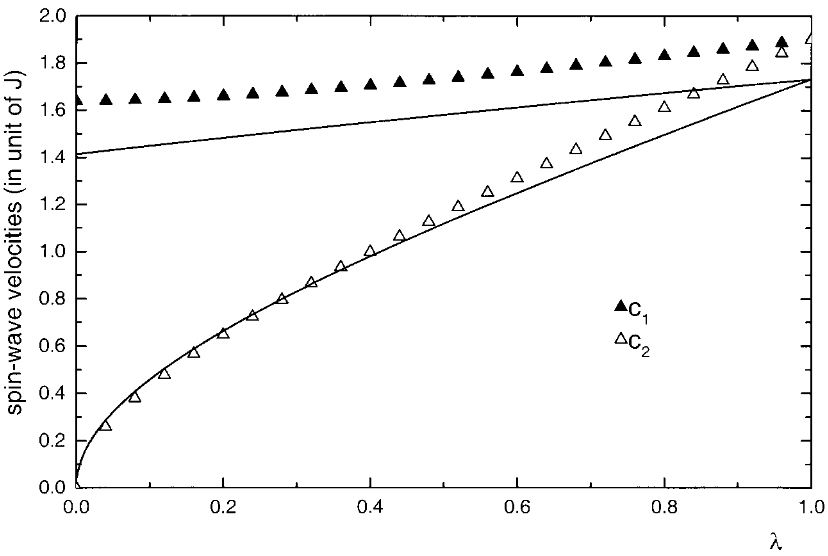

Figure 1. The interplanar coupling dependence of the anisotropic spin-wave velocities (triangles). The solid lines show the corresponding quantities calculated from the linear spin-wave theory.

At low temperature, the velocities can be expanded in powers of $k_B T/J$; the results can be given as the T^4-laws stated below:

$$\begin{aligned} c_1 &= c_{10}\{1 - A(k_B T/J)^4\} \\ c_2 &= c_{20}\{1 - B(k_B T/J)^4\} \end{aligned} \tag{17}$$

where the two coefficients can be evaluated from

$$\begin{aligned} a &= \frac{4\pi^2}{15}(2f_0 + \lambda g_0)^{-5/2} f_0^{-1} (\lambda g_0)^{-1/2} \\ b &= \frac{\pi^2}{45}(f_0 + 2\lambda g_0)(2f_0 + \lambda g_0)^{-5/2} f_0^{-1} (\lambda g_0)^{-3/2} \\ A &= \frac{1}{2}\{a(4f_0 + \lambda g_0) + b\lambda f_0\} f_0^{-1} (2f_0 + \lambda g_0)^{-1} \\ B &= \{a g_0 + b(f_0 + \lambda g_0)\} g_0^{-1} (2f_0 + \lambda g_0)^{-1} \end{aligned} \tag{18}$$

where f_0 and g_0 are the values of f and g at zero temperature. The numerical results for the two coefficients A and B as functions of the interplanar coupling are shown in figure 2. On one hand, all the coefficients are divergent in the limit of zero interplanar coupling. This property is due to the dimensionality effect, for the interplanar coupling has changed the 2D system into a 3D one, which is consistent with the Mermin–Wagner theorem. On the other hand, the coefficients decrease quickly with increasing interplanar coupling. This indicates that the thermal fluctuations are suppressed by the increasing interplanar coupling, which is just what is expected, for fluctuations are more important in low dimensions. Furthermore, the interplanar coupling has stabilized a Néel ordered state at finite temperature and, at low temperature, the sublattice magnetization also follows a power law (T^2-law):

$$m(T) = m(0) - p(k_B T/J)^2 \tag{19}$$

where $m(0)$ is the zero-temperature magnetization and is given by

$$m(0) = 1 - \frac{1}{N}\sum_{\boldsymbol{q}} \frac{1}{\sqrt{1 - (\Gamma_{\boldsymbol{q}}/\Gamma_{\boldsymbol{0}})^2}}. \tag{20}$$

The interplanar-coupling-dependent coefficient p can be evaluated from

$$p = \frac{1}{12}(2f_0 + \lambda g_0)^{-1/2} f_0^{-1} (\lambda g_0)^{-1/2}. \tag{21}$$

Figure 3 shows the zero-temperature sublattice magnetization as a function of the interplanar coupling (circles). The magnetization increases gradually from 0.303 for the 2D isotropic case to 0.422 for the 3D isotropic case, which indicates that the quantum fluctuation is suppressed with increasing interplanar coupling, consistently with the result derived above. Nevertheless, compared with that calculated from the linear spin-wave theory (triangles), the magnetization increases by a small amount due to the zero-point contribution from the magnon interactions (see figure 3). This enhancement seems to be independent of the interplanar coupling, i.e. it is uniform, although its magnitude varies continuously. This can be understood by assuming that the magnon interactions give a positive contribution to the effective exchange coupling. The coefficient p is shown in figure 2 (triangles). It is divergent in the limit of zero interplanar coupling, similarly to the other coefficients shown in figure 2. It also decreases with the interplanar coupling, but becomes nearly constant when λ is more than 0.2, which indicates again that the thermal fluctuation is suppressed by the increasing interplanar coupling.

Note that the T^2-law that we obtained is in good agreement with the NMR experiments on the AF materials [21–23], and the calculations made by the linear spin-wave approach [26]. Although these materials have different structures, they can all be seen as 2D layered

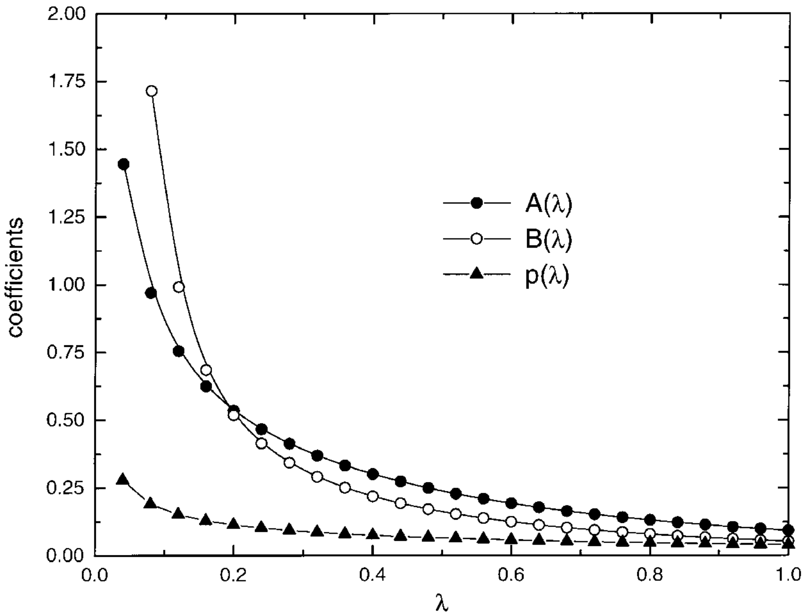

Figure 2. The interplanar coupling dependence of several coefficients. See the details in the text.

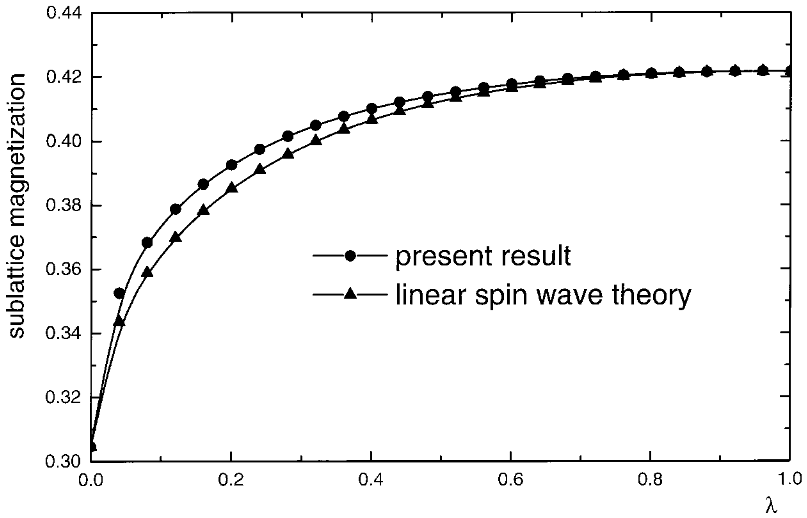

Figure 3. Zero-temperature magnetization versus the interplanar coupling (filled circles). Also shown is the same quantity calculated from the linear spin-wave theory. The solid lines are guides to the eye.

antiferromagnets with different effective interplanar couplings, which is the reason for the similar behaviours (power laws) at finite temperature. This confirms that the T^4-law for the spin-wave excitation is also reasonable at finite temperature for these antiferromagnets. What is important is that in these materials, the power laws survive up to temperature $T_N/2$. This can be easily understood if we recall that the expansions are made in powers of $k_B T/J$, which is rather small as long as T is less than T_N, for $k_B T_N/J \simeq 0.1$–0.2. The advantage of our method is that we give the evolution of the spin-wave excitation and sublattice magnetization with the interplanar coupling in an improved linear spin-wave approach, which will contribute to improvement of the precision of the measurement of coupling constants by NMR or other experiments.

The Néel temperature T_N increases with the interplanar coupling. The gradually changed Néel temperature can be estimated roughly from $m(T_N) = 0$. In the small-coupling limit, we have $T_N \approx 1.243\lambda^{1/4}J$, i.e. the Néel temperature is proportional to one quarter of the interplanar coupling. Thus, although λ can be quite small, the Néel temperature need not be very small. For La_2CuO_4, λ is about 5×10^{-5}, so T_N is estimated to be 150 K, which is qualitatively in agreement with the experimental result $T_N \approx 300$ K for La_2CuO_4.

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