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2011 J. Phys.: Condens. Matter 23 346003

(http://iopscience.iop.org/0953-8984/23/34/346003)

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **23** (2011) 346003 (8pp) doi:10.1088/0953-8984/23/34/346003

Longitudinal excitations in quantum antiferromagnets

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Received 17 May 2011
Published 12 August 2011
Online at stacks.iop.org/JPhysCM/23/346003

Abstract

By extending our recently proposed magnon-density waves to low dimensions, we investigate, using a microscopic many-body approach, the longitudinal excitations of the quasi-one-dimensional (quasi-1d) and quasi-2d Heisenberg antiferromagnetic systems on a bipartite lattice with a general spin quantum number. We obtain the full energy spectrum of the longitudinal mode as a function of the coupling constants in the original lattice Hamiltonian and find that it always has a nonzero energy gap if the ground state has a long-range order and becomes gapless for the pure isotropic 1d model. The numerical value of the minimum gap in our approximation agrees with that of a longitudinal mode observed in the quasi-1d antiferromagnetic compound $KCuF_3$ at low temperature. It will be interesting to compare values of the energy spectrum at other momenta if their experimental results are available.

1. Introduction

The low temperature properties of many two-dimensional (2d) and three-dimensional (3d) quantum antiferromagnetic systems can be understood by Anderson's spin-wave theory (SWT) and its extensions [1], which provide a correct description of the quantum corrections to the classical Néel states of the systems. For many purposes, the dynamics of these systems at low temperature can be considered as that of a dilute gas of weakly interacting spin-wave quasiparticles (magnons) with its density corresponding to the quantum correction to the classical Néel order; also present in these systems are the longitudinal fluctuations consisting of the multi-magnon continuum [2].

On the other hand, due to the strong quantum fluctuations, the isotropic 1d antiferromagnets with low quantum spin numbers exhibit different low temperature properties, such as no Néel-like long-range order in the ground state and quite different low-lying excitation states from 2d and 3d counterparts. According to the exact solutions by the Bethe ansatz, the natural low-lying excitation states of the 1d spin-1/2 Heisenberg model have been shown corresponding to the spin-1/2 objects (spinons) which always appear in pairs, and the spin-wave-like excited states are actually the triplet states of a spinon pair [3], in contrast to the doublet states from SWT. For the spin-1 Heisenberg chain, however, the triplet excitation states have a nonzero energy gap, first predicted by Haldane [4]. These theoretical predictions have been confirmed by the experimental results in the antiferromagnetic compound $KCuF_3$ for the spin-1/2 chains [5] and $CsNiCl_3$ for the spin-1 chains [6].

Strictly, all real systems are 3d when the temperature is low enough. The antiferromagnetic compounds $KCuF_3$ or $CsNiCl_3$ are actually quasi-1d systems with very weak inter-chain couplings. In particular, the spin–spin couplings are ferromagnetic in the tetragonal basal planes of $KCuF_3$ and antiferromagnetic in the hexagonal planes of $CsNiCl_3$. Many parent compounds of the high-T_c superconducting cuprates or the ion-based pnictides are also quasi-2d antiferromagnetic systems with very weak inter-plane couplings [7, 8]. Therefore, there is a 3d magnetic long-range order with a nonzero Néel temperature T_N for all these systems and one expects SWT should provide a qualitatively correct description for some, if not all, the low temperature dynamics of these quasi-1d or quasi-2d systems. One interesting question is whether or not some 1d-type excitations, such as the longitudinal part of the triplet spin-wave excitations of the pure 1d systems, can survive in the ordered phase when the inter-chain couplings are present. In fact, there is now ample evidence of the longitudinal excitation states in various quasi-1d structures with Néel-like long-range order at low temperature, including the hexagonal ABX_3-type antiferromagnets with both spin quantum number $s = 1$ ($CsNiCl_3$ and $RbNiCl_3$) [9, 10] and $s = 5/2$ ($CsMnI_3$) [11,

12] and the tetragonal structure of $KCuF_3$ with $s = 1/2$ [13]. More recently, a longitudinal mode was also observed in the dimerized antiferromagnetic compound $TlCuCl_3$ under pressure with a long-range Néel order [14]. To our knowledge, no observation of any longitudinal mode in the quasi-2d antiferromagnets has been reported yet. Clearly, such longitudinal modes, which correspond to the oscillations in the magnitude of the magnetic order parameter, are beyond the usual SWT which only predicts the transverse spin-wave excitations (magnons). There have been several theoretical investigations in these longitudinal modes, all using the field theory approach, such as a simplified version of Haldane's theory for the spin-1 systems [15] or the sine-Gordon theory for the spin-1/2 systems [16, 17], and both treating the inter-chain couplings as perturbation. A phenomenological field theory approach focusing on the spin frustrations of the hexagonal lattice of the ABX_3-type antiferromagnetic systems has also been proposed [18]. Common to all these field theory approaches is the need to take the continuum limit with a number of fitting parameters. By proper choice for the values of the fitting parameters, general agreements with the experiments mentioned earlier have been found, although there are still some disagreements particularly for the data away from the minimum energy gap at the antiferromagnetic wavevector [12].

We recently proposed a microscopic many-body theory for the longitudinal excitations of spin-s quantum antiferromagnetic systems, using the original spin lattice Hamiltonians [19]. The basic physics in our analysis is simple: by analogy to Feynman's theory on the low-lying excited states of the helium-4 superfluid [20], we identify the longitudinal excitation states in a quantum antiferromagnet with a Néel-like order as the collective modes of the magnon-density waves, which represent the fluctuations in the long-range order and are supported by the interactions between magnons. These longitudinal excitation states are constructed by the s^z spin operators, in contrast to the transverse spin operators $s^{\pm}$ of the magnon states in Anderson's SWT. These modes are referred to as the collective modes of the magnon-density waves because of the fact that s^z is the magnon-density operator in these systems. The energy spectra of these collective modes can be easily derived by a formula first employed by Feynman for the famous phonon–roton spectrum of the helium superfluid involving the structure factor of its ground state. Nevertheless, we now realize that the precise form for the definition of the longitudinal state in our earlier work is not quite correct and we have now slightly modified the definition and, indeed, we find the corresponding values of the energy spectra in an approximation using the SWT ground state are, in general, much lower than before. We extend our analysis to the 1d model and find that in the isotropic limit the longitudinal mode has a gapless spectrum. Interestingly, this gapless spectrum in our approximation is degenerate with the doublet spin-wave spectrum of SWT, hence making it triplet, in good agreement with the exact triplet spin-wave spectrum of the spin-1/2 Heisenberg model [3]. The application of our analysis to the quasi-1d and quasi-2d systems is straightforward and hence a more detailed comparison with the experiments is now possible. Our numerical results for the spin-1/2 quasi-1d compound $KCuF_3$ show the minimum gap value of the longitudinal energy spectrum in agreement with the value obtained from the experiments [13]. This is particularly satisfactory since our analysis has no fitting parameters except the coupling constants in the original lattice Hamiltonian. As our microscopic approach allows us to obtain the full spectrum of the longitudinal mode, it will be interesting to compare the values at other regions if their experimental results are available.

We present our general theory of the magnon-density waves briefly in section 2, with numerical results calculated in an approximation using the SWT ground state for the simple cubic and square lattices and its extension to the 1d models in section 3. We then discuss its application to quasi-1d and quasi-2d systems in section 4, including the quasi-1d compound $KCuF_3$. We summarize our work and discuss possible observations of the longitudinal modes in some quasi-2d systems in section 5. We also discuss the approximations employed in our analysis and their possible improvements in section 5.

2. Magnon-density waves in antiferromagnets

We consider an antiferromagnetic system as described by an N-spin Hamiltonian H on a bipartite lattice. The classical ground state is given by the Néel state with two alternating sublattices a and b, where we assume the spins on the a sublattice all point up in the z direction of the spin space and the spins on the b sublattice all point down. This Néel state describes the perfect two-sublattice long-range order. In this paper, we shall exclusively use index i for the sites of the a sublattice, index j for the sites of the b sublattice, and index l for both sublattices. The excited states are given by the spin-flipped states with respect to the Néel state and are commonly referred to as magnons, the quasiparticles of magnetic systems in general.

The quantum ground state $|\Psi_g\rangle$ of H in general differs from the classical Néel state by a correction, the long-range order is hence reduced. For many purposes, as described by the SWT [1], this quantum correction in most 2d and 3d models is well represented by a gas of magnons whose density ρ directly gives the correction as

$$\langle s_i^z \rangle_g = s - \rho, \tag{1}$$

where s is the spin quantum number, s_i^z is the z component of the spin operator on the lattice site i and the expectation $\langle \cdots \rangle_g$ is with respect to the ground state $|\Psi_g\rangle$. Similarly, $\langle s_j^z \rangle_g = -s + \rho$ for the b sublattice with the same density ρ. Therefore, operators s^z correspond to the magnon-density operators, in contrast to the spin-flip operators $s^{\pm}$ which correspond to the magnon creation/destruction operators. Clearly, there are two types of magnons due to the two sublattice structures. Anderson's SWT can be most simply formulated by bosonizing the two sets of these three spin operators, s^z and $s^{\pm}$, on the two sublattices, respectively. For

example, the quantum correction to the classical Néel state by the linear SWT gives the magnon density of $\rho = 0.078$ per lattice site for the spin-1/2 Heisenberg model on a simple cubic lattice, and of $\rho = 0.197$ per lattice site for the same model but on a square lattice.

Due to the interactions between the magnons, it may be necessary to consider the states of the magnon-density waves (MDW). These states may not be well defined in the 3d systems where the magnon density is very dilute and the long-range order is near perfect with little fluctuations. In the low-dimensional systems. However, the magnon density may be high enough to support these longitudinal waves. In terms of microscopic many-body theory, these MDW states are the longitudinal excitation states constructed by applying the density operator s^z on the ground state in a form such as $s^z|\Psi_g\rangle$, similar to Feynman's theory of the phonon–roton excitation state of the helium superfluid, where the density operator is the usual particle-density operator [20]. These longitudinal states may be compared to the quasiparticle magnon states which are constructed by the transverse spin-flip operators as $s^{\pm}|\Psi_g\rangle$. The above discussion underlines the main idea in our earlier papers [19], whose main purpose is to outline a general framework for the excitation states of both quasiparticles and quasiparticle-density waves for a general quantum many-body system in our variational coupled-cluster method [22].

In more detail, following Feynman, the MDW excitation state with momentum $\mathbf{q}$ in an antiferromagnet is given by

$$|\Psi_q^a\rangle = X_q^a|\Psi_g\rangle, \tag{2}$$

where excitation operator X_q^a, in the linear approximation, is the sublattice Fourier transformation of the magnon-density operators s_i^z of the a sublattice:

$$X_q^a = \sqrt{\frac{2}{N}}\sum_i e^{i\mathbf{q}\cdot\mathbf{r}_i}s_i^z, \qquad q > 0, \tag{3}$$

with condition $q > 0$ required because of its orthogonality to the ground state $|\Psi_g\rangle$ in which $s^z_{\text{total}} = 0$. The excitation energy spectrum in this linear approximation can be derived as

$$E^a(q) = \frac{N^a(q)}{S^a(q)}, \tag{4}$$

where $N^a(q)$ is given by a double commutator:

$$N^a(q) = \tfrac{1}{2}\langle[X^a_{-q}, [H, X^a_q]]\rangle_g, \tag{5}$$

and the state normalization integral $S^a(q)$ is, in fact, the structure factor of the a sublattice:

$$S^a(q) = \langle X^a_{-q}X^a_q\rangle_g = \frac{2}{N}\sum_{i,i'} e^{i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_{i'})}\langle s_i^z s_{i'}^z\rangle_g . \tag{6}$$

Similarly, we have the MDW excitation state $X_q^b|\Psi_g\rangle$ with the operator

$$X_q^b = \sqrt{\frac{2}{N}}\sum_j e^{i\mathbf{q}\cdot\mathbf{r}_j}s_j^z, \qquad q > 0, \tag{7}$$

and the corresponding energy spectrum $E^b(q)$ for the b sublattice. Due to the lattice symmetry, the spectra E_q^a and E_q^b are degenerate. However, these two excitation states are not orthogonal to each other because of the couplings between the spins on the a sublattice and the spins on the b sublattice. We therefore need to consider their linear combinations as

$$|\Psi_q^{\pm}\rangle = X_q^{\pm}|\Psi_g\rangle = \frac{1}{\sqrt{2}}(X_q^a \pm X_q^b)|\Psi_g\rangle, \tag{8}$$

for the coupled MDW states. The corresponding energy spectra is similarly given by $E^{\pm}(q) = N^{\pm}(q)/S^{\pm}(q)$ with $N^{\pm}(q)$ and $S^{\pm}(q)$ given by similar equations to equations (5) and (6), respectively, using excitation operators $X_q^{\pm}$ instead of X_q^a. It seems that we have two longitudinal modes. But these two states $|\Psi_q^{\pm}\rangle$ with the energy spectra $E^{\pm}(q)$ are actually the same state, with one equal to another by a substitution for the wavevector $\mathbf{q} \to \mathbf{q} + \mathbf{Q}$, where $\mathbf{Q}$ is the antiferromagnetic wavevector of the system (e.g. $\mathbf{Q} = (\pi, \pi)$ for the 2d square lattice model). This can be easily seen as the excitation operator $X_q^{\pm}$ are, in fact, nothing but the Fourier transformations of the (staggered) magnon-density operators s_l^z or $(-1)^l s_l^z$, respectively. We therefore only need to consider one of them. We choose $|\Psi_q^-\rangle = |\Psi_q\rangle$ with its energy spectrum $E^-(q) = E(q)$, and write

$$E(q) = \frac{N(q)}{S(q)}, \tag{9}$$

where $N(q)$ and $S(q)$ are calculated by using $X_q^- = X_q$ of equation (8). We notice the slight difference between this definition of the MDW states of equation (8) and that in our earlier paper [19], where we used the total density operator as $\hat{n}_i = \frac{1}{2}(2s - s_i^z + \frac{1}{z}\sum_n s_{i+n}^z)$ with z as the coordination number and n as the nearest-neighbor index. We now realize the use of the operator $\hat{n}_i$ (or its equivalent form, $s_i^z - \frac{1}{z}\sum_n s_{i+n}^z$) is not quite correct. Our current definition of equation (8) seems more natural as discussed in detail above. Indeed, as we will see later, the values of the energy spectrum of the states defined by equation (8) in our approximation are, in general, much lower than before, with the maximum energy values about half of those of the earlier results [19].

So far, in the above general analysis for the longitudinal excited states, the exact ground state $|\Psi_g\rangle$ is used for the ground state expectation values. The only approximation comes from the choice of the linear form in the excitation operators of equations (3) and (7), and is often referred to as the single-mode approximation as viewed from the general expression of the dynamic structure factor. In the case of the helium superfluid, the double commutator can be simply evaluated as $N(q) \propto q^2$, and Feynman [20] used the experimental results for the structure factor with $S(q) \propto q$ as $q \to 0$ and hence derived the low-lying phonon spectrum $E(q) \propto q$ and the gapped roton spectrum around the peak of $S(q)$. Jackson and Feenberg, however, used the variational results calculated from the Jastrow-type wavefunctions and obtained similar results [21]. In our earlier papers [19], we have demonstrated that these equations remain valid when the exact ground state $|\Psi_g\rangle$ is replaced by a variational state $|\Psi_0\rangle$ and furthermore, in the case of the quantum antiferromagnets

as discussed here, our variational ground state $|\Psi_0\rangle$ by the so-called variational coupled-cluster method in a first-order approximation reduces to that of Anderson's SWT [22]. Therefore, to this first-order approximation which is what we focus on here, we apply the SWT ground state $|\Psi^{\rm sw}\rangle$ in all of our following calculations. We like to emphasize that SWT itself in its usual form cannot produce the longitudinal MDW excitations discussed here. We will discuss this approximation and its possible improvement in section 5. We present our numerical results of the MDW spectra $E(q)$ for several models in the following two sections: section 3 contains the results for the antiferromagnetic models on the simple lattices, while section 4 contains results for the more physical quasi-1d and quasi-2d systems.

3. Results of magnon-density wave spectra in simple lattices

3.1. The spin-s XXZ Heisenberg model

In this section, we present the numerical results for the energy spectra of the MDW states as discussed in the earlier section for the spin-s XXZ Heisenberg model on a simple cubic lattice and a square lattice. We then present the results for the 1d model and discuss the convergent results in its isotropic limit.

The spin-s XXZ Heisenberg model on a bipartite lattice is given by

$$H = J\sum_{i,n}[\tfrac{1}{2}(s_i^+ s_{i+n}^- + s_i^- s_{i+n}^+) + A s_i^z s_{i+n}^z], \tag{10}$$

where the coupling parameter $J > 0$, index i runs over all a sublattice only, index n runs over the z nearest-neighbor sites and A ($\geq$1) is the anisotropy parameter. The usual isotropic Heisenberg model is given by $A = 1$. The purpose of introducing the anisotropy is twofold: it is interesting in its own right and it also provides a way to obtain convergent results for the 1d case in the isotropic limit as we will see later.

Using the usual spin commutation relations, it is straightforward to derive the following double commutator as

$$N(q) = \frac{1}{2}\langle[X_{-q},[H,X_q]]\rangle_{\rm g} = -\frac{zJ}{2}(1+\gamma_q)\langle s_i^+ s_{i+1}^-\rangle_{\rm g}, \tag{11}$$

where γ_q is defined as usual:

$$\gamma_q = \frac{1}{z}\sum_n {\rm e}^{{\rm i}\mathbf{q}\cdot\mathbf{r}_n}, \tag{12}$$

with the coordination number z and $\langle s_i^+ s_{i+1}^-\rangle_{\rm g}$ is independent of the index i due to the lattice translational symmetry. The general expression for the structure factor $S(q)$ contains an additional cross-term compared to the sublattice counterpart $S^a(q)$ as

$$S(q) = \langle X_{-q}X_q\rangle_{\rm g} = S^a(q) + \frac{2}{N}\sum_{i,j}{\rm e}^{{\rm i}\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)}\langle s_i^z s_j^z\rangle_{\rm g}. \tag{13}$$

Before we discuss any approximation, we notice that the double commutator in general behaves as, near the antiferromagnetic wavevector $\mathbf{Q}$:

$$N(|\mathbf{q}+\mathbf{Q}|) \propto q^2, \qquad q\to 0, \tag{14}$$

similar to that of the helium superfluid [20].

Now we need a specific approximation for the ground state $|\Psi_{\rm g}\rangle$ in order to evaluate the spin correlation functions $\langle s_i^+ s_j^-\rangle_{\rm g}$, $\langle s_i^z s_{i'}^z\rangle_{\rm g}$ and $\langle s_i^z s_j^z\rangle_{\rm g}$. As mentioned earlier, in this paper we use as our first-order approximation the spin-wave ground state, $|\Psi^{\rm sw}\rangle$, for these calculations. After defining the transverse spin correlation function $\tilde{g}(r)$:

$$\tilde{g}_r = \frac{1}{2s}\langle s_i^+ s_{i+r}^-\rangle_{\rm g}, \tag{15}$$

we derive the following results for its Fourier transformation:

$$\tilde{g}_q = -\frac{1}{2}\frac{\gamma_q/A}{\sqrt{1-\gamma_q^2/A^2}}, \tag{16}$$

and the sublattice structure factor:

$$\begin{aligned} S^a(q) &= \rho + \sum_{q'}\rho_{q'}\rho_{q-q'},\\ \rho_q &= \frac{1}{2}\frac{1}{\sqrt{1-\gamma_q^2/A^2}} - \frac{1}{2} \end{aligned} \tag{17}$$

with magnon density $\rho = \sum_q \rho_q$. And, finally, the full-lattice (staggered) structure factor is given by

$$S(q) = S^a(q) + \sum_{q'}\tilde{g}_{q'}\tilde{g}_{q-q'}. \tag{18}$$

We notice that, in deriving the expressions of equations (17) and (18) for the structure factors, the values for $q = 0$ are excluded due to the condition $q > 0$ in the definition of X_q from equations (3) and (7). Furthermore, the integrals in the structure factor involving the function $\gamma_{q'}\gamma_{q-q'}$ clearly indicate the couplings between magnons. In all these formulae, the summation over q is given by

$$\sum_q = \frac{1}{(2\pi)^d}\int_{-\pi}^{\pi}{\rm d}^d q, \tag{19}$$

where d is the dimensionality of the system. The energy spectrum $E(q)$ of equation (9) is obtained by calculating the values for $N(q)$ and $S(q)$ from the approximations of equations (16)–(18). This longitudinal spectrum $E(q)$ can be compared with the following transverse spin-wave spectra of the linear SWT [1]:

$$E^{\rm sw}(q) = szJA\sqrt{1-\gamma_q^2/A^2}. \tag{20}$$

In the following subsections, we present numerical results using the above approximations.

3.2. Results for the simple cubic and square lattices

We first consider the isotropic case $A = 1$ for the simple cubic lattice model for which $\tilde{g}_1$ is calculated as $\tilde{g}_1 = \langle s_i^+ s_{i+1}^-\rangle_{\rm g} \approx -0.13$. The numerical values for $E(q)$ near $q\to 0$

are similar to those given in [19], with a large energy gap of about $0.99szJ$ at $q = 0$. But at other values of q, the energies are much smaller than before due to the different definitions of the density operator of equation (8).[1] At the antiferromagnetic wavevector (AFWV) $\mathbf{Q} = (\pi, \pi, \pi)$, the spectrum has a larger gap of $1.40szJ$. As discussed before, this high energy 3d longitudinal mode may not be well defined and distinguishable from the multi-magnon continuum.

For the square lattice model at the isotropic point $A = 1$, $\tilde{g}_1 \approx -0.28$. Similar to the earlier results [19], $E(q)$ becomes gapless at both AFWV $\mathbf{Q} = (\pi, \pi)$ and $q \to 0$, due to the logarithmic behaviors from the structure factors (e.g. $S(q) \to -\ln q$, hence $E(q) \propto -1/\ln q$ as $q \to 0$). However, as discussed earlier, this logarithmic gapless spectrum of the square lattice model, in fact, is quite 'hard' in the sense that any finite-size effect, anisotropy or inter-plane coupling to be discussed later, however small, will make a nonzero gap. For example, we consider a tiny anisotropy here with a value $A - 1 = 1.5 \times 10^{-4}$, which in fact is a typical value for the parent compound of the high-T_c superconducting cuprate, La_2CuO_4 [23]. We obtain in our approximation the gap values at $E(Q) \sim 0.76szJ$ and $E(q) \sim 0.44szJ$ as $q \to 0$, both much larger than the corresponding magnon gap value of $0.02szJ$ from equation (20). We plot part of the spectrum with this anisotropy in figure 1, together with the spin-wave spectrum for comparison. The energy values at the two particular momenta $(\pi/2, \pi/2)$ and $(\pi, 0)$ deserve attention, where $\gamma_q = 0$ and the spin-wave spectrum gives the same value of szJ. The longitudinal spectrum $E(q)$ at these two point has slightly different values, $1.36szJ$ and $1.40szJ$, respectively. The energy difference at these two momenta has been used to indicate nonlinear effects due to magnon–magnon interactions in the more accurate calculations for the isotropic Heisenberg model [24]. It is interesting to note that our longitudinal mode also shows this difference.

3.3. Results for the 1d model

We next consider the 1d case. The SWT results in general for the isotropic 1d case are not reliable as most integrals suffer from the well-known infrared divergence, e.g. the magnon density $\rho \to \infty$ as $A \to 1$, an unphysical result. Nevertheless, the value of the spin-wave spectrum of equation (20) is not far off that of the exact result by the Bethe ansatz [3] for the spin-1/2 model despite the different degeneracies (i.e. the spin-wave spectrum is doublet while the exact spectrum is triplet). Infrared divergence of the spin-wave results also occurs for the parameter $\tilde{g}_1$ in the numerator of the energy spectra in equation (9). We examine the behaviors of each integral in $N(q)$ and $S(q)$ in the isotropic limit $A \to 1$ and find that they all have similar infrared divergence. For example, by numerical calculations, we find that

$$\tilde{g}_1 \propto -\frac{1}{2\pi}\ln(A-1), \qquad \text{as } A \to 1, \tag{21}$$

[1] The integrals over momentum space have bounds between $(-\pi, \pi)$ as given by equation (19). In [19], the bounds of $(0, \pi)$ were used by mistake which makes no difference for most integrals but a small difference for integrals involving $\gamma_{q'}\gamma_{q-q'}$.

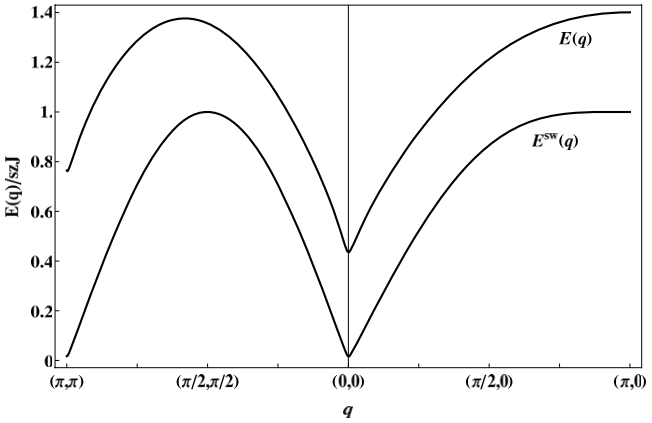

Figure 1. The energy spectrum $E(q)$ for the longitudinal mode of equation (9) for the square lattice Heisenberg model of equation (10) with an anisotropy $A - 1 = 1.5 \times 10^{-4}$, together with the linear spin-wave spectrum $E^{sw}(q)$ of equation (20) for comparison. This tiny anisotropy is a typical value for the parent compound of the high-T_c superconducting cuprate, La_2CuO_4.

agrees with the analytical results using the elliptical formula [25]. Furthermore, in the limit $q \to 0$, both $S^a(q)$ and $S(q)$ behave as

$$S(q) \to -\frac{1}{2\pi}\frac{\ln(A-1)}{\sqrt{A-1}}, \qquad \text{as } q \to 0 \quad \text{and} \quad A \to 1. \tag{22}$$

Since the divergences in the numerator $N(q)$ and the denominators $S(q)$ precisely cancel out, we obtain finite results for the energy spectrum $E(q)$ for the isotropic 1d model. Interestingly, we find that these numerical values of $E(q)$ coincide precisely with those of the linear spin-wave spectra of equation (20) for all values of q in the isotropic limit $A \to 1$. Therefore, our longitudinal spectrum and the doublet transverse spin-wave spectrum constitute a triplet, in good agreement with the following exact triplet spectrum for the spin-1/2 model by the Bethe ansatz first derived by des Cloizeaux and Pearson [3]:

$$E(q) = \frac{\pi J}{2}\sin q. \tag{23}$$

The different factor $\pi J/2$ of the above exact result compared to the value of J by the linear SWT of equation (20) with $z = 2$ clearly comes from the nonlinear effects beyond our simple approximation employed here. We also notice that our analysis here in the approximations employed is not able to produce the Haldane gap for the isotropic spin-1 chain.

For the anisotropic 1d model (i.e. $A > 1$), the triplet spectra split and the values of the longitudinal spectrum $E(q)$ are larger than those of the doublet spin-wave spectrum, similar to the cases of the 2d and 3d models discussed earlier. We plot this $E(q)$ for $A = 1.1$ in figure 2 as an example. The gaps for $E(q)$ are about $1.16szJ$ and $1.64szJ$ at $q = 0$ and π, respectively, comparing with $0.46szJ$ of the spin-wave spectrum at both points.

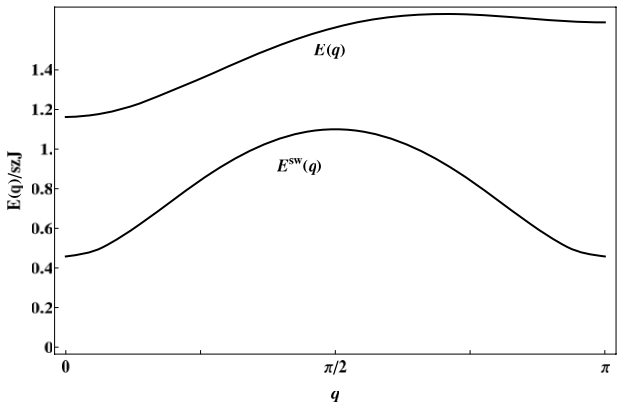

Figure 2. Similar to figure 1 but for the 1d model with anisotropy $A = 1.1$. In the isotropic limit of $A = 1$, $E(q)$ approaches $E^{\rm sw}(q)$, forming a triplet spectrum as described in detail in the text.

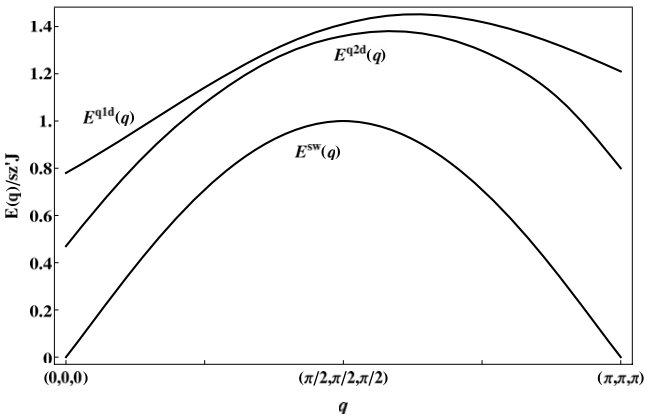

Figure 3. Similar to the earlier figures but for the quasi-1d and quasi-2d systems of equation (24), with parameter $\xi = J_\perp/J = 0.01$ for the quasi-1d spectrum $E^{\rm q1d}(q)$ and $\xi = 10^3$ for the quasi-2d spectrum $E^{\rm q2d}(q)$. The spin-wave spectrum $E^{\rm sw}(q)$ is for the quasi-1d model.

4. Magnon-density waves in quasi-1d and quasi-2d systems

4.1. Quasi-1d and quasi-2d antiferromagnets on bipartite lattices

A generic quasi-1d and quasi-2d antiferromagnetic Hamiltonian on a bipartite lattice is given by

$$H = J\sum_{i,n_1} \mathbf{s}_i \cdot \mathbf{s}_{i+n_1} + J_\perp \sum_{i,n_2} \mathbf{s}_i \cdot \mathbf{s}_{i+n_2}, \tag{24}$$

where index i as before runs over all a-sublattice sites with index n_1 over the nearest-neighbor sites along the chains and n_2 over the nearest-neighbor sites on the basal planes, J is the coupling constant along the chains and $J_\perp$ is the counterpart on the basal planes. We consider the model with both J and $J_\perp > 0$. The quasi-1d model corresponds to the case of $J_\perp/J \ll 1$, the quasi-2d model to the case of $J_\perp/J \gg 1$ and the 3d model is given by $J_\perp = J$. This Hamiltonian has been studied for the case of the quasi-1d systems with $J_\perp/J \ll 1$ by SWT [25, 26]. In particular, the SWT ground state was used to evaluate the corrections due to the kinematic interactions to the order parameter ρ. The longitudinal modes were not discussed.

All of our earlier formulae for the longitudinal mode at the isotropic point $A = 1$ remain the same after the following replacements:

$$\begin{aligned} z &\to z' = 2(1+2\xi), \\ \gamma_q \to \gamma'_q &= \frac{2}{z'}\left[\cos q_z + \xi(\cos q_x + \cos q_y)\right], \end{aligned} \tag{25}$$

where $\xi = J_\perp/J$. This is true also for the spin-wave spectrum $E^{\rm sw}(q)$ of equation (20). We notice that the spin-wave spectrum is gapless at zone boundaries. The longitudinal mode $E(q)$ of equation (9), however, has nonzero gaps for any $\xi > 0$, at which there is a long-range order [25, 26]. In figure 3, we present our results for the spectrum, denoted as $E^{\rm q1d}(q)$, of the quasi-1d model with $A = 1$ and $\xi = 1.05$ as an example, together with the spin-wave spectrum $E^{\rm sw}(q)$. The gaps for $E^{\rm q1d}(q)$ at $q \to 0$ and $\mathbf{Q} = (\pi, \pi, \pi)$ are $0.78sz'J$ and $1.21sz'J$, respectively. Figure 3 also includes our results for a quasi-2d model with $A = 1$ and $1/\xi = J/J_\perp = 10^{-3}$, denoted as $E^{\rm q2d}(q)$. The gap values for this quasi-2d spectrum at $q \to 0$ and $\mathbf{Q} = (\pi, \pi, \pi)$ are about $0.47sz'J$ and $0.80sz'J$, respectively. We also notice that at the particular two momenta $(\pi/2, \pi/2, 0)$ and $(\pi, 0, 0)$, where the linear spin-wave spectrum has the same value of $sz'J$ but the longitudinal mode has slightly different values, $1.36sz'J$ and $1.41sz'J$, respectively, due to magnon–magnon interactions as discussed earlier. This quasi-2d model may be relevant to the parent compounds of the high-T_c cuprates, where the effective interlayer couplings $J/J_\perp$ between the CuO_2 planes are estimated to be between 10^{-2} and 10^{-5} [27].

4.2. Quasi-1d model with $KCuF_3$ structure

In the experimentally well-studied quasi-1d compound $KCuF_3$, strong spin couplings along the chains are antiferromagnetic but weak couplings on the basal plane are ferromagnetic. This compound can be described by the following Hamiltonian model:

$$\begin{aligned} H = {} & J\sum_{l_a,n_1} \mathbf{s}_{l_a} \cdot \mathbf{s}_{l_a+n_1} \\ & - \frac{J_\perp}{2}\left(\sum_{l_a,n_2} \mathbf{s}_{l_a} \cdot \mathbf{s}_{l_a+n_2} + \sum_{l_b,n_2} \mathbf{s}_{l_b} \cdot \mathbf{s}_{l_b+n_2}\right), \end{aligned} \tag{26}$$

whose classical Néel state consists of two alternating planes, with all the spins on the a plane pointing up and labeled by index l_a and all the spins on the b plane pointing down and labeled by index l_b. In equation (26), the nearest-neighbor indices n_1 and n_2 are as defined before with n_1 along the chains and n_2 on the basal planes, and both J and $J_\perp > 0$. The spin-wave spectrum is derived as

$$E^{\rm sw}(q) = 2sJ\sqrt{\Delta_q^2 - \cos^2 q_z}, \tag{27}$$

where Δ_q is defined as

$$\Delta_q = 1 + 2\xi(1 - \gamma_q^{2d}), \qquad \xi = \frac{J_\perp}{J}, \tag{28}$$

with $\gamma_q^{2d} = (\cos q_x + \cos q_y)/2$. It is easy to check that, when $J = 0$, we recover the spin-wave spectrum of the 2d ferromagnetic model and that, when $J_\perp = 0$, we recover the spin-wave spectrum of the 1d antiferromagnetic model. For the longitudinal energy spectrum $E(q)$ of equation (9), the double commutator is now given by a different form as

$$N(q) = -J(1 + \cos q_z)\,\langle s_{l_a}^+ s_{l_a+n_1}^- \rangle_g + 2J_\perp(1 - \gamma_q^{2d})\,\langle s_{l_a}^+ s_{l_a+n_2}^- \rangle_g, \tag{29}$$

and the structure factor $S(q)$ is as given before by equations (6) and (13) in general forms and by equations (17) and (18) in our approximation using the similar SWT ground state with the anisotropy parameter $A = 1$. We notice that, in equation (29), the two spin operators in the first correlation function $\langle s_{l_a}^+ s_{l_a+n_1}^- \rangle_g$ are from the two sublattices, respectively, as before, but in the second correlation function $\langle s_{l_a}^+ s_{l_a+n_2}^- \rangle_g$, they are from the same sublattice. So we still name the first one as before by $\tilde{g}_1 = \langle s_{l_a}^+ s_{l_a+n_1}^- \rangle_g /2s$ but the second one as $\tilde{g}_1' = \langle s_{l_a}^+ s_{l_a+n_2}^- \rangle_g /2s$. Using the similar SWT ground state, we obtain for their Fourier transformations

$$\tilde{g}_q = -\frac{1}{2}\frac{\cos q_z}{\sqrt{\Delta_q^2 - \cos^2 q_z}} \tag{30}$$

and

$$\tilde{g}_q' = \rho_q = \frac{1}{2}\frac{\Delta_q}{\sqrt{\Delta_q^2 - \cos^2 q_q}} - \frac{1}{2}, \tag{31}$$

respectively. We notice the quite different expressions for $\tilde{g}_q$ and $\tilde{g}_q'$ as expected. We present our numerical results for $E(q)$ in figure 4, together with $E^{sw}(q)$ of equation (27) for comparison, using the experimental values for the coupling constants, $J \approx 34$ meV, $J_\perp \approx 1.6$ meV and $s = 1/2$ [13]. Different to the longitudinal modes in other systems discussed earlier, we find that $E(q)$ has a smaller gap of about $0.63J \approx 21.4$ meV at AFWV $\mathbf{Q} = (0, 0, \pi)$ and a larger gap of about $0.85J \approx 28.9$ meV at $q \to 0$. This gap value of 21.4 meV at AFWV is about 40% higher than the experimental value of about 15 meV. The field theory by Essler *et al* produces a gap value of about 17.4 meV [17]. However, there is uncertainty in the estimated value of the inter-chain coupling constant $J_\perp$. Lake *et al* seem to have used the theoretical formula (56) in [17] to obtain $J_\perp = 1.6$ meV $= 0.047J$. By different methods [29, 30], $J_\perp$ was estimated to be $0.01J \sim 0.016J$. Using this estimate of $\xi = J_\perp/J = 0.01$, we obtain the minimum gap value of 11.9 meV at AFWV and 16.8 meV at $q = 0$. Naively, if we choose about the midpoint between the values of [13] and [29], $J_\perp = 0.85$ meV with $\xi \approx 0.025$, we obtain the minimum gap value of $0.49J = 16.8$ meV at AFWV and $0.68J = 23.2$ meV at $q = 0$, in good agreement with the experiment for the minimum gap [13]. Furthermore, with this value of $J_\perp = 0.85$ meV, the linear spin-wave spectrum gap at $\mathbf{q} = (\pi, 0, \pi)$ is $E^{sw}(q) \approx 0.32J = 10.9$ meV, very close to the gap value of 11 ± 0.5 meV by the experiment [29]. The longitudinal mode $E(q)$ is nearly flat in the region $(\eta, 0, \pi) \sim (\eta, 0, \pi)$ with $\pi \leq \eta \leq 0$, with the gap value about $0.59J = 20.1$ meV at $(\pi, 0, \pi)$. It will be very interesting indeed to compare with experimental results if available for the whole spectrum.

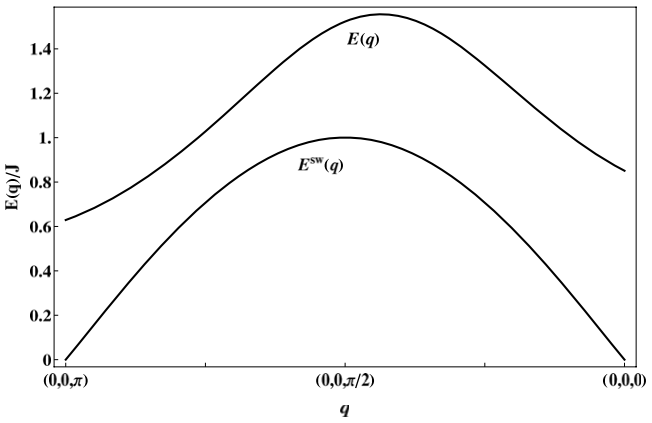

Figure 4. Similar to figure 3 but for the quasi-1d structure of $KCuF_3$ as described by the Hamiltonian of equation (26), with parameter $\xi = J_\perp/J = 1.6/34$ from the experiment [13]. The spin-wave spectrum $E^{sw}(q)$ is given by equation (27).

5. Summary and discussion

In summary, we have investigated the longitudinal excitations of various quantum antiferromagnets based on our recently proposed magnon-density waves. Our numerical results show that the longitudinal mode always has a nonzero gap so long as the system has a Néel-type long-range order and becomes gapless in the limit of the 1d isotropic model. In particular, the spectrum of the longitudinal mode in our approximation is degenerate with the doublet spin-wave spectrum of SWT in the limit of the isotropic 1d model, in agreement with the triplet spin-wave spectrum of exact results for the spin-1/2 model by the Bethe ansatz [3]. In the case of the simple cubic lattice model, the longitudinal mode with high energy values may not be well defined since there is little fluctuation in the nearly perfect classical long-range order. In the quasi-1d and quasi-2d models, where the quantum correction is large and the magnon density is significant, the magnon-density waves may be observable. Indeed, there is now ample evidence of the longitudinal modes in several quasi-1d systems as mentioned earlier in section 1. In particular, for the quasi-1d compound $KCuF_3$, our value for the minimum gap is in agreement with the experimental value [13]. It will be interesting if more experimental results for the spectrum away from the minimum are available for comparison.

It is also interesting to note that the longitudinal modes were observed in the ABX_3-type antiferromagnets with both $s = 1$ [9, 10] and $s = 5/2$ [11, 12], clearly indicating that the modes are more general in their physics, independent of the mechanism which generates the Haldane gap of the 1d model. The phenomenological field theory model with

five fitting parameters employed by Affleck is derived from Haldane's theory of the spin-1 chain [15]. It will be interesting to apply our general microscopic analysis presented here to the ABX_3-type antiferromagnets where the basal plane is hexagonal and the corresponding Néel-like state has three sublattices rather than two sublattices discussed here. Other systems where we can apply our analysis for the magnon-density waves include the quasi-2d systems where the next-nearest-neighbor antiferromagnetic couplings, in addition to the usual nearest-neighbor couplings, are present. These additional couplings cause quantum frustrations and the Néel-like order is further reduced, hence the greater the magnon density to support the magnon-density waves. Of particular current interest is the parent compounds of the newly discovered high-T_c superconducting iron-based pnictides where such next-nearest couplings are believed to be significant [28].

Finally, we want to point out that there are two major approximations in our analysis here. The first is the linear operators X_q employed in constructing the excitation states and the second is the SWT ground state $|\Psi_{sw}\rangle$ employed in evaluating all the correlation functions involved. In regard to the first approximation, it is interesting to consider the case of the phonon–roton spectrum of the helium superfluid, where after inclusion of the nonlinear terms due to the couplings to the low-lying phonons (i.e. the so-called backflow correction), the values of the roton gap are reduced by about half to near the experimental values [20, 21]. Clearly, the effects due to the couplings between the longitudinal modes and the gapless magnons in the antiferromagnetic systems also deserve further investigation. In regard to the second approximation, i.e. the SWT ground state employ in our calculations, improvement can be obtained by using better ground state functions available by more sophisticated microscopic many-body theories such as the coupled-cluster method [22, 31] and, particularly, its most recent extension where the strong correlations are included by a Jastrow correlation factor [32]. We believe the quasi-1d and quasi-2d antiferromagnetic systems as studied here are good theoretical models from both the viewpoint of the field theory approach which deal with the nonlinear effects of the 1d systems [15–17] most effectively and of the microscopic many-body theory approach which provides general, systematic techniques in dealing with many-body correlations in a plethora of quantum systems [33]. The two theoretical approaches complement one another in the study of these models and we wish to report our progress in these investigations in the near future.

Acknowledgment

Useful discussion with P Mitchell and Ch Rüegg is acknowledged.

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