Eur. Phys. J. B **32**, 489–493 (2003)
DOI: 10.1140/epjb/e2003-00129-0

THE EUROPEAN PHYSICAL JOURNAL B

Multiparticle continuum in the excitation spectrum of the S = 1 compound $CsNiCl_3$

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Received 2 October 2002 / Received in final form 19 March 2003
Published online 7 May 2003 –

Abstract. Recent neutron scattering experiments on $CsNiCl_3$ reveal some features that are not well described by the standard nonlinear σ model, nor by numerical simulations, for isolated $S = 1$ spin chains. In particular, in real systems at the antiferromagnetic point of the Brillouin zone, the intensity of the continuum of multiparticle excitations, at $T = 6$ K, is about 5 times greater than predicted. Also, the spin gap is higher and the correlation length is smaller than predicted. We propose a theoretical scenario where the interchain interaction is approximated by an effective staggered magnetic field, and that yields a correct prediction for the observed quantities.

PACS. 75.10.Jm Quantized spin models – 75.50.-y Studies of specific magnetic materials

About twenty years after the famous "Haldane conjecture" [1], quantum spin chains with Heisenberg antiferromagnetic interactions are still attracting much experimental and theoretical interest. Half-integer spin chains have no spin gap and are quantum critical at $T = 0$ with algebraic decay of correlations. Integer spin chains exhibit instead a large, dynamically generated, spin gap, and the correlation functions decay exponentially with a finite correlation length. Haldane's conjecture was proved, in the continuum limit when the isotropic Heisenberg Hamiltonian gets mapped onto an O(3) Nonlinear Sigma Model (NLσM) [2], by the Zamolodchikovs [3] for the integer-spin case and by Shankar and Read [4] for the half-odd-integer-spin case.

For $S = 1$ chains, the excitation spectrum was predicted [3] to be dominated by a well-defined, degenerate $S = 1$ magnon triplet. Such excitations have been observed in several neutron scattering experiments and, in the case of NENP [5], they constitute a large part of the total spectrum.

Subsequent theoretical work [6] has shown that, at the antiferromagnetic point of the Brillouin zone, above the single-particle excitation spectrum, there is a continuum of multiparticle excitations starting at a lower threshold of 3Δ, where Δ is the Haldane gap. Higher in energy there are further contributions due to $(2n + 1), n > 1$ multimagnon excitations. The total integrated three-magnon contribution to the spectral weight near the antiferromagnetic wave vector turns out to be about 3% of the total spectral weight.

These theoretical predictions seem to be at odds with recent experimental measurements on $CsNiCl_3$ that reveal a significant spectral weight in the incoherent multiparticle continuum above the coherent magnon peak. At 6 K the integrated intensity of the continuum around the antiferromagnetic point is about 9(2)% of the total spectral weight [7]. This result is considerably larger than the 1–3% weight predicted by numerical simulations [8] and the O(3) non-linear sigma model for the three-particle continuum [6]. The effects of the coupling between chains have been considered also in the framework of the RPA, but no significant increase of the continuum was found [6]. On the other hand, Tsvelik's Majorana Fermion theory [9] does indeed yield a consistent three-particle-scattering continuum [6], but it would require strong biquadratic exchange interactions of the form $(\mathbf{S}_i \cdot \mathbf{S}_{i+1})^2$ that are not in accordance with the measurements on $CsNiCl_3$ [7].

Other measured quantities at $T = 6$ K show appreciable deviations from analytical and numerical predictions at finite temperature on isolated chains [10,11]. In units of the intrachain exchange coupling J the measured gap Δ is higher ($\Delta = 0.54$) and the correlation length ξ is shorter ($\xi = 4.0$) than the corresponding theoretical values predicted by the NLσM for isolated chains ($\Delta = 0.49$ and $\xi = 4.6$ respectively). Moreover, with an estimated value for the spin-wave velocity of $c = 2.5$, these values violate the relation $\xi\Delta = c$ predicted by the Single-Mode Approximation [12].

Finally, recent experimental findings [7] seem to indicate clearly that the multiparticle continuum sets in at a *two*-magnon threshold. This too is at variance with the theoretical predictions, and has led some authors [7] to question the overall validity of an effective field theory

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description in terms of the NLσM and to hypothesize the presence of residual spin-1/2 excitations that could be responsible for the anomalous two-magnon threshold. However, at the present time there is no clear theoretical support for the inclusion of additional spin-1/2 degrees of freedom.

The model Hamiltonian proposed for $CsNiCl_3$ is

$$H = J \sum_{i}^{\text{chain}} \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J' \sum_{\langle ij \rangle}^{\text{plane}} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^z)^2 \ , \quad (1)$$

with an estimated intrachain coupling $J = 2.28$ meV along the c-axis that is much stronger than the interchain coupling in the basal plane $J' = 0.044$ meV [13]. The single-ion anisotropy is estimated to be $D \approx 4$ μeV, small enough that $CsNiCl_3$ may be considered purely isotropic, and we will set $D = 0$ henceforth. Below $T_{\rm N} = 4.85$ K the system undergoes 3D long-range ordering, caused by the interchain interaction. For $T > T_{\rm N}$ the system is regarded as one-dimensional and commonly considered to be a good realization of a single isotropic spin chain, although there is a sizable dispersion perpendicular to the chain direction due to the interchain coupling.

The discrepancies between the predictions of the NLσM and the experimental findings seem to persist [7] from $T = T_N$ up to about $T = T^* \sim 12$ K. Above T^* (of course not a sharp but rather a cross-over temperature) the properties of the system are basically in accordance with the theoretical predictions for an assembly of isolated chains. Similar discrepancies have been recently found also between exact diagonalization studies of $S = 1$ Heisenberg chains and neutron scattering experiments [14]. They become evident at the antiferromagnetic point and at low temperatures ($T < 15$ K), suggesting that three dimensional effects, due to the interchain coupling J', are still important in this range of temperatures. Since at the critical point the $3D$ antiferromagnetic correlation length ξ_{3D} diverges, it is natural to think that there is an intermediate range of temperatures, $T_N < T < T^*$, where ξ_{3D} remains considerably greater than the correlation length ξ_{1D} of the single chain. In other words, crossing the critical point from below does indeed imply destruction of the overall $3D$ long-range order, except on the scale of the $3D$ correlation length ξ_{3D}, as long as the latter remains substantially greater than ξ_{1D}. This fact suggests a simple theoretical scenario, that can reconcile the experimental results with the NLσM description, in which three-dimensional effects are still important and the system is arranged in large domains with non-zero magnetization, even if the total staggered magnetization is zero. Thus, in a mean-field picture, a single chain will experience, in the interval $T_N < T < T^*$, an effective staggered magnetic field generated by the neighboring chains. No matter if the field is not constant along the whole chain, because it varies slowly enough on the scale of ξ_{1D} that it may be considered constant, since the $1D$ chains are short-ranged. The same argument can be applied to the time dependence of fluctuations, which in the vicinity of the critical point are slow enough to consider the effective magnetic field as static. This assumption will not affect the physics of the single chain because of the presence of a relatively large gap (of the order of J) and because, as we will see later, the dynamical structure factor is zero for lower frequencies.

In the mean-field approximation the interchain interaction is treated then as a staggered magnetic field that takes into account the effects of the neighboring chains on the single 1D system. This amounts to the replacement

$$J' \sum_{\langle ij \rangle}^{\text{plane}} \mathbf{S}_i \cdot \mathbf{S}_j \quad \Longrightarrow \quad H_s \sum_i (-)^i S_i^z \ , \quad (2)$$

where the staggered field (we choose it along the c-axis) is evaluated self-consistently by means of the fixed-point equation [15]

$$H_s = z_c J' \, m_s(H_s) \ , \quad (3)$$

z_c is the coordination number in the basal plane and $m_s(H_s)$ is the magnetization curve of a single chain in presence of the external staggered field H_s. In our calculation we fix the energy scale $J = 1$, the interchain coupling $J' = 0.02$ as close as possible to the experimental value for $CsNiCl_3$ and we put $z_c = 3$ because the lattice in the basal plane is triangular.

So, the Hamiltonian we are going to study is

$$\mathcal{H} = \sum_i \left[J \, \mathbf{S}_i \cdot \mathbf{S}_{i+1} \ + \ (-1)^i \, H_s \, S_i^z \right] \ . \quad (4)$$

Following the Haldane mapping [2], we represent the local spin operators as

$$\mathbf{S}_i \approx S(-1)^i \mathbf{n}_i + \mathbf{l}_i \, , \qquad |\mathbf{n}_i|^2 = 1 \, , \quad (5)$$

where $\mathbf{n}_i$ represents the slowly-varying local staggered magnetization and $\mathbf{l}_i$ is the local generator of angular momentum. In this framework, the Zeeman term of equation (4) becomes essentially a linear shift in the $\mathbf{n}$-field along the z-direction, $(-1)^i H_s S_i^z \approx S \, H_s n_i^z$. The staggering factor $(-1)^i$ has the effect that small momenta of the $\mathbf{n}$-field correspond to momenta near π of the original spin variable.

Going then to the continuum limit and integrating out the fluctuation field $\mathbf{l}$ we obtain the O(3) NLσM plus a linear term [16]

$$\mathcal{L} = \frac{1}{2gc} \left(c^2 |\partial_x \mathbf{n}|^2 + |\partial_\tau \mathbf{n}|^2 \right) - S \, H_s n^z - \mathrm{i}\lambda \left(\mathbf{n}^2 - 1 \right) \ , \quad (6)$$

where $g = 2/S$ and $c = 2JS$. A Lagrange multiplier $\lambda(x, \tau)$ has been introduced to implement the local constraint $\mathbf{n}^2(x, \tau) = 1$.

The experiments were performed at temperatures small enough if compared with the exchange interaction $J/k_{\rm B} \approx 26\,{\rm K}$ along the chain. Therefore, the 1D chains (but not the background, of course) are effectively in a low-temperature phase, and this justifies the use of ground state calculations, arguing that a finite temperature treatment [10] will give simply a small correction in the calculated quantities.

For a constant staggered field, the associated saddle point will correspond to a constant value of λ and the self-consistency equation at $T = 0$ is

$$\frac{3g}{2\pi} \ln \left\{ \Lambda\xi + \sqrt{1 + (\Lambda\xi)^2} \right\} = 1 - \left(\frac{Sg}{c} \right)^2 H_s^2 \xi^4 \ , \quad (7)$$

where ξ is the correlation length and Λ is the momentum cutoff of the theory. In the passage to the continuum limit we have lost the correct renormalization of the parameters. In order to restore them, we fix the zero-field values of $\Delta_0 = 0.41048\ J, \xi = 6.03$ (which correspond to $c = \xi\Delta_0 = 2.48\ J$ [17]), known from the DMRG studies [18]. The NLσM coupling has been left to its analytical value $g = 2$. As the field is varied, the cutoff Λ is held fixed to its zero-field value $\Lambda = 0.2072$ and we use equation (7) to determine the value of $\xi = \xi(H_s)$.

The magnetization per site is given by [16]

$$m_s(H_s) = S \langle n^z \rangle = \frac{gS^2}{c} \xi^2(H_s)\ H_s \ . \quad (8)$$

Putting this back into equation (3), we find a stable solution for $H_s = 0.027$, corresponding to $m_s = 0.45$ and $\xi = 4.54$. It is remarkable that even for a small staggered field the response of the system is strong, generating an appreciable magnetization, meanwhile the gap renormalizes upward and the correlation length is getting shorter.

The staggered field breaks explicitly the O(3) symmetry down to O(2) and the quasi-particle propagators in the longitudinal (in the direction of the field) and in the transverse channels differ notably. The transverse propagator agrees with the SMA and is given by

$$G_T(q, \omega_n) = \frac{S^2 gc}{\omega_n^2 + \varepsilon^2(q)} \ , \quad (9)$$

where $\omega_n = 2\pi n/\beta$ are the Matsubara frequencies and $\varepsilon(q)$ is the single-particle energy

$$\varepsilon(q) = \sqrt{c^2 q^2 + \Delta_T^2} \ , \quad (10)$$

where $\Delta_T = c\xi^{-1} = 0.545$ is the transverse gap and $q = 0$ corresponds to the antiferromagnetic point. In the transverse channel the theory is purely bosonic and the spectral weight is fully exhausted by this magnon excitation. In this case, after the analytical continuation to the real axis, the dynamical structure factor

$$S_T(q, \omega) \equiv \frac{1}{\pi} \operatorname{Im} G_T(q, \omega) \quad (11)$$

is simply given by

$$S_T(q, \omega) = \frac{gcS^2}{2\varepsilon(q)} \{ \delta(\omega - \varepsilon(q)) - \delta(\omega + \varepsilon(q)) \} \cdot \quad (12)$$

On the other hand, the longitudinal propagator must be evaluated more carefully considering its connected part and explicit calculations [19] give

$$G_L(q, \omega) = G_T(q, \omega)\ \frac{3\widetilde{\Gamma}(q, \omega)}{3\widetilde{\Gamma}(q, \omega) + 2m_s^2 G_T(q, \omega)} \ , \quad (13)$$

where $\widetilde{\Gamma}(q, \omega)$ is the Fourier transform of the polarization bubble, *i.e.* of the product

$$\Gamma(\mathbf{x} - \mathbf{x}') = \frac{1}{S^4} G_T(\mathbf{x} - \mathbf{x}') G_T(\mathbf{x}' - \mathbf{x}) \ . \quad (14)$$

From the study of the analytic structure of the longitudinal propagator [19] it is possible to calculate the relative longitudinal poles $\omega = \pm\varepsilon_L(q)$, given by the solution of

$$\varepsilon_L^2(q) = \varepsilon_T^2(q) + \frac{(m_s/S)^2}{\Gamma_1(q, \varepsilon_L(q))} \ , \quad (15)$$

where $\Gamma_1(q, \varepsilon_L(q))$ is the real part of the polarization bubble at $\omega = \pm\varepsilon_L(q)$. In particular, at the antiferromagnetic point, we can define a longitudinal gap $\varepsilon_L(0) = \Delta_L$ and the quantity $\Gamma_1(0, \Delta_L)$ may be written as

$$\Gamma_1(0, \Delta_L) = \frac{3}{4} g \int_{2\Delta_T}^{+\infty} \frac{\mathrm{d}\omega}{\pi} \frac{1}{\sqrt{(\omega^2 - 4\Delta_T^2)}} \frac{1}{\omega^2 - \Delta_L^2} \cdot \quad (16)$$

For the field strength that we have estimated for $CsNiCl_3$, we find the value $\Delta_L = 0.779$, which is greater than Δ_T and consistent with polarized-neutron observations for weak staggered fields [20]. It is important to stress that the presence of the continuum invalidates, as already stated previously [16,19], the applicability of the SMA, that establishes the relation $\chi_L = Sgc/\Delta_L^2$ between the longitudinal susceptibility $\chi_L = \mathrm{d}m_s/\mathrm{d}H_s$ and the gap.

For $|\omega| < 2\Delta_T$ the dynamical structure factor in the longitudinal channel has well-defined poles corresponding to single particle excitations

$$S_L(q, \omega) = \gamma \frac{gcS^2}{2\varepsilon_L(q)} \{ \delta(\omega - \varepsilon_L(q)) - \delta(\omega + \varepsilon_L(q)) \} \quad (17)$$

where the prefactor γ is less than unity for $H_s \neq 0$ and gives the reduction of the quasi-particle weight. Since we are going to consider the antiferromagnetic point $q = 0$, here the longitudinal quasi-particle pole $\varepsilon_L(q)$, given by equation (15), is set equal to Δ_L. It turns out [19] that γ at $q = 0$ is given by the formula

$$\gamma = \left\{ \left[1 + \frac{(m_s/S)^2}{\Gamma_1^2(\omega^2)} \frac{\mathrm{d}\Gamma_1(\omega^2)}{\mathrm{d}\omega^2} \right]_{\omega = \Delta_L} \right\}^{-1} \quad (18)$$

which shows a rapid decrease as a function of H_s, starting from unity at $H_s = 0$ and saturating to a finite value ($\gamma = 0.279$) for high fields. In equation (18) we have written $\Gamma_1(\omega^2)$ in place of $\Gamma_1(0, \omega)$ to emphasize its explicit dependence on ω^2.

As the field increases, the spectral weight that is lost from the pole gets transferred to the multi-particle continuum, which starts at the two-particle threshold $\omega = 2\Delta_T(H_s)$, which is consistent with the first moment sum rule [19] for the dynamical structure factor. We can observe this phenomenon by calculating the continuum contribution to the dynamical structure factor for some values of the staggered field. In Figure 1 the quantity $S(q, \omega)$ is

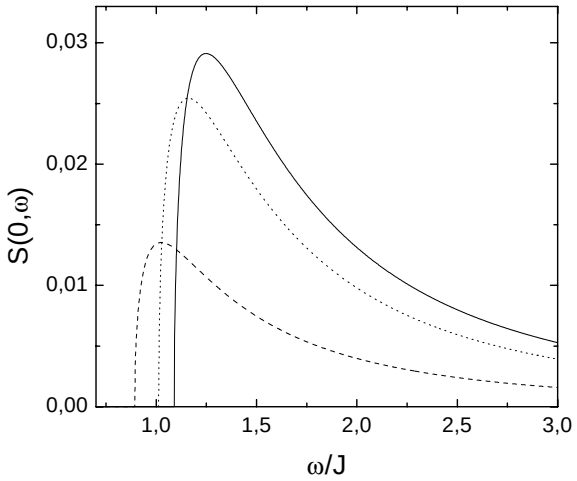

Fig. 1. Continuum contribution to the spin dynamical structure factor at the antiferromagnetic point $q = 0$, for $H_s = 0.010$ (dashed line), $H_s = 0.020$ (dotted line) and $H_s = 0.027$ (solid line). As the staggered field increases, the integrated intensity grows and the two-magnon threshold $2\Delta_T(H_s)$ shifts forward.

plotted as a function of ω at the antiferromagnetic point $q = 0$, for $H_s = 0.027$, $H_s = 0.020$ and $H_s = 0.010$. The integrated intensity grows as the staggered field increases. We also notice that this contribution starts always at the two-magnon threshold $\omega = 2\Delta_T(H_s)$, consistently with experimental data [7,20] in which the continuum starts well before the three magnon contribution as predicted by the NLσM for a single chain in the absence of an external staggered field [6]. The plots of $S(0,\omega)$ have been obtained calculating the imaginary part of the longitudinal propagator for $|\omega| > 2\Delta_T(H_s)$, explicitly [19]

$$S(0,\omega) = \frac{g\,c\,m^2\,\Gamma_2(\omega)}{(\Delta_T^2-\omega^2)^2(\Gamma_2(\omega))^2 + (m/S + (\Delta_T^2-\omega^2)\Gamma_1(\omega))^2} \quad (19)$$

where $\Gamma_2(\omega) = g^2c/(2\omega\sqrt{\omega^2-4\Delta_T^2})$ is the imaginary part of the polarization bubble at $q = 0$. Consistently with the main assumption of this paper, just above T_N the O(3) symmetry is restored by a slowly-varying staggered field, so the splitting of the structure factor in two channels must be understood in an adiabatic way, justified by the fact that the single chains involve fast processes as compared with those taking place in the background right above T_N.

At $H_s = 0.027$ the value of the prefactor is $\gamma = 0.746$. Hence, using the first moment sum rule we find that the relative weight of the incoherent contribution to the structure factor is

$$\frac{1-\gamma}{3} \approx 8.5\% \quad (20)$$

which is consistent with the experimental value of 9(2)%. In equation (20) the denominator 3 comes from the equiprobability for an unpolarized neutron to create an excitation in the longitudinal channel or in the two transversal ones.

The present theory predicts the existence of a longitudinal mode as well as of two (degenerate) transverse modes with different energies. This fact would be consistent with the presence of two peaks in the inelastic scattering measurements with unpolarized neutrons. However, in the experiments only one peak is observed. Remembering that a similar problem arises in the case of R_2BaNiO_5 [20], we argue that the longitudinal mode could be masked by its proximity to the transversal one, by its reduced weight (less than 1/2 of the transverse weight) and by the wide broadening. We recall also that the persistence of a longitudinal mode below the ordering temperature T_N has been predicted in reference [21] again for the $CsNiCl_3$ compound, in the framework of a Ginzburg-Landau model.

In conclusion, we have proposed a theoretical scenario that is able to explain both qualitatively and quantitatively the gap increase and the anomalous multiparticle excitation weight observed in recent neutron scattering experiments on $CsNiCl_3$. This latter quantity turns out to be much greater than the theoretical prediction for isolated $S = 1$ chains. The results of the present work seem to show that there is no need of invoking additional spin-1/2 excitations, as suggested by some authors [7]. We assume the existence of a temperature range above T_N, where the 3D effects are not at all negligible, although the overall 3D long-range order is destroyed, and we take into account the interaction with the background by modeling it in terms of an effective staggered magnetic field. We find that, even in the framework of a simple mean-field theory, this scenario may account for the generation of quite a sizable continuum weight, in good agreement with the experimental results, as well as for an upward gap renormalization and a reduction of the correlation length, breaking also the SMA correspondence between these two quantities. Finally, we have to mention that a more complete theory of $CsNiCl_3$ should take into account the effects of frustration caused by the triangular arrangement of the 1D chains, and that they might contribute to the enhancement of the gap for $T > T_N$. However, we believe that the sizable continuum starting from two-magnon contributions that the experimentalists observe is clearly described by the picture outlined in the present paper, and it seems to us hard to think that frustration could lead to similar results. In our attempt to interpret the apparently anomalous experimental data on $CsNiCl_3$ in the temperature range $T_N < T < T^*$ ($T^* \simeq 12$ K) we have tried to stress that their origin can be traced to the fact that there are actually, in this temperature range, two competing length and time scales, namely those related to the 3D order, that is destroyed above T_N, and that play a relevant role at large distances (small momenta) and times (low frequencies) as compared with the length and time scales proper of the single 1D chains, that are set by the 1D correlation length and by the inverse of the Haldane gap, and which are the scales at which the interesting physics of $CsNiCl_3$ manifests itself. That is why we believe that, from the Néel temperature and up to around the cross-over temperature T^*, despite the fact that the overall O(3) symmetry has been restored, the single chains

still experience, on their characteristic scales, an effective staggered field. Stressing once again that a more complete theory is needed, the fact that the predictions of this simple model fit quite well the experimental data appears to lend support to the model we are presenting here, and that it should be the starting point for future investigations.

We would like to thank A.A. Nersesyan, P. Pieri and G. Sierra for useful discussions.

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