Model for Quasi-One-Dimensional Antiferromagnets: Application to $CsNiCl_3$

Ian Affleck

Canadian Institute for Advanced Research and Physics Department, University of British Columbia, Vancouver, British Columbia, Canada V6T 2A6

(Received 7 October 1988)

The approximate mapping of the antiferromagnetic Heisenberg chain onto a (1+1)-dimensional quantum field theory indicates that the ground state is disordered with a finite gap for integer spin. We study a lattice of coupled (1+1)-dimensional field theories as a model for weakly coupled spin chains. This model offers a possible resolution to the mystery of the upper mode in the low-temperature ordered phase of $CsNiCl_3$, whose polarization is inconsistent with spin-wave theory.

PACS numbers: 75.10.Jm, 75.30.Ds, 75.50.Ee

It was first argued by Haldane that integer-spin antiferromagnetic chains should have a finite gap above the ground state, to a massive triplet excitation.[1] After some initial controversy this has been verified by numerical simulation[2,3] and by a rigorous proof for a solvable model.[4]

$CsNiCl_3$ is a highly one-dimensional and highly isotropic spin-1 antiferromagnet. Neutron-scattering data above the Néel temperature is indicative of a gap for the one-dimensional system.[5] Interchain couplings lead to a significant dispersion for the gap in the directions transverse to the chains. They are also responsible for Néel order at low enough temperatures. In Ref. 5 the ordered phase was analyzed by use of standard spin-wave theory. On the whole this seemed to give a reasonable description. However, in order to fit the observed ordered-phase finite-gap excitation branch, it was necessary to assume a rather large anisotropy parameter, too large to be consistent with the observed canting angle or spin-flip field. Furthermore, this excitation has a different polarization than that predicted by spin-wave theory.[6] In the disordered phase at higher T, a simple model of coupled chains was used to explain the existence of a finite gap and a transverse dispersion.

In this paper we will develop a more complete theory of coupled chains which should be applied in both the Néel *and* disordered phases. Whether or not the system orders at $T=0$ depends on the strength of the interchain coupling, relative to the gap of the single chain. In the ordered phase the massive triplet of the single chain splits up into two massless Goldstone modes and a massive "longitudinal mode" (corresponding to oscillations in the *magnitude* of the sublattice magnetization) with a finite decay width. We identify the observed finite-gap branch with this longitudinal mode. An increase in the temperature leads to a renormalization of the parameters such that a transition to the disordered phase eventually occurs. The gap of the longitudinal mode should vanish at T_N. Above T_N the Goldstone modes and longitudinal mode are combined into a triplet, with a temperature-dependent gap which increases from zero at T_N.

As shown in Ref. 1, the large-s *integer-spin* chain, $H=\sum_i [2J\mathbf{S}_i\cdot\mathbf{S}_{i+1}+D(S_i^z)^2]$, is described at low energies by the O(3) nonlinear σ model, with Lagrangian density

$$L_1=\tfrac{1}{2}g[(\partial\boldsymbol{\phi}/\partial t)^2/v-v(\partial\boldsymbol{\phi}/\partial z)^2]-Ds^2(\phi^z)^2.$$

(We set $\hbar=1$, z is the distance along the chain.) Here the triplet field $\boldsymbol{\phi}$ obeys the constraint $\boldsymbol{\phi}^2=1$. $\boldsymbol{\phi}$ and the rotation generator, $\mathbf{l}\equiv\boldsymbol{\phi}\times\boldsymbol{\phi}$, represent the Fourier modes of the spin operators near momentum π, the ordering wave vector, and 0, respectively. $\mathbf{S}(z)\approx(-1)^z s\boldsymbol{\phi}+\mathbf{l}/v$. The continuum-limit field theory gives a description of the low-energy modes occurring at these two points in momentum space. The velocity of "light" of the quantum field theory is that of spin-wave theory: $v=4Js$. The coupling constant measures the strength of the quantum fluctuations: $g=2/s$. A perturbative treatment of this field theory involves expanding around the ordered state $\boldsymbol{\phi}\approx(\phi_1,\phi_2,1)$. The fields ϕ_1 and ϕ_2 are massless Goldstone modes, for $D=0$. However, it is known on general grounds that the severe infrared divergences of (1+1)-dimensional quantum field theory prevent the spontaneous breaking of continuous symmetries, and invalidate perturbation theory. Our understanding of the actual behavior of this model is based on a combination of renormalization-group arguments, the exact solution in the large-n limit (where $\boldsymbol{\phi}$ is taken to be an n-component unit vector)[7] and the proposed exact S matrix.[8] The spectrum consists of a triplet of energy gap $\Delta\approx ve^{-\pi s}$.

The corresponding correlation length, $e^{\pi s}\gg 1$ at large s, justifying the continuum approximation. This mapping may not be very accurate at $s=1$ but should be at least qualitatively correct and provides a useful phenomenological model. For $s=1$ the correlation length, $\xi\approx v/\Delta$, is about five lattice spacings, long enough to partially justify the continuum approximation.

We now add a nearest-neighbor interchain coupling, J'. For $J'\ll J$, and $s\gg 1$, we may represent the low-energy degrees of freedom with $k_z\approx 0$ or π by a set of σ fields. Representing the ith chain by a σ field $\boldsymbol{\phi}_i$, we obtain the system of coupled (1+1)-dimensional field

theories:

$$L=\sum_i L_1(\phi_i)-J's^2\sum_{\langle i,j\rangle}\phi_i(z)\cdot\phi_j(z),\quad \phi_i(z)^2=1\,,$$

where the sum is over the nearest-neighbor chains, each pair occurring *twice* and $L_1(\phi_i)$ is the Lagrangian density for a single chain. Since the model is now (3+1) dimensional, spontaneous symmetry breaking is possible. However, we do not expect it to occur for sufficiently small J' since the disordered phase is stabilized by the existence of a finite mass. It is apparently not possible to find the exact solution of this model but various more or less reliable approximations are possible. One approach is to take the large-n limit. This will be discussed in detail elsewhere. A simpler and more intuitive approach is to relax the constraint on the field ϕ and introduce an explicit mass term and a ϕ^4 term for stability:

$$L=\sum_i[(\partial\phi_i/\partial t)^2/2v-v(\partial\phi_i/\partial z)^2/2-(\Delta^2/2v)\phi_i^2-2Ds(\phi_i^z)^2-(\lambda/4)(\phi_i\cdot\phi_i)^2]-2J's\sum_{\langle i,j\rangle}\phi_i(z)\cdot\phi_j(z)\,.$$

Δ represents the mass gap which arises dynamically in the nonlinear σ model. Here we have rescaled ϕ by a factor of $\sqrt{g}$ so that $\mathbf{S}(z)\approx(-1)^z(2s\phi)^{1/2}+(2/s\phi)^{1/2}\times\dot{\phi}/v$. We see that the ground state will have spontaneous symmetry breaking (nonzero ϕ) for J' bigger than a critical value. For $D=0$, $J's>k\Delta^2/v$, where k is a pure number which depends on the lattice type. λ controls the size of the sublattice magnetization, $\langle\phi\rangle$.

Of course other terms could be added to H as well, for instance higher powers of ϕ. This particular form is used in the spirit of a Landau-Ginsburg Hamiltonian. The five phenomenological parameters, v, J', Δ, λ, and D can be fitted to experiment (or numerical simulations) as discussed below.

We now consider the particular case of a triangular lattice of chains as occurs in $CsNiCl_3$. Following the standard notation of Ref. 5, the chains are aligned along the z axis with spacing $c/2$ between neighboring Ni atoms. The primitive vectors for the triangular lattice are $a(1,0,0)$, $a(-\frac{1}{2},\sqrt{3}/2,0)$, and $a(-\frac{1}{2},-\sqrt{3}/2,0)$. We set $c/2$ and a to 1 so that the wave vector $\mathbf{k}$ is dimensionless. $\mathbf{k}$ is related to the $\mathbf{Q}$ variables used in Ref. 5 by $Q_a\equiv k_x/2\pi$, $Q_b=(-k_x+\sqrt{3}k_y)/4\pi$, and $Q_c=k_z/\pi$.

If we assume a three-sublattice spin ordering, with $\langle\mathbf{l}\rangle=0$, corresponding to Néel order along the chains, and $\langle\phi\rangle$ having the values ϕ_i $(i=1,2,3)$ on the three inequivalent chains, then the semiclassical ground state is obtained by minimizing the energy per chain, per unit length:

$$V(\phi_i)=\tfrac{1}{3}\sum_{i=1}^{3}[\Delta^2\phi_i^2/2v+2Ds(\phi_i^z)^2+\lambda\phi_i^4/4]+4J's[\phi_1\cdot\phi_2+\phi_1\cdot\phi_3+\phi_2\cdot\phi_3]\,.$$

This classical potential is a special case of the one considered in Ref. 9. For $D=0$, the minimum of V has $\phi_i=0$, for $\Delta^2>12J'sv$. For $\Delta^2<12J'sv$, $\phi_i^2\equiv\phi_0^2=(12J's-\Delta^2/v)/\lambda$ and the ϕ_i point in three different directions lying in a plane with relative angles $2\pi/3$. We choose the particular ground state $\phi(\mathbf{x})=\phi_0(\sin\mathbf{k}_2\cdot\mathbf{x},0,\cos\mathbf{k}_2\cdot\mathbf{x})$, where $\mathbf{k}_2\equiv(2\pi/3,2\pi/\sqrt{3},0)$. For $D<0$, ϕ_1 points along the positive z axis and ϕ_2 and ϕ_3 make an angle $\pm\theta$ with the negative z axis which is less than $\pi/3$: $\cos\theta=\frac{1}{2}(1+D/6J')$ (for $-D<3J'$). (This is identical to the result in Ref. 5 after the correction of a factor of 2 error.) The spin components in the xy plane line up with the lattice vectors due to weak sixth-order anisotropic terms[10] in V, which we omit from this discussion.

$CsNiCl_3$ displays this type of order at $T<4.4$ K. This implies that the interchain coupling is sufficiently strong to overwhelm the Haldane gap: $12J'vs>\Delta^2$. The observed value of θ at $T=1.6$ K is 59 K, corresponding to $D/6J'=-0.0292$. An estimate based on the spin-flop field gives[6,11] $D/6J'=-0.07$. These values of D are negligible and we will set $D=0$ in what follows. In order to fit spin-wave theory to the observed finite-gap mode in the ordered phase, a much larger "effective" D was used in Ref. 5: $D/6J'=-0.36$. It was speculated in Ref. 5 that this larger "effective" D reflected the importance of the one-dimensional many-body effects, i.e., the Haldane gap. In this work we will explain this mode by taking into account the one-dimensional effects and using $D=0$.

We now turn to the dispersion relation for this system, in the ordered phase. This is calculated in the semiclassical approximation where we simply expand the Lagrangian to quadratic order in the small fluctuations of the fields away from their expectation values. The resulting Hamiltonian can be diagonalized exactly. These small fluctuations are of two distinct types: transverse fluctuations where the magnitude of ϕ is unchanged but its direction oscillates, and longitudinal fluctuations where the direction is unchanged but the magnitude oscillates. In the semiclassical approximation, these two types of fluctuations are decoupled. The transverse fluctuations are precisely the usual excitations of spin-wave theory, in a low-energy approximation, valid for $J'\ll J$ and $k_z\approx 0$ or π. The longitudinal fluctuations, however, are not obtained in standard spin-wave theory, since the quantum spins are of fixed length: $\mathbf{S}^2=s(s+1)$. However, it is well known that upon making a renormalization-group transformation, such "hard spins" are usually turned into soft ones. The spectrum of the one-dimensional model in the low-energy, large-s limit is known[7,8] to consist of a triplet. Essentially the constraint on the magnitude of the field ϕ becomes irrelevant at low energies and the Landau-Ginsburg theory gives the right spectrum. We expect this triplet to break up into a pair of Goldstone modes plus a longitudinal mode when we pass into the ordered phase; i.e., the sys-

tem retains a "memory" of its triplet spectrum. Although the longitudinal mode will have a finite decay width, the width-to-mass ratio vanishes as the critical point is approached from the ordered side. This can be seen from the fact that the zero-temperature phase transition is in the four-dimensional universality class and thus the critical theory is the zero-coupling limit of the Landau-Ginsburg model. The width-to-mass ratio is proportional to the coupling constant, and so vanishes at the critical point. This argument would apply equally well to a three-dimensional antiferromagnet which was sufficiently close to its zero-temperature disordering transition due to frustration. Since λ vanishes logarithmically slowly as the critical point is approached, the issue of whether a given system will have a well-defined longitudinal mode becomes a somewhat heuristic one.

We write the small fluctuations of $\boldsymbol{\phi}$ as $\boldsymbol{\phi}_i = (1+\psi_L/\phi_0)[\psi_1, \psi_2, \psi_0 - (\psi_1^2+\psi_2^2)/2\phi_0]$, for i on the first sublattice. ψ_1 and ψ_2 are transverse fluctuations and ψ_L is longitudinal. On the second and third sublattice the parametrization is obtained from this by rotating by $\pm 2\pi/3$. By our expanding L to quadratic order in small fluctuations we find it is diagonal with frequencies:

$$\omega_1(\mathbf{k}) = \{(vk_z)^2 + 4vJ's[3 - f(\mathbf{k}_\perp)]\}^{1/2},$$

$$\omega_2(\mathbf{k}) = \{(vk_z)^2 + 4vJ's[3 + 2f(\mathbf{k}_\perp)]\}^{1/2},$$

$$\omega_L(\mathbf{k}) = \{(vk_z)^2 + 4vJ's[9 - f(\mathbf{k}_\perp)] - 2\Delta^2\}^{1/2},$$

where, following Ref. 5, $f(\mathbf{k}_\perp) \equiv \cos k_x + \cos(-k_x + \sqrt{3}k_y)/2 + \cos(-k_x - \sqrt{3}k_y)/2$. Note that the introduction of σ-model variable doubles the length of the unit cell in the z direction so that k_z and $k_z + \pi$ are identified. $\mathbf{k}_\perp$ runs over the entire paramagnetic Brillouin zone; folding it back into the antiferromagnetic zone gives a total of nine branches, corresponding to a triplet from each of the three chains in the unit cell. ω_1 vanishes at $\mathbf{k} = \mathbf{0}$, and ω_2 at $\mathbf{k} = \pm \mathbf{k}_2$. These three gapless modes correspond to the three Goldstone bosons resulting from the complete breaking of the rotational symmetry. The longitudinal mode has its minimum frequency, $\Delta_L = (24J'vs - 2\Delta^2)^{1/2}$ at $\mathbf{k} = \pm \mathbf{k}_2$. This gap vanishes when J' has its critical value at which Néel order disappears.

The neutron-scattering cross section for k_z near π, ignoring finite width effects discussed below,

$$S(\mathbf{k},\omega) \propto g(\mathbf{k})^2(1-\hat{k}_y^2)[1/\omega_2(\mathbf{k}')]\delta[\omega - \omega_2(\mathbf{k}')]$$
$$+ g(\mathbf{k})^2(1+\hat{k}_y^2)\tfrac{1}{4}\sum_{\pm}\{[1/\omega_1(\mathbf{k}' \pm \mathbf{k}_2)]\delta[\omega - \omega_1(\mathbf{k}' \pm \mathbf{k}_2)] + [1/\omega_L(\mathbf{k}' \pm \mathbf{k}_2)]\delta[\omega - \omega_L(\mathbf{k}' \pm \mathbf{k}_2)]\},$$

where $\mathbf{k}' \equiv (k_x, k_y, k_z - \pi)$ and $g(\mathbf{k})$ is the neutron-scattering form factor. The ω_1 and ω_2 terms are those of ordinary spin-wave theory,[5] in our approximation ($J' \ll J$, k_z near π), but the ω_L mode is new. It has purely xz polarization since it corresponds to fluctuations of the spins along their ordering directions. *Thus it provides a possible explanation of the finite-gap mode, observed in $CsNiCl_3$ which could not be understood using conventional spin-wave theory.* The finite D spin-wave spectrum is shown in Fig. 1 and the predictions of the present theory, together with the experimental data are shown in Fig. 2. From the observed transverse modes we estimate $v \approx 4J = 1.38$ Thz, $J' = 0.006$ THz, essentially the same values as in Ref. 5. The observed longitudinal gap, $\Delta_L \approx 0.19$ THz, then implies a gap for a single chain of $\Delta \approx 0.285$ THz, essentially identical to the numerical finite-chain estimate[3] of $0.40(2J)$. Given the very approximate nature of our model, this agreement must be regarded as somewhat fortuitous.

The decay amplitude of the longitudinal mode is given, to lowest order in the coupling constant λ by the cubic coupling between the longitudinal and transverse fields in the Néel phase: $L_{\text{int}} = \psi_L \sum_{a=1}^{2} (\dot{\psi}^2 a^2/v - v\psi' a^2)/\phi_0$. We find a decay rate at $\mathbf{k} = \mathbf{k}_0$ (full width at half maximum) $\Gamma \approx \Delta_L/16\phi_0^2 \approx 0.09$ THz, less than the experimental resolution in Ref. 5.

We will discuss the finite-temperature behavior of this model in detail elsewhere. However, the main conclusions are fairly transparent. Increasing the temperature will restore the rotational symmetry. This effect can be described in terms of temperature-dependent renormalizations of the parameters in the Hamiltonian; in particular, we expect Δ to increase with T. An analysis

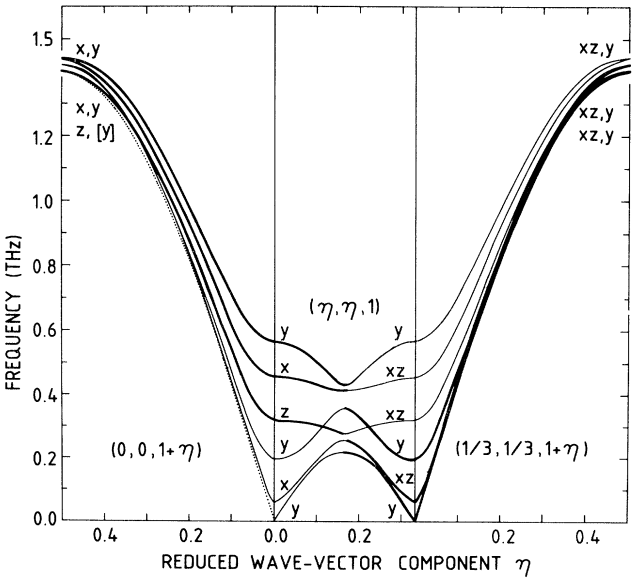

FIG. 1. The spectrum predicted by conventional spin-wave theory with $D/6J' = -0.36$, plotted vs (Q_a, Q_b, Q_c) from Ref. 5. Polarizations are indicated and line thickness corresponds to intensity.

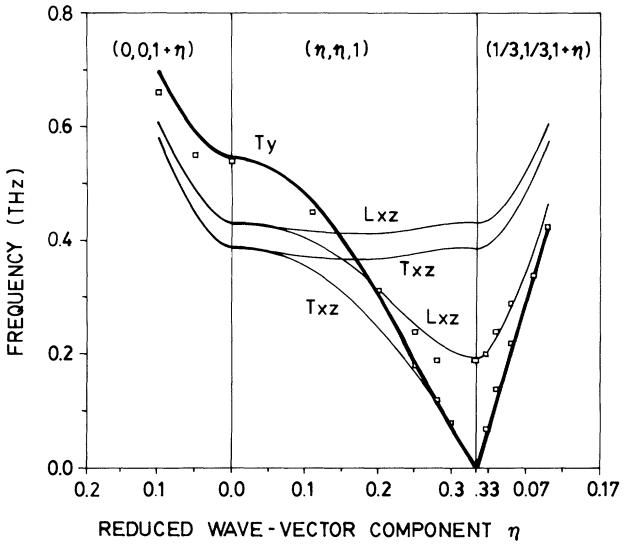

FIG. 2. The spectrum predicted by the present theory with $D=0$. T and L label transverse and longitudinal modes.

of the classical potential, with increasing Δ shows, in agreement with experiment, that for $D<0$ an intermediate phase is first reached in which the z but not the x components of the spins are ordered. At higher T the paramagnetic phase is reached. This follows from the analysis of Ref. 9, since our classical potential is a special case of the one considered there. In the paramagnetic phase, the spectrum consists of a massive triplet, with $\omega_t=[(vk_z)^2+\Delta^2+8f(\mathbf{k}_\perp)J'vs]^{1/2}$, and hence a gap: $\Delta_t=[\Delta^2-12J'vs]^{1/2}$, where all parameters are temperature dependent. Δ_t varies with T, and in particular should vanish as $T\rightarrow T_{N1}$, the upper Néel temperature. The formula for ω_t is identical in form to the one in Ref. 5. A best fit with the data occurs with $J'v(T)=0.00153$ THz, $\Delta(T)=0.32$ THz, at $T\approx 7$–10 K, the same value of Δ estimated in Ref. 5. Thus Δ has renormalized up from 0.28 to 0.32 and J' down from 0.00207 to 0.00153. This is a remarkably gradual renormalization over a temperature range of about 1 THz.

Another prediction of our model is the maximum value of J'/J for which the system will be paramagnetic even at $T=0$ due to the one-dimensional fluctuations: $J'/J<(\Delta/J)^2/48$. By use of the value found above, $(\Delta/J)\approx 0.8$, this gives $J'/J<0.013$. In $CsNiCl_3$, $J'/J\approx 0.017$. $Ni(C_2H_8N_2)_2NO_2ClO_4$, has $J'/J\approx 0.0004$ and may represent an example of such a quantum disordered system.[12]

I would like to thank Bill Buyers and Zin Tun for sharing their unpublished data, many stimulating discussions, and much encouragement and enthusiasm. This research was supported in part by the Natural Sciences and Engineering Research Council of Canada.

[1]F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983), and Phys. Rev. Lett. **50**, 1153 (1983), and J. Appl. Phys. **57**, 3359 (1985); I. Affleck, Nucl. Phys. **B257**, 397 (1985).

[2]R. Botet and R. Jullien, Phys. Rev. B **27**, 613 (1983); R. Botet, R. Jullien, and M. Kolb, Phys. Rev. B **28**, 3914 (1983); M. Kolb, R. Botet, and R. Jullien, J. Phys. A **16**, L673 (1983); M. P. Nightingale and H. W. J. Blote, Phys. Rev. B **33**, 659 (1986); H. J. Schulz and T. A. L. Ziman, Phys. Rev. B **33**, 6545 (1986).

[3]J. B. Parkinson, J. C. Bonner, G. Muller, M. P. Nightingale, and H. W. J. Blote, J. Appl. Phys. **57**, 3319 (1985).

[4]I. Affleck, T. Kennedy, E. H. Lieb, and H. Tasaki, Phys. Rev. Lett. **59**, 799 (1987), and Commun. Math. Phys. **115**, 477 (1988).

[5]W. J. L. Buyers, R. M. Morra, R. L. Armstrong, M. J. Hogan, P. Gerlach, and K. Hirakawa, Phys. Rev. Lett. **56**, 371 (1986); R. M. Morra, W. J. L. Buyers, R. L. Armstrong, and K. Hirakawa, Phys. Rev. B **38**, 543 (1988).

[6]M. Steiner, K. Kakurai, J. K. Kjems, D. Petitgrand, and R. Pynn, J. Appl. Phys. **61**, 3953 (1987); H. Kadowaki, K. Ubukoshi, and K. Hirakawa, J. Phys. Soc. Jpn. **56**, 751 (1987); Z. Tun, W. J. L. Buyers, R. L. Armstrong, E. D. Hallman, and D. Arovas, to be published.

[7]E. Brezin and J. Zinn-Justin, Phys. Rev. B **14**, 3110 (1976).

[8]A. B. Zamalodchikov and A. B. Zamalodchikov, Ann. Phys. (N.Y.) **120**, 11 (1979).

[9]M. L. Plumer and A. Caille, to be published; M. L. Plumer, K. Hood, and A. Caille, Phys. Rev. Lett. **60**, 45 (1988).

[10]Xiadong Zhu and M. B. Walker, Phys. Rev. B **36**, 3820 (1987).

[11]E. Cohen and M. D. Sturge, Solid State Commun. **24**, 51 (1977).

[12]J. P. Renard, M. Verdaguer, L. P. Regnault, W. A. C. Erkelens, J. Rossat-Mignod, and W. G. Stirling, Europhys. Lett. **3**, 945 (1987).