Continuum in the Excitation Spectrum of the $S=1$ Compound CsNiCl$_3$

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Recent neutron scattering experiments on CsNiCl$_3$ reveal some features which are not well described by the nonlinear $\sigma$ model nor by numerical simulations on isolated S=1 spin chains. In particular, in real systems the intensity of the continuum of multiparticle excitations, at $T = 6$ K, is about 5 times greater than predicted. Also the gap is slightly higher and the correlation length is smaller. We propose a theoretical scenario where the interchain interaction is approximated by a staggered magnetic field, yielding to a correct prediction of the observed quantities.

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About twenty years after the famous “Haldane conjecture”[2], quantum spin chains with Heisenberg antiferromagnetic interactions are still attracting much experimental and theoretical interest. Half-integer spin ferromagnetic interactions are still attracting much experimental and theoretical interest. In particular, in real systems the intensity of the continuum of multiparticle excitations, at $T = 6$ K, is about 5 times greater than predicted. Also the gap is slightly higher and the correlation length is smaller. We propose a theoretical scenario where the interchain interaction is approximated by a staggered magnetic field, yielding to a correct prediction of the observed quantities.

For $S=1$ chains, the excitation spectrum is dominated by a well-defined magnon triplet carrying spin 1. These excitations have been observed in several neutron scattering experiments and in the case of NENP, they constitute a large part of the total spectrum.

However, recent experimental measurements on CsNiCl$_3$ 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[1]. This result is considerably larger than the 1-3% weight predicted by numerical simulations[3] and the O(3) non-linear sigma model for the 3-particle continuum[4]. The effects of the coupling between chains has been considered in the framework of RPA, but no significative increase of the continuum has been found[5]. On the other hand, Majorana fermion theory yields a prominent three-particle-scattering continuum[6], but this would require strong biquadratic exchange interactions of the form $(S_i \cdot S_{i+1})^2$ which are not in accordance with the measurements on CsNiCl$_3$[2].

Furthermore, some measured quantities at $T = 6$ K show appreciable deviations from analytical and numerical predictions at finite temperature on isolated chains[1][2], i.e. the measured gap $\Delta$ is higher and the correlation length $\xi$ is shorter than the corresponding theoretical values, breaking also the single mode approximation (SMA) correspondence $\Delta \xi = c$, where $c$ is the spin-wave velocity.

The model Hamiltonian proposed for CsNiCl$_3$ is

$$H = J \sum_{i=1}^{\text{chain}} S_i \cdot S_{i+1} + J' \sum_{\langle ij \rangle} S_i \cdot S_j + D \sum_{i} (S_i^z)^2,$$

where the coupling $J = 2.28$ meV along the c-axis is much stronger than the exchange interaction in the basal plane $J' = 0.044$ meV[1]. The single-ion anisotropy is estimated to be $D \approx 4 \mu$eV, small enough that CsNiCl$_3$ may be considered purely isotropic, hence we put $D = 0$ from now on. Below $T_N = 4.85$ K the system undergoes 3D long-range ordering, caused by the interchain interaction. For $T > T_N$ 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.

Here comes our main assumption:

In some range of intermediate temperatures above the ordering one, $T_N < T < T^*$, the interchain coupling induces a (short-range) three-dimensional antiferromagnetic order which varies on a length scale $\xi_{3D}$ which is greater than the correlation length $\xi_{1D}$ of the single chain.

In other words, a little above $T_N$, even if the total staggered magnetization is zero, three-dimensional effects are still important and the system is arranged in large domains with non-zero magnetization. Thus, in a mean-field picture, a single chain experiences a staggered magnetic field generated by the neighboring chains. No matter if the field is not constant along the whole chain, because it varies so slowly that it may be considered constant, since the 1D chains are gapped and short-ranged.

Consistently, we suppose the existence of a temperature $T^*$ above which the antiferromagnetic order is so reduced that the 1D system may be viewed as isolated. To support this hypothesis we mention that in the real compound CsNiCl$_3$, some experimentalists[6] found that for $T \gtrsim 12$ K the measurements turn out to be in accordance with the theoretical predictions on isolated chains.

In the mean-field approximation the interchain interaction is treated as a staggered magnetic field that takes
into account the effect of the neighboring chains on the single 1D system

\[ J' \sum_{\langle ij \rangle} S_i \cdot S_j \quad \Rightarrow \quad H_s \sum_i (-)^i S_i^z , \]  

(2)

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

\[ H_s = z_c J' m_s(H_s) , \]  

(3)

with \( z_c \) 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

\[ H = \sum_i \left[ J S_i \cdot S_{i+1} + (-)^i H_s S_i^z \right] . \]  

(4)

Following the Haldane mapping, we represent the local spin operators as \( S_i = S(-)^i n_i + i \), with \( n_i^2 = 1 \), where \( n_i \) represents the slowly-varying local staggered magnetization and \( i \) is the local generator of angular momentum. In this framework, the Zeeman term of Eq.(4) becomes essentially a linear shift in the \( n \)-field along the \( z \)-direction, \((-)^i H_s S_i^z \approx S H_s n_i^z \).

Going then to the continuum limit and integrating out the fluctuation field \( i \) we obtain the O(3) NL\( \sigma \)M plus a linear term

\[ \mathcal{L} = -\frac{1}{2gc} \left( c^2 |\partial_x n|^2 + |\partial_x n|^2 \right) - S H_s n^z - i\lambda \left( n^2 - 1 \right) , \]  

(5)

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

The experiments were performed at temperatures small enough if compared with the exchange interaction \( J \) along the chain, so we mainly consider the zero-temperature case arguing that a calculation at finite temperature 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 \( \lambda \) and the self-consistency equation at \( T = 0 \) is

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

(6)

where \( \xi \) is the correlation length and \( \Lambda \) 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 \( c = \xi \Delta_0 = 2.48 J \)), known from the DMRG studies. The NL\( \sigma \)M coupling has been left to its analytical value \( g = 2 \). As the field is varied, the cutoff \( \Lambda \) is held fixed to its zero-field value \( \Lambda = 0.2072 \) and we use Eq.(6) to determine the value of \( \xi = \xi(H_s) \).

The magnetization per site is given by

\[ m_s(H_s) = S \langle n^z \rangle = \frac{g S^2}{c} \xi^2(H_s) H_s , \]  

(7)

Putting it back in Eq.(3), we find a stable solution for \( H_s = 0.027 \), corresponding to \( m_s = 0.45, \xi = 4.54 \) and \( \Delta_T = \xi^{-1} = 0.545 \). 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 symmetry and the quasi-particle propagator splits in one longitudinal channel (i.e. in the direction of the field) and two transverse channels. The transverse propagator is given by

\[ G_T(q, \omega) = \frac{S^2 gc}{\omega^2 + c^2 q^2 + \Delta_T^2} , \]  

(8)

and has simple poles at \( \omega = \pm \varepsilon(q) \) with

\[ \varepsilon(q) = \sqrt{c^2 q^2 + \Delta_T^2} , \]  

(9)

where \( q = 0 \) 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, the dynamical structure factor \( S_T(q, \omega) \equiv \text{Im} G_T(q, \omega) / \pi \) is simply given by

\[ S_T(q, \omega) = \frac{g c S^2}{2 \varepsilon(q)} \left\{ \delta(\omega - \varepsilon(q)) - \delta(\omega + \varepsilon(q)) \right\} \]  

(10)

On the other hand, the longitudinal propagator must be evaluated more carefully considering its connected part and the calculation\[1\] gives

\[ G_L(q, \omega) = G_T(q, \omega) \frac{3 \tilde{\Gamma}(q, \omega)}{3 \tilde{\Gamma}(q, \omega) + 2m_s^2 G_T(q, \omega)} , \]  

(11)

where \( \tilde{\Gamma}(q, \omega) \) is the Fourier transform of the product of
two propagators
\[ \Gamma(x - x') = \frac{1}{S^4} G_T(x - x') G_T(x' - x). \]

From the study of the analytic structure of the longitudinal propagator it is possible to calculate the longitudinal gap solving the following equation for \( \Delta_L \):
\[ \Delta_L^2 = \Delta_T^2 + \frac{(m_s/S)^2}{\Gamma_1(0, \Delta_L)}, \]
where \( \Gamma_1(0, \Delta_L) \) is the real part of the "polarization bubble" (in \( q = 0 \) and \( \omega = \Delta_L \)) and may be written as
\[ \Gamma_1(0, \Delta_L) = \frac{3}{4} g \int_{2\Delta_T}^{+\infty} \frac{d\omega}{\pi} \frac{1}{\sqrt{(\omega^2 - 4\Delta_T^2)}} \frac{1}{\omega - \Delta_L^2}. \]

For the field strength that we have estimated for CsNiCl\(_3\), we find the value \( \Delta_L = 0.779 \), which is greater than \( \Delta_T \), as we expected. It is important to stress that the presence of the continuum makes inapplicable the SMA, which establishes the relation \( \chi_L = S g c / \Delta_L^2 \) between the longitudinal susceptibility \( \chi_L = dm_s / dH_s \) 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
\[ S_L(q, \omega) = \gamma \frac{g c s^2}{2\varepsilon_L(q)} \{ \delta(\omega - \varepsilon_L(q)) - \delta(\omega + \varepsilon_L(q)) \} \]
where the prefactor \( \gamma \) is less than unity and gives the reduction of the quasi-particle weight. From the calculation \( \[11\] \), it turns out that \( \gamma \) is a decreasing function of the staggered field, as displayed in Fig.1.

![FIG. 2: Continuum contribution to the spin dynamical structure factor at the antiferromagnetic point, for \( H_s = 0.027 \) (solid line), \( H_s = 0.020 \) (dotted line) and \( H_s = 0.010 \) (dashed line). The integrated intensity grows as the staggered field increases.](image)

As the field increases, the spectral weight that is lost from the pole gets transferred to the multi-particle continuum, as required by the first moment sum rule \( [11] \) of 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 Fig 2 the quantity \( S(q, \omega) \) is plotted as a function of \( \omega \) at the antiferromagnetic point, for \( H_s = 0.027, H_s = 0.020 \) and \( H_s = 0.010 \) (dashed line). The integrated intensity grows as the staggered field increases. We also notice that this contribution starts at the two-magnon threshold \( \omega > 2\Delta_T(H_s) \), consistently with experimental data \( [12, 13] \) in which the continuum starts well before the three magnon contribution as predicted by the NLσM for a single chain \( [11] \).

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
\[ \frac{1 - \gamma}{3} \approx 8.5\% \]
which is a considerable amount, very close to the experimental measure of \( 9(2)\% \).

The present theory predicts the existence of a transversal mode and a longitudinal one with different energies. This fact would be consistent with the presence of two well-defined peaks in the inelastic scattering measurements with unpolarized neutrons. However, in the experiments only one peak is observed. Remembering that a similar problem arise in the case of \( R_2\text{BaNiO}_5 \) \( [12] \), we argue that the longitudinal mode could be masked by its proximity to the transversal one, by its reduced weight.

![FIG. 1: The relative quasi-particle weight of the pole of the longitudinal propagator as a function of the staggered field, at \( q = 0 \).](image)
(less than 1/2 of the transverse weight) and by the wide broadening.

In conclusion, we have proposed a theoretical mechanism to explicate the anomalous multiparticle excitation weight observed in recent neutron scattering experiments. This quantity turns out to be much greater than the theoretical prediction on isolated $S = 1$ chains. We assume the existence of a temperature region above $T_N$, where the 3D effects are not at all negligible, although the long range order is destroyed, and we take into account of the interaction with the neighboring chains by means of a staggered magnetic field. We found that, even in the framework of a simple mean-field theory, this scenario may account for the generation of a sizable continuum weight, as well as an upward gap renormalization and a reduction of the correlation length, breaking also the SMA correspondence between these two quantities. Further and more detailed analytical calculation and numerical tests will be treated in a future paper.

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