Incommensurate nodes in the energy spectrum of weakly coupled antiferromagnetic Heisenberg ladders

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Heisenberg ladders are investigated using the bond-mean-field theory [M.Azzouz, Phys.Rev.B 48, 6136 (1993)]. The zero inter-ladder coupling energy gap, the uniform spin susceptibility and the nuclear magnetic resonance spin-relaxation rate are calculated as a function of temperature and magnetic field. For weakly coupled ladders, the energy spectrum vanishes at incommensurate wavevectors giving rise to nodes. As a consequence, the spin susceptibility becomes linear at low temperature. Our results for the single ladder successfully compare to experiments on SrCu$_2$O$_3$ and (VO)$_2$P$_2$O$_7$ materials and new predictions concerning the coupling to the magnetic field are made.

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Among materials that contributed to the increased interest in low-dimensional spin systems, one finds ladder compounds like vanadyl-pyrophosphate (VO)$_2$P$_2$O$_7$ or Cu-O layers of SrCu$_2$O$_3$. These can be modeled by the Heisenberg Hamiltonian on a ladder geometry. Here, the general Heisenberg model

$$H = J_\perp \sum_{i,j} S_{i,2j} \cdot S_{i,2j+1} + J'_\perp \sum_{i,j} S_{i,2j+1} \cdot S_{i,2j+2} + J \sum_{i,j} S_{i,j} \cdot S_{i+1,j} - h \sum_{i,j} S_i^z$$

for an array of coupled ladders in a uniform magnetic field $h$ along the z-direction is considered. The quantities $J$, $J'_\perp$ and $J'_\parallel$ stand respectively for intrachain, transverse intra-ladder and inter-ladder positive exchange coupling constants. It is now well established that any finite transverse coupling opens an energy gap, $E_g$, in elementary spin excitations for the single ladder ($J'_\perp = 0$). As a consequence of this energy gap, thermodynamical quantities like the uniform susceptibility, $\chi'$, and the nuclear magnetic resonance (NMR) spin-relaxation rate, $T_1^{-1}$, show an activated behavior at low temperatures, $T < E_g$. Recently an important numerical work has been reported by Sandvik et al. who used Quantum Monte Carlo (QMC) technique to calculate the magnetic susceptibility and the NMR relaxation rate as a function of temperature for the SrCu$_2$O$_3$ material. However, comparisons to experiments did not lead to a clear conclusion concerning the magnitude of the longitudinal, $J$, and transverse, $J'_\perp$, exchange couplings. The best fits to Azuma et al. results they reported for $\chi'$ and $T_1^{-1}$ were obtained using different values, namely $J = J_\perp = 850$ K for $\chi'$ and $J = J'_\perp = 1200$ K for $1/T_1$. It is therefore interesting to address the question of the relevance of the Heisenberg ladder for this system. Frischmuth et al. numerically studied (also using QMC method) the temperature dependence of the uniform susceptibility for Heisenberg ladders with up to 6 legs. They came to the conclusion that the gap rapidly decreases when the number of legs increases (for even number of legs).

The Hamiltonian (1) is studied by the bond-mean-field theory (BMFT) explained in detail in Refs. [1,2]. In this approach, (1) becomes a spinless fermion Hamiltonian

$$H = \frac{J}{2} \sum_{i,j} c_{i+1,j}^\dagger c_{i,j} e^{-i\phi} + \frac{J}{2} \sum_{i,j} c_{i,2j+1}^\dagger c_{i,2j} e^{-i\phi_\perp} + \frac{J'_\parallel}{2} \sum_{i,j} c_{i,2j+2}^\dagger c_{i,2j+1} e^{-i\phi'_\parallel} + J \sum_{i,j} (n_{i,j} - \frac{1}{2})(n_{i+1,j} - \frac{1}{2}) + J'_\perp \sum_{i,j} (n_{i,2j+1} - \frac{1}{2})(n_{i,2j+2} - \frac{1}{2}) - h \sum_{i,j} c_{i,j}^\dagger c_{i,j}$$

using the two-dimensional (2D) generalization of Jordan-Wigner transformation where the phases $\Phi$, $\Phi_\perp$ and $\Phi'_\parallel$ are readily derived. The BMFT treatment consists, first and foremost, to approximate the sum of these phases on each plaquette by $\pi$. Then, quartic terms corresponding to Ising interactions are decoupled by introducing the bond parameters $Q = \langle c_{i,j} c_{i+1,j}^\dagger \rangle$, $P = \langle c_{i,2j} c_{i,2j+1}^\dagger \rangle$ and $P' = \langle c_{i,2j+1} c_{i,2j+2}^\dagger \rangle$ that are calculated by minimising the mean-field free energy. The dispersion relation is found to be

$$E_{\pm}(k) = \pm \frac{1}{2} \left( 4J_1^2 \sin^2 k_x + 2J_{\perp 1} J_{\perp 2} \cos^2 k_y + J_1^2 + J_{\perp 1}^2 + 4J_1 (J_{\perp 1} - J_{\perp 2}) \sin k_x \sin k_y \right)^{1/2} - h$$
with \( J_1 = J(1+2Q) \), \( J_{11} = J_1(1+2P) \) and \( J_{12} = J_1'(1+2P') \).

In this work, elementary spin excitations are described in a fermionic like picture. Consequently, the number of fermions in excitations that contribute to thermodynamical functions is found to be conserved just as in normal Fermi systems. This result indicates that only the \( S^z = 0 \) component of the triplet excited state is relevant to thermodynamics, and is similar to what has been already verified both experimentally and theoretically in the case of spin-Peierls materials.\[1\]\[2\]\[3\]\[4\]\[5\]\[6\]\[7\]\[8\]\[9\]\[10\]\[11\]\[12\]\[13\]\[14\]\[15\]\[16\]\[17\]\[18\]\[19\]\[20\]\[21\]\[22\]\[23\]\[24\]\[25\]

For example, this property is reflected by the fact that the magnetic Zeeman term is missing in the magnetic field dependence of the zero inter-ladder coupling energy gap \( E_g(T,h) = |J_{11} - J_{12}|/2 \). The simple physical reason behind this effect is that thermal fluctuations cannot create spin configurations with \( S^z = \pm 1 \) because up and down spins are equally distributed. This also remains true at zero temperature because \( S^z = 0 \) due to quantum fluctuations. The energy gap, \( E_g(T,h) \), of the single ladder (\( J_{11}' = 0 \)) reported in Fig. 1 as a function of temperature never vanishes, but instead decreases with increasing temperature. In other words, the effect of the gap is washed out by thermal fluctuations when the temperature becomes larger than a characteristic value of the order of the zero temperature energy gap. This is in perfect agreement with the absence of a phase transition in the two real ladder materials we mentioned above.\[3\]\[4\] Also, from Fig. 1, it is seen that \( E_g(T,h) \) remains unaffected by the magnetic field at low temperature. As we explained before, this result is reminiscent of the fact that only the \( S^z = 0 \) component of the triplet excited state will contribute to thermodynamical quantities, and is a consequence of the absence of the magnetic Zeeman term in \( E_g(T,h) \). The magnetic field dependence is implicitly included in the parameters \( Q, P \) and \( P' \) and vanishes for \( T \to 0 \) as can be noticed in Fig. 1. We should mention here that in the single ladder limit (\( J_{11}' = 0 \)), the gap at \( k_z = 0 \) is the same as that at \( k_z = \pi \). This contrasts with exact diagonalization results\[2\] where the gap at \( k_z = 0 \) is twice the gap at \( k = \pi \). Our mean-field gap finds however a compromise by being half way between these gaps.

As for inter-ladder coupling, from Eq. 3, one can distinguish three regimes: i) \( J_{11}' = 0 \) where the only transverse nodes are \( k_y = 0, \pi \). The third term under the square root in the dispersion relation, that becomes identical to that of the ladder system, vanishes. ii) \( J_{11}' = J_{12} \), if \( J_{11} \sim J \) then the AF order parameter has to be included.\[2\] In the controversial limit \( J_{11} \ll J \), the ground state is either ordered antiferromagnetically or is a gapless spin liquid state. The later is well described by Eq. 2 where \((J_{11} - J_{12}) \sin k_x \sin k_y = 0 \) since \( J_{11} = J_{12} \). iii) \( J_{11}' \ll J_{12} \), this is an interesting regime. The dispersion relation is found to present nodes at incommensurate wavevectors. The third term under the square root, which is absent in cases i and ii, is responsible for such a behavior. For \( J_{11}' = 0.01 \) for example, the zeros of the spectrum occur at \((k_x, k_y) = (3.661212, \pm \pi/2), (k_x, k_y) = (5.763566, \pm \pi/2)\) and their images with respect to the symmetry centre \((\pi, \pi)\) as shown in figure 2.

Now, we calculate the momentum transfer- and frequency-dependent response function, \( \chi(q, \omega) \), by evaluating the space and time Fourier transform of the correlation function \( \langle T(S_{i,j}^z(\tau)S_{i,j}^z(\tau')) \rangle \). We obtain:

\[
\chi(q, \omega) = \frac{1}{8} \sum k \left\{ \frac{f_+(k+q) - f_+(k)}{E_+(k+q) - E_+(k) - \omega} + \left( + \rightarrow - \right) \theta_+ \right\} \left\{ \frac{f_-(k+q) - f_-(k)}{E_-(k+q) - E_+(k) - \omega} + \left( + \rightarrow - \right) \theta_- \right\}
\]

where \( f_\pm \) is the Fermi factor and \( \theta_\pm = 1 \pm \cos(\alpha_k - \alpha_{k+q}) \) with \( \alpha_k \), satisfying tan \( \alpha_k = \frac{2J_1 \sin k_x + (J_{11}' - J_{12}) \sin k_y}{(J_{11} + J_{12}) \cos k_y} \), is the phase of the coherence factors required in the phase diagonalization of the mean-field Hamiltonian. The static and uniform spin susceptibility, \( \chi(T, h) \), is obtained by evaluating \( \lim_{q \to 0, \omega \to 0} \chi(q, \omega) \). This yields the following expression: \( \chi(T, h) = \frac{\beta}{4} \sum k, p = \pm 1 + e^{\beta E_0(k)} - 2 e^{-\beta E_0(k)} \) where \( \chi''(q, \omega) \) is the imaginary part of \( \chi(q, \omega) \) and \( A(q) \) is the nuclear hyperfine form factor that will be determined by fitting experimental data. \( h, g \) and \( \gamma_N \) are respectively, the Bohr magneton, the Landé and gyromagnetic factors. In Fig. 3, we display \( \chi(T, h) \) as a function of temperature for several values of the magnetic field for the single ladder limit. From this figure, one can notice that the magnetic field manifests itself only at low temperature contrary to the energy gap that is rather affected at high temperature. The dominant contribution to \( \chi(T) \) when \( T \ll E_g \) that is given by \( \cosh(3h) e^{-\beta E_g} \) (\( E_g \) is field independent for \( T \sim 0 \)) explains such a behavior. In order to confirm these predictions in experiment, we suggest to carry on a measurement of the spin susceptibility as a function of temperature and magnetic field on a ladder material. Note finally that a sizable effect of the magnetic field begins at \( h \sim 0.1 E_g(T = 0) \). For weakly coupled ladders, as shown in Fig. 4 for \( J_{11}' = 0.01 \) and \( J_{11} = J = 1 \), the energy spectrum is found to show nodes (zero energies) at incommensurate wavevectors, \((k_x, k_y) = (3.661212, \pm \pi/2), (k_x, k_y) = (5.763566, \pm \pi/2)\) and their images. As a result of these nodes, \( \chi(T) \sim T \) at low temperature. Note that in a work reported by Gopalan et al.\[12\] the gap vanishes only for \( J_{11}'/J_1 \geq 0.25 \) at commensurate wavevectors. However, as their mean-field theory favors the opening of an energy gap, the question of the right threshold value (whether it is 0 or 0.25) will remain open. An argument in favor of our result is given by the fact that nodes occur at incommensurate wavevectors as a consequence of coupling anisotropy.

Let us compare with one experiment. First we would like to address the temperature dependence of the energy gap. In Fig. 5, one notes that the gap decreases with increasing temperature. However, contrary to previous investigations,\[3\]\[4\]\[5\]\[6\]\[7\]\[8\]\[9\]\[10\]\[11\]\[12\]\[13\]\[14\]\[15\]\[16\]\[17\]\[18\]\[19\]\[20\]\[21\]\[22\]\[23\]\[24\]\[25\]
where the zero temperature value of the gap is used for all temperatures, one should take into account the decrease of the gap. The data of Johnston et al. concerning the uniform magnetic susceptibility for the \( \text{VO}_2\text{P}_2\text{O}_7\) compound are fitted in Fig. 2 using the single ladder limit. These data have already been fitted by Barnes and Riera using exact diagonalization with \( J = 7.82\) meV and \( J_1 = 7.76\) meV. We have noted that our calculations for the single ladder are more accurate in this case than for \( \text{SrCu}_2\text{O}_3\) (we do not report the fit for \( \text{SrCu}_2\text{O}_3\) because of lack of space). Concerning the NMR relaxation rate, we plot in Fig. 3 both Azuma et al.’s experimental results of the \( \text{SrCu}_2\text{O}_3\) compound and the best fit obtained using the same coupling constants as for the susceptibility. This suggests, as an answer to the problem rose in Dagotto et al.’s work, that both \( \chi’ \) and \( 1/T_1 \) can be modeled using the same set of parameters of the Heisenberg single ladder. For the magnetic field, a sizable effect in this material is expected for fields \( h \sim 0.1E_g(T = 0) \sim 100\) T that are not accessible in experiment. One can instead use the \( \text{VO}_2\text{P}_2\text{O}_7\) material since the energy gap \( E_g \approx 50\) K is much smaller. This yields \( h \sim 10\) T.

The departure from experimental data at high temperature may be attributed to the fact that our spectrum is the same for either \( k_\perp = 0 \) or \( k_\perp = \pi \), and gaps at \( k = 0 \) and \( k = \pi \) are equal. As stressed before, the gap at \( k = 0 \) is twice that at \( k = \pi \). Another point is that since energy gaps decrease with increasing temperature, contributions from the continuum of the spectrum above the gap at \( k = 0 \) could become not negligible already for temperatures \( T \sim E_g(T = 0) \). This is obvious for \( \text{VO}_2\text{P}_2\text{O}_7\) where deviations start at around \( T \sim 50\) K. However, it is less clear for \( \text{SrCu}_2\text{O}_3\) where deviations are found to occur at \( T \sim 250\) K which is almost half the gap (\( E_g \approx 468\) K). From these above remarkes, we conclude that our theory can be applied to explain experiments at low temperature \((T \leq E_g(T = 0))\).

In conclusion, we have used the bond-mean-field theory to investigate AF Heisenberg ladder spin systems. The uniform spin susceptibility and the NMR spin-relaxation rate are calculated as a function of temperature and uniform magnetic field. For a single ladder, the energy gap remains unaffected by the magnetic field at low temperature in agreement with the fact that the relevant elementary spin excitations to thermodynamics (as seen here for \( \chi’ \) and \( T_{1}^{-1} \)) conserve the total \( S^2 = 0 \) component. Our results successfully compare to experiments on \( \text{SrCu}_2\text{O}_3\) and \( \text{VO}_2\text{P}_2\text{O}_7\) compounds for \( T \leq E_g\). Several predictions concerning the magnetic field effect that can be verified in simple experiments are also made. Inter-ladder coupling is found to drastically change the behavior of the uniform susceptibility, \( \chi’ \), at very low temperature. For \( J_1^{-1} = 0.01J_\perp \) for example, \( \chi’ \) becomes linear in \( T \) as a consequence of the appearance of nodes in the spectrum at incommensurate wavevectors, \((k_x, k_y) = (3.661212, \pm \pi/2), (k_x, k_y) = (5.763566, \pm \pi/2)\) and their images. It is worthy of note to mention, in the end of this work, that Gopalan et al. suggested that the inter-ladder coupling in \( \text{SrCu}_2\text{O}_3\) is ferromagnetic. They based their argument on the fact that two \( \text{Cu} \) ions on adjacent ladders are connected through an \( \text{O} \) site by \( 90^\circ \) bonds. A calculation in this case of inter-ladder coupling will be reported in the near future.

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FIG. 1. (a) The susceptibility is drawn for $J_{\perp}' = 0$ as a function of temperature for several values of $h$. In the inset, $E_g$ is drawn as a function of temperature for $h = 0$.

FIG. 2. The dispersion relation for weakly coupled ladders is plotted as a function of $k_x$ and $k_y$; $J_{\perp}' = 0.01J_{\perp}$.

FIG. 3. In (a), the fit to Johnston et al. susceptibility data is reported for $(VO)_2P_2O_7$. The fit to Dagotto et al.’s experimental results for $1/T_1$ is shown in (b) in the case of SrCuO$_3$. Data are drawn in crosses.