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NEUTRON SCATTERING EXPERIMENTS ON THE HALDANE CONJECTURE

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Abstract. – Polarized neutron study of the spin dynamics for a Heisenberg antiferromagnet with easy-axis anisotropy, CsNiCl₃, clearly demonstrates the almost isotropic character of the system. The study also revealed a difference between the observed and the calculated spin wave dispersion using the classical spin wave theory for $T < 4.8$ K and $T > 4.8$ K. This may be interpreted as a quantum effect on the spin dynamics in one and three dimension, as has been suggested by Haldane for one dimension.

Introduction

The spin wave (SW) dispersion in CsNiCl₃, being a quasi one-dimensional (1-D) near Heisenberg antiferromagnetic (AF) system, has attracted attention because of its possible relevance to the Haldane conjecture [1]. CsNiCl₃ forms antiferromagnetic chains along the hexagonal $c$-axis, whose magnetic properties can be described by

$$H = -J \sum_i S_i S_{i+1} - A \sum_i (S^z_i)^2 - J' \sum_{i,j} S_i S_j$$

with $S = 1$, $J$ and $J'$ being the intra- and interchain exchange parameter, respectively, and $A$ the single site anisotropy. The systems undergoes two successive magnetic phase transitions at $T_{N1} = 4.8$ K and $T_{N2} = 4.3$ K [2]. Buyers et al. [3] deduced the microscopic parameters $J / k_B = -16.61$ K, $J' / k_B = -0.29$ K and $A / k_B = 0.63$ K from the SW dispersion in the 3-D ordered phase ($T < T_{N2}$). They applied a six sublattice model assuming a nearly 120° – AF ordered state in the $ac$-plane proposed by Yelon and Cox [2]. The observed gap in 1-D phase ($T > T_{N1}$) then could not be explained by this easy axis anisotropy $A$. The additional energy beyond the expected energy gap due to the anisotropy was then attributed to the existence of the Haldane gap in the 1-D, $S = 1$ AF Heisenberg system. One should note that this anisotropy value does not agree with the value derived from other experimental findings. Firstly the anisotropy energy estimated from the measured spin flop field [4] is much smaller than the one obtained by Buyers et al. Secondly the canting angle $\theta$ in the 3-D ordered state, which is the angle between the $c$-axis and the spins not pointing along this easy $c$-axis, deduced from the neutron diffraction study [2] is 59°. Using the parameters derived from the SW dispersion would mean a canting angle $\theta = 54°$, a much bigger deviation from the isotropic 60° case than observed.

To solve this discrepancy which might be essential to the interpretation of the observed energy gap at (001) in the 1-D phase by the Haldane conjecture, we performed polarized neutron inelastic scattering experiments to determine the eigenvector of the different SW branches as a much more detailed test for the theoretical predictions.

Experimental

We performed polarized neutron experiments on the IN20 3-axis intrument at ILL equipped with Heusler monochromator and analyser. Energy scans in constant $Q$-mode with fixed $E_I = 5.4$ and $14.7$ meV were performed. A PG filter was installed after the sample to suppress the second order contamination. The single crystal sample CsNiCl₃ of a size $\sim 2$ cm$^3$ was oriented with its [110] and [001] axis in the scattering plane. For experiments in the 3-D ordered phase the sample was mounted in a vertical field cryomagnet. To obtain a single domain state we applied a field of 8 kG.

For experiments in the 1-D phase the sample was mounted with the same orientation in a flow-cryostat in a Helmholtz configuration, where the field direction of the sample could be chosen arbitrarily by setting the currents in the different Helmholtz coils.

Results and discussions

In figure 1 the measured dispersion in [110] direction with the observability of the different branches in spin flip (SF) and non spin flip (NSF) is shown. The dispersion in the chain direction measured by the unpolarized neutron scattering experiment [5] is also included. Disregarding the polarization of the SW the observed SW peak positions are the same as observed by Buyers et al. [2]. They associated to the flat branch in [110]-[110].

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direction around 0.2 THz the second lowest \( \langle S^zS^z \rangle \) mode which then resulted in the rather large value of \( A/k_B = 0.63 \) K. A \( \langle S^zS^z \rangle \) mode would have shown up in our polarized experiment in the NSF channel. The experiment however unambiguously showed that the mode at 0.2 THz shows up in the SF channel.

We performed a SW calculation using the six sublattice model. The equation of motion derived from the Hamiltonian [1] is solved by a numerical diagonalisation and the SW energies are obtained from the eigenvalues. By adjusting the next higher, flat \( x^2 - y^2 \) branch, which would show up in the SF channel, to the observed flat SF branch at 0.2 THz, we obtain \( J'/k_B = -0.10 \) K and \( A/k_B = -0.035 \) K when we keep \( J/k_B = -16.6 \) K (deduced from the zone boundary energy in the chain direction). The calculated SW dispersion with these parameters are indicated by the lines in figure 1. Only the branches also observed by the neutron scattering experiment are shown. This \( J' \) and \( D \) yield the canting angle \( \theta = 59^\circ \) in accordance with the neutron diffraction experiment [2]. The \( A \) value agrees also well with the value derived from the spin flop field of 2 Tesla [5]. Thus by adjusting the SW branch of correct polarization to the observed energy the above mentioned discrepancy is resolved. Recently the successive phase transitions in CsNiCl\(_3\) were studied carefully by NMR [6]. Their interpretation of the intermediate phase requires a quasi-degeneracy in the canting angle \( \theta \), which is only possible if \( |A/J'| \ll 1 \) in accordance with the parameters derived here.

To demonstrate that CsNiCl\(_3\) is really a nearly isotropic case we measured the different correlation functions for \( T > T_{N1} \). In figure 2 we show an example of the polarization analysis results at the 1-D zone center along \((\zeta, \zeta, 1)\). Applying a small guide field vertical to \( Q \) we observe \( \sin^2 \alpha \langle S^zS^z \rangle + \cos^2 \alpha \langle S^yS^y \rangle \), \( \alpha \) being the angle between \( c^* \) and \( Q \), in SF and \( \langle S^yS^y \rangle \) in NSF channel. By applying the guide field along \( Q \) and looking at the NSF channel we obtain the background. Since \( \langle S^yS^y \rangle = \langle S^zS^z \rangle \) in the easy-axis 1-D phase it can be directly compared with the \( \langle S^zS^z \rangle \). One concludes from the measurement \( \langle S^zS^z \rangle \) and \( \langle S^yS^y \rangle \) to be identical within the errorbar of 8 %.

All experimental evidence indicate that the axial anisotropy is very small indeed, and that the energy gap at \( (001) \) in the 1-D phase, which would require \( A/k_B = 1.78 \) K, cannot be of single site origine. To our surprise the energy gap at \( (001) \) is too large not only in 1-D phase, but even in 3-D ordered phase, if one takes the polarization of the SW-branches and the above mentioned experimental facts into account. Two reasons for this surprising observation can be thought of. Either the assumed Hamiltonian is missing some important terms or the quantum effect also modifies the dynamics in the 3-D ordered state. The first possibility seems to be very unlikely because even the complicated successive transitions can be explained using this Hamiltonian [6]. Considering the second possibility one should remember that the classical SW calculation requires a well defined ground state, as have been assumed here with the six sublattice model. Yelon and Cox [2] already noted that the observed small ordered moment at low temperature is probably due to quantum fluctuations. Mattis [7] has also pointed out that the Haldane gap just scales with dimensionality and the conjecture therefore may be well valid for all dimensions. Whether this observation can be interpreted by the Haldane conjecture [1] or the dynamical dimerization [8] requires more extended studies in other related systems.