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Spin dynamics along the chain direction in an $S = \frac{3}{2}$, quasi-one-dimensional Heisenberg antiferromagnet $CsVCl_3$

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Abstract. Inelastic neutron scattering experiments investigating the spin dynamics along the chain direction of an $S = \frac{3}{2}$ quasi-one-dimensional Heisenberg antiferromagnet $CsVCl_3$ were performed up to the zone boundary energy. We observed the quantum renormalization in the exchange constant and found that the scattering function cannot be described by either the classical δ-function or the quantum Heisenberg model for $S = \frac{1}{2}$. The spin excitations have apparent linewidths and they are compared with the theory of the classical isotropic Heisenberg system at finite temperatures.

1. Introduction

The spin dynamics in one-dimensional (1D) Heisenberg antiferromagnets (HAFs) is one of the most attractive problems in physics and many theoreticians and experimentalists have been tackling this problem. Quantum theories for 1D HAF systems with $S = \frac{1}{2}$ have been developed intensively and many important features have been predicted; the classical Néel state is not the ground state [1], there is a quantum renormalization such that the excitation energy is $\frac{1}{2}\pi$ times as large as that of the classical spin wave [2] and the spin-wave dispersion curve is the lower boundary of a triplet spin-wave double continuum [3]. These features have been confirmed in real materials [4, 5]. Haldane predicted the spin-value-dependent dynamics in 1D HAF systems; half-integer-spin systems show gapless excitations and power-law decay in the spin correlation and integer-spin systems show an excitation gap and exponentially decaying correlation [6]. Since he presented this conjecture, the dynamical properties in $S = 1$ systems have been intensively investigated both theoretically and experimentally [7]. On the other hand it is well established that $S = \frac{5}{2}$ systems behave classically [8]. We are interested especially in the crossover behaviour in the spin dynamics from the quantum to the classical limit [7]. $S = \frac{3}{2}$ systems are expected to be suitable for such investigations.

$CsVCl_3$ is a compound recognized as an excellent realization of a 1D HAF system with $S = \frac{3}{2}$ above the three-dimensional ordering temperature $T_N = 13.3$ K [9]. In the inelastic neutron measurements using a triple-axis spectrometer installed at a steady state neutron

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source, the spin-wave dispersion relation along the chain up to 20 meV, which corresponds to only 20% of the Brillouin zone, was measured [9, 10]. From these results the intrachain exchange constant was estimated to be $J = -169$ K applying the classical interpretation [9]. On the other hand, J was estimated to be -115 K from bulk susceptibility measurements [11]. This discrepancy is interpreted as a quantum renormalization similar to the factor $\frac{1}{2}\pi$ in the spin-wave excitation energy expected in $S = \frac{1}{2}$ systems [9].

In this paper we report inelastic neutron scattering experiments of the spin-wave excitations along the chain direction of a $CsVCl_3$ single-crystal sample up to the zone boundary energy using a chopper spectrometer installed at a pulsed neutron source.

2. Experiment

Inelastic neutron scattering experiments were performed on the MARI spectrometer installed at the spallation neutron source (ISIS Facility) at the Rutherford Appleton Laboratory [12]. The MARI spectrometer is a direct-geometry chopper spectrometer coupled with the pulsed neutron source. Pulsed polychromatic beams generated at the source are monochromated by a mechanical chopper synchronized with the repetition of neutron pulses, where the chopper is placed between the source and the sample. Incident beam energies E_i can be selected in the wide range from the thermal to the epithermal neutron regime. Scattered neutrons are detected at a detector system covering a wide range of scattering angles from 3° to 135°. The energy transfer and the momentum transfer are determined from the time of flight of the detected neutrons and the nominal instrumental resolution of the energy transfer is about 1% of E_i.

Our sample consists of four single crystals of which the c^* axes were aligned in the same direction and the total volume of the sample was about 3 cm^3. The mosaicity of the sample including the misalignment among the single crystals was less than 2°. The c^* axis of the sample was mounted parallel to the incident beam direction. In this configuration the 1D Bragg plane is perpendicular to the incident wavevector k_i, and the 1D momentum transfer q, which is the momentum transfer along the chain direction and defined as a distance from the 1D Bragg plane, does not vary very much in a small-scattering-angle regime and the energy transfer is independent of the scattering angle. Therefore adding the intensities counted at the small scattering angle between 3° and 12° does not make the resolution of either energy transfer or 1D momentum transfer worse [5].

We performed four measurements with incident energies E_i of 100, 150, 200 and 300 meV at $T = 40$ K above T_N. A background measurement was performed by turning the sample into the position where the c^* axis is perpendicular to k_i. Because of the change in the sample geometry, the self-shielding effect of the sample had to be accounted for. By comparing this background with the intensity in the actual measurement at the large-scattering-angle detectors between 30° and 120° where there is no magnetic contribution due to the magnetic form factor, an empirical formula including self-shielding and multiple scattering was established to deduce the background from the signals at the large-scattering-angle detectors. The factor k_f/k_i which generally appears in the inelastic neutron cross section was also corrected in the spectrum.

3. Results and discussion

The excitation spectrum consisted of well-defined peaks. In order to determine the peak position, each peak was fitted to a Gaussian. Figure 1 shows all the observed excitation peak positions on the scan locus in the $q\omega$-space. The peak positions were well fitted to the classical spin-wave dispersion relation $\hbar\omega(q) = \epsilon_{ZB}|\sin(\pi q)|$, and the excitation energy ϵ_{ZB} at the zone boundary was obtained to be 86 ± 3 meV. In the classical model ϵ_{ZB} is equal to $4S|J|$, therefore the exchange constant was determined to be $J = -167 \pm 6$ K. On the other hand, J was estimated to be -115 K from the susceptibility measurements [11] applying the classical interpretation. This discrepancy suggests that the quantum effect might be significant even in a system with a larger spin value ($S = \frac{3}{2}$) than $\frac{1}{2}$. Our result supports the preliminary result presented from the measurement at a steady state neutron source [9].

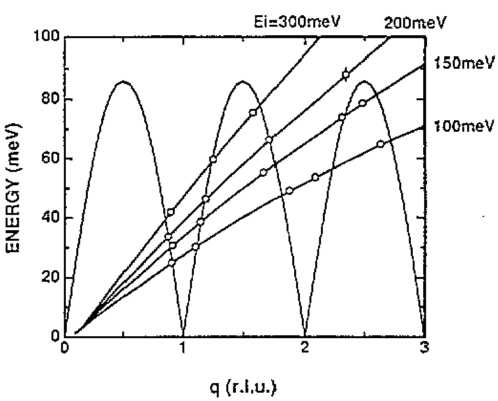

Figure 1. Dispersion relation of the spin-wave excitation. All the observed peaks on the scan locus of $E_i = 100$, 150, 200 and 300 meV are indicated. The sinusoidal curve is the fitted dispersion relation.

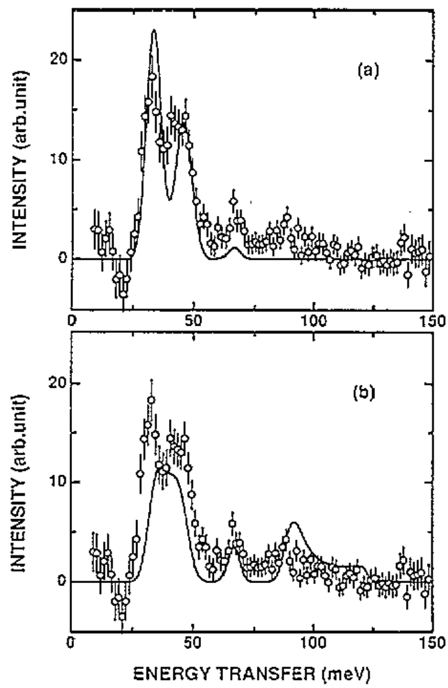

Figure 2. Background-corrected inelastic spectrum with $E_i = 200$ meV and comparison with model calculations: (a) the classical δ-function; (b) the quantum Heisenberg model for $S = \frac{1}{2}$.

The classical spin-wave theory for a 1D HAF where the spins are interacting with their nearest neighbours yields the following scattering function (see, e.g., [13]):

$$S(q, \omega) = [n(\omega) + 1]F(Q)^2|\tan(\tfrac{1}{2}\pi q)|\delta[\hbar\omega - \hbar\omega(q)] \qquad (1)$$

where $F(Q)$ is the magnetic form factor [14] and Q is the scattering vector. The temperature factor $n(\omega) = [\exp(\hbar\omega/k_BT) - 1]^{-1}$ is negligible in the energy region where excitation peaks were observed. $\hbar\omega(q)$ is the dispersion relation determined above. We performed the convolution of the scattering function in equation (1) with the resolution function [15] calculated by the experimental geometry and compared the convoluted scattering function with the experimental spectra. The solid curve in figure 2(a) is the convoluted scattering function with $\epsilon_{ZB} = 86$ meV. The calculated curve was adjusted to the experimental spectrum by parametrizing only the constant proportional to the intensity. In the spectrum with $E_i = 200$ meV the positions of the lowest double peak and the next peak in the experimental data are expected by the calculation, however, the peak at around $\hbar\omega = 90$ meV is not expected and all the spectra have apparent widths. We also calculated intensities for the quantum model for 1D HAF systems with $S = \frac{1}{2}$ [3] as shown in figure 2(*b*). The calculation curve was adjusted in the same way as in the classical calculation. The shape of the lowest double peak is quite different, that is there is no valley in the calculation; moreover a large continuum appears at a higher energy transfer in the calculation. The excitation spectrum cannot be expressed by either the classical δ-function or the quantum theory.

We therefore introduced finite linewidths and assumed the scattering function where the δ-function in equation (1) is replaced by the Lorentzian

$$(\Gamma/\pi)/\{[\hbar\omega - \hbar\omega(q)]^2 + \Gamma^2\}. \tag{2}$$

It is difficult to determine the q-dependence of Γ in this experimental geometry because the value of q varies on the scan locus. First, therefore, we assumed Γ to be independent of q and tried to fit the data to the resolution-convoluted Lorentzian scattering function by parametrizing ϵ_{ZB}, Γ and the constant proportional to the intensity. Although each spectrum was fitted to the Lorentzian better than either the classical δ-function or the quantum model, unique values of ϵ_{ZB} and Γ were not obtained for all the spectra with different incident energies. This suggests the q-dependence of Γ. Next, in order to determine the q-dependence, $\Gamma(q)$, the value of Γ was obtained for each excitation peak. For example, the spectrum with $E_i = 200$ meV was divided into four intervals around the peaks of $\hbar\omega = 24$–38, 38–60, 60–75 and 75–110 meV and each interval was fitted to the above-mentioned scattering function with Γ independent of q. During the fitting, the value of ϵ_{ZB} was kept at a constant value of 86 meV determined from the dispersion relation. Figure 3 shows the $\Gamma(q)$ obtained by this procedure.

Each interval is affected by the nearest peaks. Around $q = 1$ rlu (reciprocal lattice unit) the linewidths of 2–7 meV were obtained by the fitting in two intervals into which the first double peak was divided and each interval was well fitted. At around $q = 2$ rlu the widths were obtained by the fitting of the peaks in the higher-energy transfer. Although the spectrum at around $q = 2$ rlu was well fitted, in the fitting the constant proportional to the intensity was obtained to be much larger than that around $q = 1$ rlu. The above feature of the fitting is common to all the spectra with different incident energies.

We compared the experimental result in figure 3 with the existing classical theory on spin-wave damping. The full curve in figure 3 is the $\Gamma(q)$ in the theory for an isotropic 1D HAF system [16], which was calculated with the parameters $S = \frac{3}{2}$, $\epsilon_{ZB} = 86$ meV and $T = 40$ K. If the present system is regarded as a classical system, this theory can be applied to the present system because the single-ion anisotropy (0.52 K [10]) is much smaller than the thermal energy ($T = 40$ K) [17]. The features of the q-dependent increase and decrease in Γ in this theory seem to agree qualitatively with the experimental results in figure 3. However, the observed linewidths are systematically larger than those predicted by the classical theory for an isotropic 1D HAF system at $T = 40$ K. One should mention that for $CsNiCl_3$ (an $S = 1$, nearly isotropic 1D HAF system) only resolution-limited excitations have been observed in the chain direction at $T = 7$ K [18]. $(T - T_N)k_B/J$ in that case is 0.17 and it is the same as the value of 0.16 at $T = 40$ K for $CsVCl_3$. Whether the larger disagreement around $q = 2$ rlu is a consequence of the q-dependence of the linewidths or

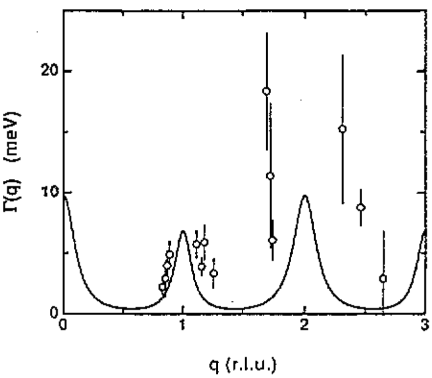

Figure 3. $\Gamma(q)$, the q-dependence of the linewidths of the scattering function determined from the excitation spectra: ——, curve calculated by the classical theory for an isotropic Heisenberg system. The parameters $S = \frac{3}{2}$, $\epsilon_{ZB} = 86$ meV and $T = 40$ K were used in the calculation.

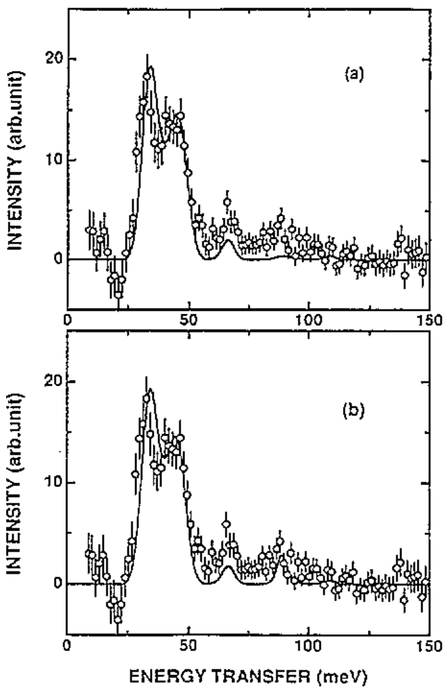

Figure 4. The inelastic spectrum with $E_i = 200$ meV and the classical theory in which $\Gamma(q)$ is described by the full curve in figure 3. The scattering function for (a) $\epsilon_{ZB} = 86$ meV and (b) $\epsilon_{ZB} = 89$ meV were calculated.

is due to the problem in the fitting procedure as mentioned above is not clear yet and will be explored in future experiments. Using $\Gamma(q)$ in this theory, we calculated the excitation spectrum as shown in figure 4(a). The calculated curve was adjusted by parametrizing only the constant proportional to the intensity. The first double peak was explained much better than that in either the classical δ-function or the quantum theory with $S = \frac{1}{2}$. However, the observed intensity in the higher-energy transfer is larger than the calculation. These features are common to all the spectra with different incident energies. We found that the peak at around 90 meV can be more emphasized if we chose $\epsilon_{ZB} = 89$ meV in this theory as shown in figure 4(b). The value of 89 meV is the upper limit of ϵ_{ZB} within the error in the present determination of the dispersion curve. The observed intensities of the peaks at higher energies are larger than the calculated values and the observed linewidths are obviously larger. It seems that the classical spin-wave damping is not enough to account fully for the experimental findings in this $S = \frac{3}{2}$ spin chain.

4. Summary

We have measured the spin-wave excitation propagating along the chain direction of an $S = \frac{3}{2}$ quasi-1D HAF $CsVCl_3$, up to the zone boundary by using a chopper spectrometer installed at a pulsed neutron source. We determined the dispersion relation of the spin-wave excitation and found that the exchange constant deduced from the excitation energy agreed well with the result from the earlier measurement at a steady state neutron source. We have therefore confirmed the quantum renormalization of the exchange constant determined by applying the classical interpretation. We found that the scattering function is not described by either the classical δ-function or the quantum theory and that the excitation spectra have apparent widths. The qualitative q-dependence of the linewidths was similar to that expected by a classical theory for an isotropic 1D HAF system at a finite temperature but the widths were systematically larger. We need to carry out experiments in another more sensitive geometry to study the linewidths of the excitations.

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