Dynamics of Coupled Quantum Spin Chains

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Static and dynamical properties of weakly coupled antiferromagnetic spin chains are treated using a mean-field approximation for the interchain coupling and exact results for the resulting effective one-dimensional problem. Results for staggered magnetization, Néel temperature, and spin wave excitations are in agreement with experiments on $KCuF_3$. The existence of a narrow longitudinal mode is predicted. The results are in agreement with general scaling arguments, contrary to spin wave theory. [S0031-9007(96)01025-3]

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One-dimensional quantum spin chains are interesting objects to study for a number of reasons. On the one hand, experimental systems are generally very well described by simple yet nontrivial Hamiltonians involving very few unknown parameters. The standard example is the Heisenberg model, with only one free parameter, the exchange constant. Comparison between theory and experiment then becomes a particularly stringent test, as a variety of data have to be explained by one single parameter. Moreover, both a large number of exact theoretical results and powerful analytical and numerical methods are available, making this comparison particularly interesting. On the other hand, in spite of their simplicity, models of quantum spin chains have lead to a number of unexpected and unconventional predictions. For example, the exact solution of the antiferromagnetic spin-$1/2$ Heisenberg chain shows that the low-lying excitations are spin-$1/2$ objects [1] (now called spinons), quite different from standard spin waves. This prediction has been confirmed experimentally quite recently in $KCuF_3$ [2]. Another example is Haldane's prediction of a gap in the excitation spectrum for integer-S antiferromagnets [3], which again has found experimental confirmation [4,5].

Strictly one-dimensional models of course do not exhibit phase transitions into states with a broken symmetry. It is nevertheless clear that in any real compound, like $KCuF_3$ [2,6,7], Sr_2CuO_3 [8,9], or Yb_4As_3 [10] some form of interchain coupling is present. Then three-dimensional magnetic long-range order can appear below a Néel temperature T_N. In the present paper I show that in this case a conceptually simple approach, namely treating the interchain coupling in the mean-field approximation and treating the resulting effective one-dimensional problem as exactly as possible [11,12], gives a coherent description of the ordered state and produces nontrivial quantitative predictions for static and dynamic quantities that can be successfully compared to experiments, in particular on $KCuF_3$ where detailed neutron scattering results are available [2,6].

I start with the natural model for coupled spin-$1/2$ antiferromagnetic chains, namely, a spatially anisotropic Heisenberg model for parallel chains forming a square lattice (i.e., the lattice has tetragonal symmetry):

$$H = J \sum_{i,\boldsymbol{r}} \boldsymbol{S}_{i,\boldsymbol{r}} \cdot \boldsymbol{S}_{i+1,\boldsymbol{r}} + J_\perp \sum_{i,\boldsymbol{r},\boldsymbol{\delta}} \boldsymbol{S}_{i,\boldsymbol{r}} \cdot \boldsymbol{S}_{i,\boldsymbol{r}+\boldsymbol{\delta}} . \quad (1)$$

Here i and $\boldsymbol{r}$ label lattice sites along the chain (z) and perpendicular (x, y) directions, $\boldsymbol{\delta}$ is summed over the two nearest neighbor vectors in the transverse directions, and $\boldsymbol{S}_{i,\boldsymbol{r}}$ is a spin-$1/2$ operator at lattice site $(i,\boldsymbol{r})$. The longitudinal and transverse exchange constants are J (>0) and $J_\perp < 0$, where in order to be close to the situation in $KCuF_3$ a ferromagnetic interchain coupling is used (but all of the subsequent results apply with minor modifications also to $J_\perp > 0$).

I now treat antiferromagnetic order using a mean-field treatment of the interchain coupling. Assuming the order to be oriented along the z direction in spin space, the Hamiltonian (1) transforms into an effective single-chain problem described by

$$H_1 = J \sum_i \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} - h \sum_i (-1)^i S_i^z - 2NJ_\perp m_0^2 . \quad (2)$$

Here N is the number of sites in the chain, $m_0 = (-1)^i \langle S_i^z \rangle$ is the staggered magnetization, and $h = -4J_\perp m_0$. We thus have a one-dimensional antiferromagnet in an effective staggered field h, with the order parameter m_0 to be determined by minimizing the energy. The next step is to transform (2) into a fermionic model by a Jordan-Wigner transformation and to go to the continuum limit. The resulting fermionic model then is described by

$$H_2 = \int dz \, [-iv(\psi_L^\dagger \partial_z \psi_L - \psi_R^\dagger \partial_z \psi_R) - h(\psi_L^\dagger \psi_R + \psi_R^\dagger \psi_L) + 2g \psi_L^\dagger \psi_R^\dagger \psi_R \psi_L] - 2NJ_\perp m_0^2 . \quad (3)$$

Here $\psi_{L,R}$ are standard fermion field operators for left- and right-moving fermions. Equation (3) can be recognized as the massive Thirring model for which an exact Bethe ansatz solution exists [13]. The parameter v determines the velocity of the low-lying excitations, and by

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comparing with the exactly known (for $h = 0$) spectrum of H_1 can be fixed as $v = \pi Ja/2$ where a is the lattice constant. The proper identification of g is more delicate because the underlying fermionic lattice model has rather strong interaction, and a naive transition to the continuum limit therefore is uncontrolled. One can, however, notice that the exact solution of H_2 has a mass gap [13]

$$\Delta = he^{\Lambda(1-\gamma)} \frac{\tan \pi\gamma}{\pi(\gamma - 1)}, \tag{4}$$

where $\pi/\gamma = 2\,\mathrm{arccot}(-g/2v)$ parametrizes the interaction and Λ is the "rapidity cutoff" [13]. Requiring further that the total particle number is independent of m_0, one has $\Lambda \propto \ln(1/h)$, i.e., $\Delta \propto h^\gamma$. On the other hand, standard scaling relations for the lattice Hamiltonian H_1 imply a mass gap $\propto h^{2/3}$, i.e., we have $\gamma = 2/3$. Thus the parameters in H_2 are

$$g = 2v = \pi Ja\,. \tag{5}$$

It is now straightforward to obtain the variation of the ground state energy per site with m_0 as

$$E(m_0) - E(m_0 = 0) = -2J_\perp m_0^2 - \frac{7}{5\sqrt[3]{4}} a v n_0^{2/3} \left(\frac{h}{v}\right)^{4/3}, \tag{6}$$

where n_0 is the fermion density, equal to $1/(2a)$ in the original lattice model. Minimizing with respect to m_0 one finds immediately the equilibrium value of the staggered magnetization as

$$m_0 = \frac{28}{15}\left(\frac{14}{15\pi}\right)^{1/2}\left(\frac{|J_\perp|}{J}\right)^{1/2} \approx 1.017\left(\frac{|J_\perp|}{J}\right)^{1/2}. \tag{7}$$

The mass gap then is

$$\Delta = \frac{56\sqrt{3}}{5\pi}|J_\perp| \approx 6.175|J_\perp|\,. \tag{8}$$

The results (7) and (8) will be compared to experiment below.

We now turn to the spin dynamics. The appropriate generalization of the above mean-field approximation then is an RPA treatment of the interchain interaction. In the ordered phase translational symmetry is broken, and therefore there are umklapp processes coupling modes at wave vectors $\boldsymbol{q}$ and $\boldsymbol{q} + \boldsymbol{Q}$ [where $\boldsymbol{Q} = (0, 0, \pi/a)$]. The transverse susceptibility then is a 2×2 matrix given by

$$\chi(\boldsymbol{q}, \omega) = \frac{\chi(q_z, \omega)}{1 - 2|J_\perp|(\cos q_x + \cos q_y)\chi(q_z, \omega)}, \tag{9}$$

where

$$\chi(q_z, \omega) = \begin{pmatrix} \chi_n(q_z, \omega) & \chi_u(q_z, \omega) \\ \chi_u(q_z, \omega) & \chi_n(q_z + \pi, \omega) \end{pmatrix} \tag{10}$$

is the susceptibility matrix of the one-dimensional model H_1. Explicit expressions for $\chi_{n,u}$ are not known; however, a great deal can be learned from general symmetry properties of H_1 and H_2: (i) By spin rotational invariance the magnitude of the staggered magnetization is independent of the orientation of the staggered field in H_1; this implies $\chi_n(\pi, 0) = -1/(4J_\perp)$ and guarantees the existence of a Goldstone mode (spin wave) in the ordered state. (ii) From the equation of motions derived from H_1 one finds $\chi_n(0, \omega) = (h^2/\omega^2)[\chi_n(\pi, \omega) - \chi_n(\pi, 0)]$, $\chi_u(0, \omega) = \chi_u(\pi, \omega) = (h/\omega)[\chi_n(\pi, \omega) - \chi_n(\pi, 0)]$, at $q_z = 0, \pi$ everything is thus determined by $\chi_n(\pi, \omega)$ alone. (iii) When all relevant energies $(\omega, \Delta, \ldots)$ are much smaller than J the relativistic invariance of H_2 can be used: The operators $S + (q_z \approx \pi)$ and $S + (q_z \approx 0)$ are a Lorentz scalar and vector, respectively. Consequently, $\chi_n(\pi + q, \omega)$ is a function of $\omega^2 - v^2q^2$ only, and $\chi_u(q, \omega) = [h\omega/(\omega^2 - v^2q^2)][\chi_n(\pi + q, \omega) - \chi_n(\pi, 0)]$. The full three-dimensional $\chi(\boldsymbol{q}, \omega)$ thus is entirely determined by $\chi_n(\pi + q, \omega)$ alone, as long as q_z is in the vicinity of 0 or π and $\omega \ll J$.

We can further use the known spectrum of H_2 [13] to determine the form of $\chi_n(\pi + q, \omega)$: This function involves intermediate states where the z component of the magnetization has increased by $\Delta S^z = 1$, corresponding to an added fermion in the Thirring model language. These excitations have energy Δ, leading to a pole. There are of course also multiple-excitation contributions, leading to a continuous spectrum. The lowest such excitation comes from a combination of the elementary $\Delta S^z = 1$ excitation with an $\Delta S^z = 0$ excitation which also has energy Δ (see the discussion of the longitudinal excitations below), thus leading to a threshold at $\omega = 2\Delta$. Notice that the elementary excitations creating this continuum are thus quite different from the spinons responsible for the lowest continuum of an isolated chain [1]. Multiple-excitation continua of course also exist, with thresholds $n\Delta, n \geq 3$. One thus can write

$$\chi_n(\pi + q, \omega) = \frac{z}{\Delta^2 + v^2q^2 - \omega^2} + f(\omega^2 - v^2q^2), \tag{11}$$

where the unknown function $f(x)$ contains contributions from the many-excitation continua, has a threshold singularity at $x = 4\Delta^2$, and is real below the threshold. The constant z ensures that $\chi_n(\pi, 0) = -1/(4J_\perp)$.

The excitation spectrum now is given by the singularities of $\chi(\boldsymbol{q}, \omega)$, Eq. (9), and in particular the low-energy $(\omega < 2\Delta)$ states are found from poles. One finds the following: (i) For propagation along the chain there is a spin wave mode with $\omega(0, 0, q_z) = v(J_\perp)|q_z|$, where the spin-wave velocity is only weakly affected by interchain coupling: e.g., neglecting f in Eq. (11) (the "single mode approximation," SMA), $v(J_\perp) = v/\sqrt{1 + h^2/\Delta^2}$. (ii) For transverse wave vector $(\pi, 0)$ and more generally on the whole line $\cos q_x + \cos q_y = 0$ the spin wave frequency is entirely determined by the one-dimensional result, and therefore from Eq. (11) $\omega(\pi, 0, \pi) = \Delta$, i.e., the mass gap is directly accessible experimentally and proportional to $J_\perp$. (iii) In the SMA the spin-wave dispersion is $\omega(\boldsymbol{q})^2 = \Delta^2[1 - (\cos q_x + \cos q_y)/2] + v^2q_z^2$. In the

relevant case $|J_\perp| \ll J$ the $\boldsymbol{q}$ dependence of the transverse part that of standard spin wave theory, taking into account f will lead to a modified relation in the transverse directions. (iv) In the SMA, the static susceptibility is $\chi(0,0) = 14\pi/(405J) \approx 0.109/J$, taking into account f will decrease this value. One can notice that for $J_\perp \to 0$ one expects to recover the purely one-dimensional result $\chi(0,0) = 1/(\pi^2 J) \approx 0.101/J$. This suggests that the SMA is a rather good description of low-energy properties.

There also are longitudinal excitations corresponding to oscillations of m_0 about its mean value. Within the RPA, the longitudinal susceptibility χ_L is given by a formula analogous to Eq. (9). From Eq. (6) $\chi_L(\pi,0) = -1/(12J_\perp)$, and the frequency dependence can again be obtained from the excitation spectrum of the massive Thirring model: Here excitations with $\Delta S^z = 0$ intervene, i.e., particle-hole pairs. Naively, one expects a continuum above a gap 2Δ; however, because of the interaction in H_2, the lowest excitation is actually an excitonic bound state at energy Δ, [13] i.e., at the same energy as the lowest $\Delta S^z = \pm 1$ excitation [14]. The one-dimensional χ_L then has the same form as χ_n, Eq. (11), and in the SMA, there then is a longitudinal mode at $\omega_L^2(\boldsymbol{q}) = \Delta^2[1 - (\cos q_x + \cos q_y)/6] + v^2 q_z^2$. Going beyond the SMA, the already relatively weak dispersion in the transverse directions would be further reduced. Notice that at $\boldsymbol{q}_\perp = (\pi,0)$ longitudinal and transverse modes are degenerate.

In this approximation, the longitudinal mode is well defined. This is, however, an oversimplification: A longitudinal mode can decay into two spin waves [15]. To get a quantitative estimate of this effect I have obtained an effective Ginzburg-Landau description of the model via a Hubbard-Stratonovich transformation of the interchain interaction in Eq. (1). At the Gaussian level expressions like Eq. (10) for the transverse and longitudinal susceptibilities are obtained, but higher order corrections can also be studied systematically. In particular, a longitudinal excitation can now decay into two spin waves, and there is also important spin-wave–spin-wave scattering. Spin rotation invariance imposes that the matrix elements for these processes at low energies are given by $\chi_L(\pi,0)/h$ and $\chi_L(\pi,0)/h^2$, respectively. Taking these processes into account in an RPA-like fashion, the longitudinal susceptibility takes the form

$$\chi_L(\boldsymbol{q},\omega) = \frac{\chi_L(q_z,\omega) + \Sigma(\boldsymbol{q},\omega)}{1 - 2|J_\perp|(\cos q_x + \cos q_y)[\chi_L(q_z,\omega) + \Sigma(\boldsymbol{q},\omega)]}. \tag{12}$$

This form still retains one-dimensional relativistic invariance: $\chi_L((\boldsymbol{q}_\perp, q_z),\omega) = \chi_L((\boldsymbol{q}_\perp,0),(\omega^2 - v^2 q_z^2)^{1/2})$. Numerical results for $\mathrm{Im}[\chi_L]$ (which determines the neutron scattering intensity) are shown in Fig. 1. One notices in particular a very sharp feature very close to the pole of the unrenormalized ($\Sigma = 0$) propagator. Numerically the width of this peak is found of order 0.01Δ, the transverse mode remains thus rather well defined even when decay into spin waves is taken into account. This can be attributed to the low frequency of the mode and the limited phase space available for decay into spin waves. In addition to the peak, one also notices an incoherent background which for $q_z = \pi$ extends down to zero energy.

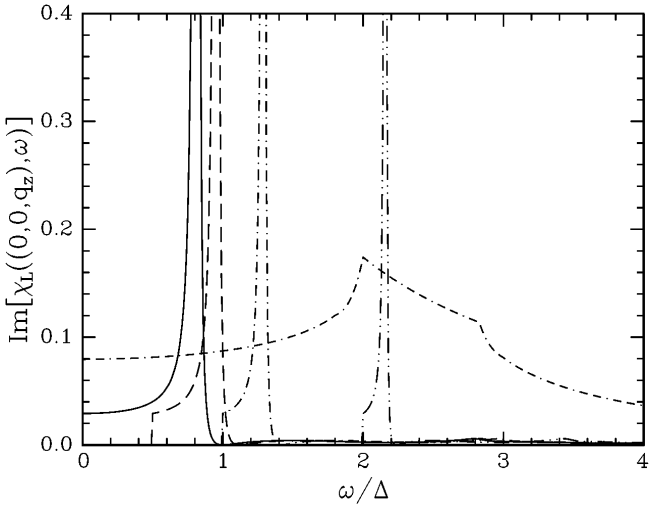

FIG. 1. Frequency dependence of the imaginary part of the longitudinal susceptibility (in units of $1/|J_\perp|$) at $\boldsymbol{q} = (0,0,\pi + q_z)$ for $q_z = 0$ (full line), $vq_z = \Delta/2$ (dashed), $vq_z = \Delta$ (dash-dotted), and $vq_z = 2\Delta$ (dash-double-dot). Also shown is the two-spin-wave cross section at $q_z = 0$, according to Ref. [7] (double-dash-dot).

Finally, the Néel temperature can be determined in this approach from the divergence of the static susceptibility at $\boldsymbol{q} = (0,0,\pi)$ as a function of temperature. The single-chain susceptibility is given by [16,17] $\chi(\pi,0;T) = A/(JT)\ln^{1/2}(\Lambda J/T)$, where numerical calculations give [18] $A \approx 0.32, \Lambda \approx 5.8$. Equation (9) then implies the relation

$$|J_\perp| = \frac{T_N}{4A\ln^{1/2}(\Lambda J/T_N)}. \tag{13}$$

The above results for staggered magnetization, excitation spectrum, and Néel temperature are now compared to experimental results on $KCuF_3$. From the excitation spectrum above T_N the exchange constant along the chains is $J = 34\,\mathrm{meV}$. From the measured [6] $m_0 = 0.25$ Eq. (7) then gives $J_\perp = -J/16 = -2.1\,\mathrm{meV}$. From Eq. (8) then follows the first prediction of the present theory: $\Delta = 13\,\mathrm{meV}$. From Eqs. (9) and (10) Δ is the spin wave frequency at $\boldsymbol{q}_1 = (\pi,0,\pi)$. Experimentally [6] $\omega(\boldsymbol{q}_1) = 11.5\,\mathrm{meV}$, in rather good agreement with the prediction. A second nontrivial prediction of the present theory is the existence of a longitudinal mode at $\omega_L(0,0,\pi) \approx \sqrt{2/3}\Delta \approx 10\,\mathrm{meV}$. It is then tempting to associate the

sharp rise in the energy-dependent neutron scattering intensity (which does not differentiate between transverse and longitudinal modes) observed around $\omega = 10$ [6,7] with this mode. On the other hand, it is not clear how the two-spin-wave process proposed in Ref. [7], which is rather featureless around $\omega = \Delta$ (see Fig. 1), can account for this result. It would clearly be of interest to study this point in more detail.

Using the estimated values for J, $J_\perp$ Eq. (13) gives an estimate for the Néel temperature: $T_N \approx 60\,\mathrm{K}$, overestimating the experimental result ($T_N = 39\,\mathrm{K}$) by about 50%. This discrepancy is in part due to the fact that logarithmic correction terms, enhancing the tendency to ordering, are included in Eq. (13) but neglected in the initial fermionic model, Eq. (3). In the fermionic language, the logarithmic terms come from an extra umklapp interaction [19] $g_u(\psi_R^\dagger \psi_R^\dagger \psi_L \psi_L + \text{H.c.})$ in Eq. (3). These terms destroy the solvability of the fermionic model, but can be taken into account perturbatively [17]. To lowest order then the $h^{4/3}$ term in Eq. (6) is multiplied by a factor $[1 + y_0 \ln(v/\Delta)]^{1/3}$, with $y_0 = g_u/(\pi v)$. With $y_0 \approx 0.25$ [17] the experimental value of m_0 leads to $J_\perp = -1.6$ meV. From this $\Delta = 11.4$ meV, very close to the experimental value, and $T_N = 47\,\mathrm{K}$. The remaining discrepancy between theory and experimental values of T_N may well be related to thermal fluctuation effects neglected here and which can be estimated to be on the level of 10% [12]. Experiment indeed indicates persistence of short-range order well above T_N [6,7].

It is also interesting to consider the case of Sr_2CuO_3 [8,9]. From the experimental $T_N \approx 5\,\mathrm{K}$ and the estimated $J \approx 220$ meV one gets $J_\perp \approx 0.12$ meV. This then leads to a predicted spin wave energy at $\boldsymbol{q} = (\pi, 0, \pi)$ of $\Delta = 1.1$ meV and $m_0 = 0.036$. This last number is consistent with the experimental upper limit $m_0 \leq 0.05$ [9]. It should, however, be noticed that in Sr_2CuO_3 there is considerable structural anisotropy in the directions perpendicular to the chains, and the value derived for $J_\perp$ therefore is an average over transverse directions.

The continuum limit used in the present treatment is valid if the correlation length $\xi = v/\Delta \approx v/(\pi T_N)$ is large compared to the lattice constant. For the two compounds discussed above one finds $\xi = 5a$ and $\xi = 320a$, satisfying this criterion. The mean-field treatment of the interchain coupling leads to $T_N \propto J_\perp$, $m_0 \propto \sqrt{|J_\perp|/J}$, up to logarithmic corrections. These relations are consistent with general scaling arguments [20]. On the other hand, the frequently used renormalized spin wave theory [21] predicts $T_N \propto \sqrt{J|J_\perp|}$, $m_0 \propto 1/\ln(J/|J_\perp|)$, in clear contradiction with scaling. Apart from the quantitatively satisfying description of $KCuF_3$, the present theory thus also seems more satisfactory on grounds of consistency with generally valid arguments.

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