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Upward renormalization of the antiferromagnetic mode in the paramagnetic phase of $CsMnBr_3$

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We study the renormalization of the excitation energy in the paramagnetic phase of a hexagonal Heisenberg model with strong exchange interaction along the c axis and weak exchange interaction in the c plane by a Green function approach. We compare our results with experiments of inelastic neutron scattering on $CsMnBr_3$, a quasi-one-dimensional antiferromagnet with $S=5/2$. The upward renormalization of the antiferromagnetic mode observed in a neutron scattering experiment in the paramagnetic phase is obtained by our approach which is based on the absence of long-range order. On the contrary, downward renormalization of the long-wavelength modes is predicted in agreement with experimental data on $CsMnBr_3$.

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It is well known that an increase of temperature causes downward renormalization of the elementary excitation energy of a magnetic system in the ordered phase. It is interesting to check whether the same occurs in the disordered phase but the quick increase of the energy width makes the experiment usually very hard.

An attempt was made a long time ago to perform inelastic neutron scattering on a single crystal of Praseodymium, a rare-earth metal without long-range order above $T=0.4$ K (Ref. 1). The experiment showed upward renormalization of the magnetic excitations at increasing temperature above $T=7$ K. A self-consistent random phase approximation (RPA) that accounted for the strong crystal field anisotropy and the long-range nature of the spin-spin interactions described well the observed upward renormalization of the magnetic excitation energy.[2] The dominant character of the crystal field potential in rare-earth metals implies the use of a standard basis operator representation[3] or the use of spin-tensor-operator Green functions.[4]

Only recently was high-resolution inelastic neutron scattering made to investigate the thermal renormalization of the magnetic excitations in the paramagnetic phase of compounds of the ABX_3 family where A is an alkali element, B a transition-metal ion, and X a halogen. Indeed the experimental data on $CsNiCl_3$ (Ref. 5) and $CsMnBr_3$ (Ref. 6) have shown that the energy width of the excitations remains small compared to the excitation energy so that measurements can be pushed well beyond the critical temperature. A common feature of the ABX_3 compounds is their quasi-one-dimensional character due to the interchain coupling which is more than two orders of magnitude weaker than the intrachain coupling. Moreover, the single-ion easy-axis or easy-plane anisotropy is of the same order of magnitude as the interchain coupling, so that the ABX_3 compounds may be regarded as a good realization of quasi-one-dimensional Heisenberg antiferromagnets. The interchain exchange and single-ion anisotropy play a key role in determining the kind of long-range order (LRO) characterizing the phase below the critical temperature T_N which is of few kelvin degrees for the most part of the ABX_3 compounds.

Finally, the variety of the spin exhibited by the magnetic ions of the ABX_3 family—for instance, $S=1$ for $B=\mathrm{Ni}, S=3/2$ for $B=\mathrm{V}$, and $S=5/2$ for $B=\mathrm{Mn}$—makes these compounds good candidates to check the Haldane conjecture[7] according to which the integer-spin antiferromagnetic Heisenberg chains have a gapped excitation spectrum, while the half-integer-spin chains have ungapped spectra. The upward renormalization of short-wavelength modes observed in $CsNiCl_3$ (Ref. 5) for $T>T_N=4.84$ K was traced back to the existence of the Haldane gap because of the integer-spin value on the Ni ion. However, an analogous upward renormalization observed in $CsMnBr_3$ (Ref. 6) for $T>T_N=8.3$ K cannot be explained in the same way because of the half-integer value of the spin on the Mn ion. Any attempt to explain this scenario by the renormalized spin-wave theory was in vain since any renormalized spin-wave theory applied to the Heisenberg Hamiltonian even taking dipolar long-range interactions into account[6] provides downward renormalization for any wave vector.

Here we present an extension to a three-dimensional Heisenberg model with a generic S of an approach developed a long time ago[8] for a Heisenberg chain with $S=1/2$. Our approach is very sound for $T>T_N$ and cannot be pushed into the ordered phase because it is based on the absence of LRO. The results we obtain agree very well with the upward renormalization of the modes $(\xi,\xi,1)$ at increasing temperature in $CsMnBr_3$. Indeed a good overall agreement for all wave vectors tested in experiment is achieved. In particular, a downward renormalization at increasing temperature is found for wave vectors $(0,0,\zeta)$ with $\zeta \lesssim 2/3$ in contrast to an upward renormalization found for $\zeta \gtrsim 2/3$. We stress that this scenario differs from that observed in Praseodymium where upward renormalization was found for all investigated wave vectors.

The Hamiltonian for the Heisenberg model is

$$\mathcal{H} = -\sum_{j,\boldsymbol{\delta}} J_{\boldsymbol{\delta}} \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+\boldsymbol{\delta}}, \tag{1}$$

where $J_{\boldsymbol{\delta}}$ is the exchange interaction between spins $\boldsymbol{\delta}$ apart. Periodic boundary conditions are assumed. The Green function equation of motion reads

$$\omega\langle\langle S_i^+;S_0^-\rangle\rangle = \frac{1}{\pi}\delta_{i,0}\langle S_0^z\rangle + \langle\langle [S_i^+,\mathcal{H}];S_0^-\rangle\rangle, \tag{2}$$

where the angular momentum operators $S_i^{\pm}=S_i^x \pm iS_i^y$ satisfy the commutation rules $[S_i^+,S_j^-]=2S_i^z\delta_{i,j}$ and $[S_i^{\pm},S_j^z] = \mp S_i^{\pm}\delta_{i,j}$. The frequency-dependent Green function $\langle\langle S_i^+;S_0^-\rangle\rangle$ is the time-Fourier transform of the advanced or retarded double-time Green function depending whether the imaginary part of the complex frequency is positive or negative.[9] In the paramagnetic phase LRO is absent—that is, $\langle S_0^z\rangle=0$—and Eq. (2) becomes

$$\omega\langle\langle S_i^+;S_0^-\rangle\rangle = -2\sum_{\boldsymbol{\delta}} J_{\boldsymbol{\delta}}(\langle\langle S_i^z S_{i+\boldsymbol{\delta}}^+;S_0^-\rangle\rangle - \langle\langle S_{i+\boldsymbol{\delta}}^z S_i^+;S_0^-\rangle\rangle). \tag{3}$$

The equations of motion for the three-operator Green functions appearing on the right-hand side of Eq. (3) lead to a four-operator Green function equation of motion. A high-temperature decoupling (HTD) approximation is used:

$$\langle\langle S_l^z S_j^z S_i^+;S_0^-\rangle\rangle = g(|\boldsymbol{r}_l - \boldsymbol{r}_j|)\langle\langle S_i^+;S_0^-\rangle\rangle \tag{4}$$

and

$$\langle\langle S_l^+ S_j^- S_i^+;S_0^-\rangle\rangle = 2g(|\boldsymbol{r}_l - \boldsymbol{r}_j|)\langle\langle S_i^+;S_0^-\rangle\rangle + 2g(|\boldsymbol{r}_i - \boldsymbol{r}_j|)\langle\langle S_l^+;S_0^-\rangle\rangle, \tag{5}$$

where $g(|\boldsymbol{r}_i-\boldsymbol{r}_j|)=\langle S_i^z S_j^z\rangle=\frac{1}{2}\langle S_i^- S_j^+\rangle=\frac{1}{2}\langle S_i^+ S_j^-\rangle$ is the isotropic correlation function between spins located at site i and j, and $g(0)=S(S+1)/3$. With these approximations the Green function equation of motion becomes

$$\begin{aligned}\omega^2\langle\langle S_i^+;S_0^-\rangle\rangle &= \frac{1}{\pi}\sum_{\boldsymbol{\delta}} 4J_{\boldsymbol{\delta}}g(|\boldsymbol{\delta}|)(\delta_{i,0}-\delta_{i+\boldsymbol{\delta},0})\\ &\quad + 8\sum_{\boldsymbol{\delta}}\sum_{\boldsymbol{\delta}'} J_{\boldsymbol{\delta}}J_{\boldsymbol{\delta}'}[g(|\boldsymbol{\delta}+\boldsymbol{\delta}'|)(\langle\langle S_i^+;S_0^-\rangle\rangle\\ &\quad - \langle\langle S_{i+\boldsymbol{\delta}}^+;S_0^-\rangle\rangle) - g(|\boldsymbol{\delta}|)(\langle\langle S_{i+\boldsymbol{\delta}'}^+;S_0^-\rangle\rangle\\ &\quad - \langle\langle S_{i+\boldsymbol{\delta}+\boldsymbol{\delta}'}^+;S_0^-\rangle\rangle)].\end{aligned} \tag{6}$$

Defining the momentum-Fourier transform of the Green function,

$$\langle\langle S_i^+;S_0^-\rangle\rangle = \frac{1}{N}\sum_{\boldsymbol{k}} G_{\boldsymbol{k}}(\omega)e^{i\boldsymbol{k}\cdot\boldsymbol{r}_i}, \tag{7}$$

the solution of the equation of motion for $G_{\boldsymbol{k}}(\omega)$ is

$$G_{\boldsymbol{k}}(\omega) = \frac{1}{\pi}\frac{\sum_{\boldsymbol{\delta}} 4J_{\boldsymbol{\delta}}g(|\boldsymbol{\delta}|)(1-e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}})}{\omega^2-\omega_{\boldsymbol{k}}^2}, \tag{8}$$

where

$$\omega_{\boldsymbol{k}}^2 = 8\sum_{\boldsymbol{\delta}}\sum_{\boldsymbol{\delta}'} J_{\boldsymbol{\delta}}J_{\boldsymbol{\delta}'}(1-e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}})[g(|\boldsymbol{\delta}+\boldsymbol{\delta}'|)-g(|\boldsymbol{\delta}|)e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}'}]. \tag{9}$$

Using the well-known relationship between the correlation function and the corresponding Green function,[9]

$$\begin{aligned}g(|\boldsymbol{\delta}|) &= \frac{1}{2N}\sum_{\boldsymbol{k}} e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}}\lim_{\eta\to 0} i\int_{-\infty}^{+\infty} d\omega[G_{\boldsymbol{k}}(\omega+i\eta)\\ &\quad - G_{\boldsymbol{k}}(\omega-i\eta)]\frac{1}{e^{\omega/k_BT}-1},\end{aligned} \tag{10}$$

Eqs. (9) and (10) give the high-temperature behavior of the magnetic excitations of a Heisenberg model on any lattice. For CsMnBr$_3$ the spin is $S=5/2$ and the nearest-neighbor (NN) intrachain exchange interaction is $2J_{\parallel}=-1.78$ meV. The NN interchain interaction is assumed to be $J_{\perp}\simeq 0.002J_{\parallel}$ and the weak anisotropy is neglected.[6] Defining $\omega_{\boldsymbol{k}}=2|J_{\parallel}|\epsilon_{\boldsymbol{k}}$ one has

$$\begin{aligned}\epsilon_{\boldsymbol{k}}^2 &= (1-\cos ck_z)[g(0)+g(2c)-2g(c)\cos ck_z]\\ &\quad + 6(J_{\perp}/J_{\parallel})\{(1-\cos ck_z)[g(\sqrt{a^2+c^2})-g(c)\gamma_{\boldsymbol{k}}]+(1-\gamma_{\boldsymbol{k}})\\ &\quad \times[g(\sqrt{a^2+c^2})-g(a)\cos ck_z]\}+3(J_{\perp}/J_{\parallel})^2(1-\gamma_{\boldsymbol{k}})\\ &\quad \times[g(0)+g(2a)+2g(a)+2g(\sqrt{3}a)-6g(a)\gamma_{\boldsymbol{k}}],\end{aligned} \tag{11}$$

with $a=7.61$ Å and $c=3.26$ Å which are the distance between NN spins in and out of plane, respectively, and

$$\gamma_{\boldsymbol{k}} = \frac{1}{3}\left[\cos ak_x + 2\cos\left(\frac{1}{2}ak_x\right)\cos\left(\frac{\sqrt{3}}{2}ak_y\right)\right]. \tag{12}$$

The self-consistent equations for the correlation functions are

$$\begin{aligned}g(c) &= -\frac{1}{N}\sum_{\boldsymbol{k}}\cos ck_z\frac{g(c)(1-\cos ck_z)+3g(a)\frac{J_{\perp}}{J_{\parallel}}(1-\gamma_{\boldsymbol{k}})}{\epsilon_{\boldsymbol{k}}}\\ &\quad \times\coth\frac{\epsilon_{\boldsymbol{k}}}{\theta},\end{aligned} \tag{13}$$

$$\begin{aligned}g(2c) &= -\frac{1}{N}\sum_{\boldsymbol{k}}\cos 2ck_z\frac{g(c)(1-\cos ck_z)+3g(a)\frac{J_{\perp}}{J_{\parallel}}(1-\gamma_{\boldsymbol{k}})}{\epsilon_{\boldsymbol{k}}}\\ &\quad \times\coth\frac{\epsilon_{\boldsymbol{k}}}{\theta},\end{aligned} \tag{14}$$

$$g(a) = -\frac{1}{N}\sum_{\boldsymbol{k}}\gamma_{\boldsymbol{k}}\frac{g(c)(1-\cos ck_z)+3g(a)\frac{J_{\perp}}{J_{\parallel}}(1-\gamma_{\boldsymbol{k}})}{\epsilon_{\boldsymbol{k}}}\coth\frac{\epsilon_{\boldsymbol{k}}}{\theta}, \tag{15}$$

$$\begin{aligned}g(\sqrt{3}a) &= -\frac{1}{N}\sum_{\boldsymbol{k}}\frac{1}{3}\left(\cos\sqrt{3}ak_y + 2\cos\frac{3}{2}ak_x\cos\frac{\sqrt{3}}{2}ak_y\right)\\ &\quad \times\frac{g(c)(1-\cos ck_z)+3g(a)\frac{J_{\perp}}{J_{\parallel}}(1-\gamma_{\boldsymbol{k}})}{\epsilon_{\boldsymbol{k}}}\coth\frac{\epsilon_{\boldsymbol{k}}}{\theta},\end{aligned} \tag{16}$$

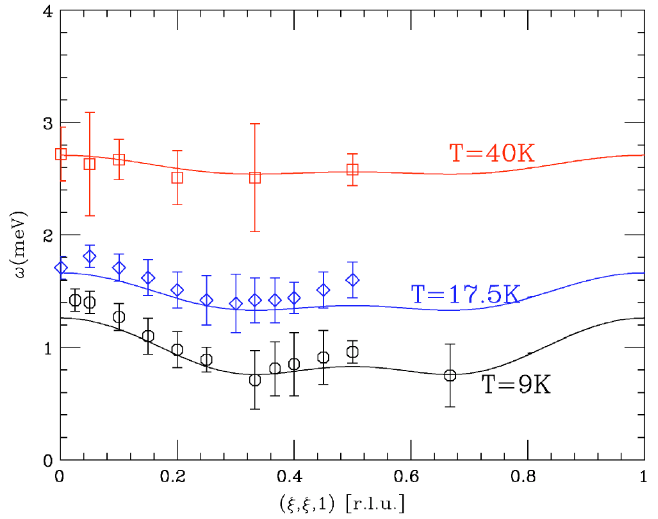

FIG. 1. Dispersion relation for CsMnBr$_3$ along the [1,1,0] direction for temperatures $T=9$, 17.5, and 40 K. Data points are taken from the neutron scattering experiment of Ref. 6: $T=9$ K (circles), 17.5 K (diamonds), and 40 K (squares).

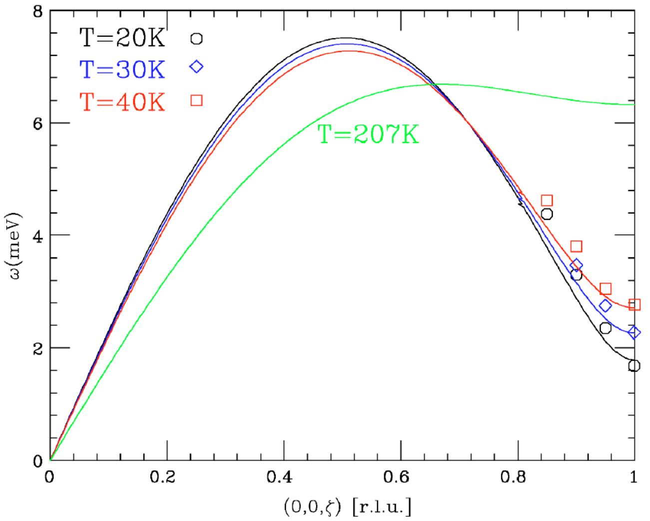

FIG. 2. Dispersion relation for CsMnBr$_3$ along [0,0,1] direction for temperatures $T=20$, 30, and 40 K. Data points are taken from the neutron scattering experiment of Ref. 6: $T=20$ K (circles), 30 K (diamonds), and 40 K (squares).

$$g(2a) = -\frac{1}{N}\sum_{\boldsymbol{k}} \frac{1}{3}(\cos 2ak_x + 2\cos ak_x \cos\sqrt{3}ak_y) \times \frac{g(c)(1-\cos ck_z) + 3g(a)\frac{J_\perp}{J_\parallel}(1-\gamma_{\boldsymbol{k}})}{\epsilon_{\boldsymbol{k}}}\coth\frac{\epsilon_{\boldsymbol{k}}}{\theta}, \tag{17}$$

$$g(\sqrt{a^2+c^2}) = -\frac{1}{N}\sum_{\boldsymbol{k}} \gamma_{\boldsymbol{k}} \cos ck_z \times \frac{g(c)(1-\cos ck_z) + 3g(a)\frac{J_\perp}{J_\parallel}(1-\gamma_{\boldsymbol{k}})}{\epsilon_{\boldsymbol{k}}}\coth\frac{\epsilon_{\boldsymbol{k}}}{\theta}, \tag{18}$$

where $\theta = k_B T/2|J_\parallel|$.

It has to be noted that the spectrum $\epsilon_{\boldsymbol{k}}$ has a minimum at $\boldsymbol{k} = (4\pi/3a, 0, \pi/c)$ corresponding to $(\frac{1}{3}, \frac{1}{3}, 1)$ in reciprocal lattice units [r.l.u.]. This minimum goes to zero at $\theta_m = 0.066\,26$. This means that the present theory cannot be pushed below $T_m = 1.4$ K. On the other hand, the HTD approach is expected to hold for $T > T_N = 8.3$ K. At $T=9$, 17.5, and 40 K we find $\omega(0,0,1) = 1.261$, 1.663, and 2.710 meV and $\omega(\frac{1}{3}, \frac{1}{3}, 1) = 0.757$, 1.331, and 2.541 meV.

In Fig. 1 we show the spectrum given by Eq. (11) compared with the experiment on CsMnBr$_3$ (Ref. 6). The data points for ω in Fig. 1 are obtained from Fig. 3 of Ref. 6 where $\Omega = \sqrt{\omega^2 + \Gamma^2}$ is shown. The energy width Γ is $\simeq 0.6$, 0.8, and 1.43 meV for $T=9$, 17.5, and 40 K, respectively.[10]

In Fig. 2 we show the spectrum given by Eq. (11) along the [0,0,1] direction for $T=20$, 30, and 40 K. The data points are obtained from Fig. 2 of Ref. 6 in the same way as in Fig. 1.[10] The theoretical expectation for a very high temperature ($T=207$ K) is also quoted. Note that the excitation energies corresponding to modes with $\zeta \lesssim 2/3$ renormalize downward whereas those corresponding to $\zeta \gtrsim 2/3$ renormalize upward. This result agrees with the renormalization observed[10] over the whole Brillouin zone at $T=9$ and 17.5 K. Collins and Gaulin[11] reported inelastic neutron scattering measurements in the paramagnetic phase of CsMnBr$_3$ between $T=15$ and 45 K along the [0,0,1] direction. They did not find renormalization for the mode at $Q=(0,0,1.3)$ corresponding to the mode (0,0,0.7) of Fig. 2 in agreement with our expectation even though the experiment gives an energy of about 7 meV to be compared with 6 meV of our calculation. On the contrary upward renormalization was found for the mode at $Q=(0,0,1.1)$ corresponding to the mode (0,0,0.9) of Fig. 2. In this case the agreement is complete since the energy obtained from the experiment is about 3 meV. Weak upward renormalization is also reported for the excitation at $Q=(0,0,1.15)$ (with an energy of about 4.2 meV) and at $Q=(0,0,1.2)$ (with an energy of about 5.2 meV) corresponding to the modes (0,0,0.85) and (0,0,0.8) of Fig. 2. The authors were not able to explain the high value of the gap observed at $Q=(0,0,1)$ for $T=15$ K (1.7 ± 0.2 meV) using the spin-wave spectrum even accounting for dipolar interactions. This is not surprising in view of our calculation where an upward renormalization is obtained for the isotropic Heisenberg model neglecting any anisotropy source, in particular the dipolar interaction, which is, however, responsible for the gap in the ordered phase. A similar upward renormalization of the spin-wave energy at wave vectors around $Q=(0,0,1)$ was also observed in TMMC,[12] a quasi-one-dimensional Heisenberg antiferromagnet with $S=5/2$. For both compounds the upward renormalization of the (0,0,1) mode cannot be traced back to the Haldane scenario because of their half-integer spin.

In Fig. 3 the energy of modes at (0,0,1) and $(\frac{1}{3}, \frac{1}{3}, 1)$ is shown as a function of temperature. For comparison we have reported the experimental data taken from Ref. 6. As one can see the agreement is very good.

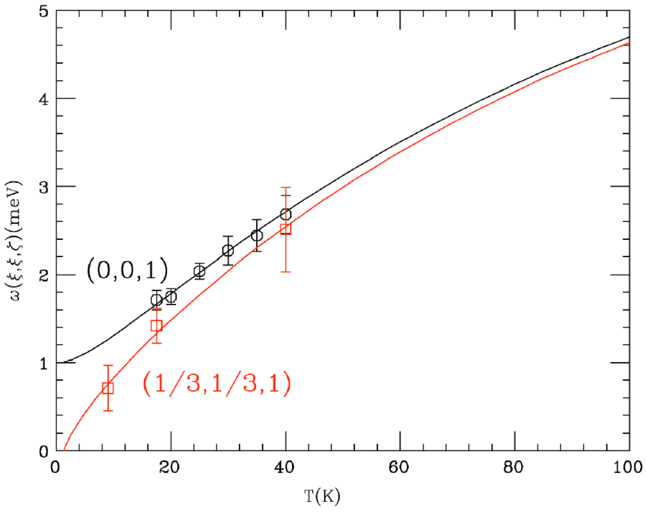

FIG. 3. Frequency of the modes (0,0,1) and $(\frac{1}{3},\frac{1}{3},\ 1)$ versus temperature. Data points for $\mathrm{CsMnBr_3}$ (circles and squares) are taken from Ref. 6.

In the very-high-temperature limit we find that the correlation functions are given by $g(c)\simeq-[S(S+1)/3]^2/\theta$, $g(2c)\simeq[S(S+1)/3]^3/\theta^2$, $g(a)\simeq(J_\perp/J_\parallel)g(c)$, $g(\sqrt{a^2+c^2})\simeq 2(J_\perp/J_\parallel)g(2c)$, $g(\sqrt{3}a)\simeq(2/3)(J_\perp/J_\parallel)^2 g(2c)$, and $g(2a)\simeq(J_\perp/J_\parallel)^2 g(2c)$. This high-temperature behavior agrees within 10%–20% with the numerical solution of the self-consistent equations (13)–(18) for $\theta\gtrsim 10$ ($T\gtrsim 200$ K) where there are no experimental data. In the limit of infinite temperature the spectrum reduces to

$$\omega_{\mathbf{k}}=4|J_\parallel|[S(S+1)/3]^{1/2}\sqrt{1-\cos ck_z+3(J_\perp/J_\parallel)^2(1-\gamma_{\mathbf{k}})}. \tag{19}$$

In one dimension we recover the first term of the exact high-temperature expansion of the correlation function of the Heisenberg quantum model.[13]

In summary we have shown that the upward renormalization at increasing temperature observed in the paramagnetic phase of $\mathrm{CsMnBr_3}$ can be explained by a HTD Green function approach. The presence of well-defined elementary excitations in the paramagnetic phase of the $\mathrm{CsMnBr_3}$ is obviously relied to the strong intrachain exchange interaction that supports short-range order at temperatures well beyond the critical temperature. However, the existence of well-defined excitations for wave vectors perpendicular to the chain supported by the weak interchain coupling suggests that one be cautious in describing the ABX_3 compounds in the paramagnetic phase $T_N\lesssim T\lesssim 3T_N$ as genuine one-dimensional (1D) spin models. In particular the magnetic susceptibility of $\mathrm{CsMnBr_3}$ measured at a field of 3.5 kOe (Ref. 14) shows a temperature dependence in agreement with the exact solution of a genuine 1D classical ($S\rightarrow\infty$) Heisenberg antiferromagnet[15] only for temperatures higher than ~50 K. In other words it seems well established that 3D character is maintained for temperatures well beyond the critical temperature. For these reasons it is not surprising that a Green function approach, where the absence of LRO has been taken into account, is successful in giving the thermal renormalization of a single-particle-like excitation energy in agreement with experimental data.

Moreover, we observe that inelastic neutron scattering measurements on $\mathrm{CsVCl_3}$, a ABX_3 compound with $S=3/2$, in the paramagnetic phase at $T=40\ \mathrm{K}\simeq 3T_N$ along the [0,0,1] direction[16] confirm our expectation of a downward renormalization at intermediate wave vectors with respect to the excitation spectrum in the ordered phase.[17] It should be interesting to investigate the excitations close to the antiferromagnetic mode (0,0,1) where we expect an upward renormalization on the basis of our approach.

Finally, we stress that the upward renormalization of the antiferromagnetic mode is a common feature of $\mathrm{CsMnBr_3}$, TMMC ($S=5/2$), and $\mathrm{CsNiCl_3}(S=1)$. Our approach gives upward renormalization of the antiferromagnetic mode and downward renormalization of the long-wavelength modes independent of the spin value. We expect that spin-dependent quantum effects which are dramatic in genuine 1D Heisenberg models at zero temperature could be less important in actual compounds in the paramagnetic phase where the 3D character survives well beyond the critical temperature. We recall that a quantum field theory in the continuum limit ($k\rightarrow 0$) applied to a 1D Heisenberg antiferromagnet with $S=1$ at zero temperature[18] gives a neutron cross section characterized by a rounded peak at energy $\omega_m=\sqrt{6\Delta^2+v^2k^2}$ somewhat higher than a threshold $\omega_{th}=\sqrt{4\Delta^2+v^2k^2}$ which is about twice the gap at the antiferromagnetic wave vector $\omega_\pi=\sqrt{\Delta^2+v^2(k-\pi)^2}$ (Haldane gap). This threshold excitation corresponds to the lower bound of a two-magnon band. The existence of an excitation of energy ω_{th} was confirmed by numerical simulations on a finite chain of 32 spins with $S=1$ (Ref. 19). Indeed an excitation of order 2Δ was found at $k=\pi/16$, the minimum wave vector for a chain of 32 spins. No indication of a two-magnon band was pointed out in such a calculation. As for experiments on $\mathrm{CsNiCl_3}$ (Ref. 5), no energy band seems to have been observed. For these reasons inelastic neutron scattering on $\mathrm{CsNiCl_3}$ for small wave vectors in the paramagnetic phase should be crucial to check the importance of the quantum effects of a genuine 1D system with integer spin. Notice that in $\mathrm{CsNiCl_3}$ the spin-wave frequencies for $T>T_N$ show a clear dependence on the wave vector component perpendicular to the chain (c axis) as one can see from Fig. 3 of Ref. 20. This is also confirmed by a comparison of the upward renormalization of the mode at $(\frac{1}{3},\frac{1}{3},\ 1)$ shown in Fig. 2 of Ref. 21 and in Fig. 8 of Ref. 22 compared to the upward renormalization of the mode at (0.81, 0.81, 1) as shown in Fig. 3 of Ref. 5. We are confident that the HTD Green function approach could be reliable also for integer-spin compounds because of the 3D character of the ABX_3 compounds well beyond their critical temperature, which is in the region where the experiments were performed.

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