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Magnetic Correlations in $CsVBr_3$

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The static magnetic correlation function, $S(q)$, in an $S = 3/2$, one-dimensional Heisenberg antiferromagnet, $CsVBr_3$, was measured at temperatures between 40 K and 200 K above the three-dimensional ordering temperature ($T_N = 20.3$ K), on a chopper spectrometer install at a pulsed neutron source. The observed $S(q)$ was well fitted to a Lorentzian function convoluted with the instrumental resolution function. The inverse correlation length, $\kappa(T)$, determined as a half width of the correlation function, showed a linear temperature dependence in the observed temperature range, and, qualitatively well agreed with classical theory. This is consistent with current theory, also, with $\kappa(T)$ reported for a similar system, $CsVCl_3$.

KEYWORDS: one-dimensional Heisenberg antiferromagnet, spin dynamics, inelastic neutron scattering, pulsed neutron, chopper spectrometer

§1. Introduction

It is well known that the spin dynamics in one-dimensional (1D) Heisenberg antiferromagnets strongly depends on the value of spin (S). We have been studying the spin dynamics in S=3/2 systems, in order to investigate crossover behavior from a quantum state in a small S system to a classical state in a large S system.[1)] $CsVCl_3$ and $CsVBr_3$ are compounds recognized as excellent realizations of an S=3/2 system above the three-dimensional (3D) ordering temperature, $T_N = 13.3$ K and 20.3 K, respectively.[2–4)] We have investigated the spin dynamics in these compounds.[1)] In order to discuss one-dimensionality in these real systems, investigations of the dynamical properties at temperatures (T) higher than T_N is required, where the properties in the 3D ordered state should be negligible. First, we measured the dynamical structure factor, $S(q,\omega)$, over the entire Brillouin zone at low T (40 K for $CsVCl_3$ and 25 K for $CsVBr_3$) on the chopper spectrometers installed at pulsed neutron sources.[1)] From the observed dispersion relation, using the renormalizationconstant calculated by the quantum Monte Carlo method,[5)] we obtained the exchange constants, $J = 119 \pm 3$ K and 99 ± 3 K for $CsVCl_3$ and $CsVBr_3$, respectively, in good agreement with those deduced from bulk-susceptibility measurements.[2,4)] We found that the magnetic excitations at the magnetic zone center spread up to approximately 10 meV corresponding to the energy scale of J in both compounds. This energy spread is consistent with the excitation continuum predicted by the quantum Monte Carlo calculation for S=3/2.[6)] Next, we measured the T dependence of the energy width of magnetic excitations, Γ, for these compounds, and Γ was observed to be independent of the 1D momentum transfer (q) at $q > 0.07$ rlu (reciprocal lattice unit) at low T. We found that $\Gamma(T)$ at the q-range at $T > J$ is proportional to T as predicted by classical theory,[7)] but Γ becomes finite at low $T(< J)$. The finite Γ at low T indicates some fluctuations, which is consistent with the excitation continuum as mentioned above. Therefore, we concluded that an S=3/2 system exhibits the crossover from the quantum state at low T to the classical state at high T in a temperature range easily accessible by a usual experiment.[1)]

In the above discussions, the T dependence of the static correlation function, $S(q)$, is important. Classical theory has predicted that Γ is proportional to the inverse correlation length (κ) at low T.[7)] In order to determine the origin of the finite Γ observed at low T, we performed the measurement of $\kappa(T)$ in $CsVCl_3$ by using a crystal-analyzer spectrometer installed at a pulsed neutron source.[1)] Inelastic spectrum was taken with the constant-q scan, then we could deduce $S(q)$ by integrating $S(q,\omega)$ over the energy spread of all magnetic excitations by correcting such an energy-dependent prefactor in the inelastic neutron scattering cross section as the kinematical factor k_f/k_i, where k_i and k_f are the incident and the final wave vectors. A conventional method of the measurement of $S(q)$ can be realized with a double-axis mode on a triple axis spectrometer at a steady-state neutron source. Since in a double-axis scan the energy integration is performed without correcting the energy-dependent prefactor, the energy-integrated spectrum can be approximated to be $S(q)$ only if the energy spread of excitations is much smaller than the incident neutron energy. Therefore, an accurate $S(q)$ in a material having the larger energy range of excitations can be obtained only in the way as described above by using a spectrometer at an intense pulsed neutron source. The measured $S(q)$ in $CsVCl_3$ was well fitted to the Lorentzian function and $\kappa(T)$ determined as a half width of the correlation function was found to be proportional to T. The obtained result is consistent with current theory.[8,9)] Therefore, the observed finite energy width at low T should not be attributed to the scaling of $\Gamma(T)$ with $\kappa(T)$ in terms of classical theory, but rather indicates some fluctuations surviving even at low T. At present, we performed an inelastic neutron scattering experiment in order to determine $\kappa(T)$ in the other compound, $CsVBr_3$.

§2. Experiment

An inelastic neutron scattering experiment was performed on the chopper spectrometer, INC,[10] installed at the pulsed spallation neutron source of the Neutron Science Laboratory (KENS) at the High Energy Accelerator Research Organization (KEK). Pulsed polychromatic beams generated at the neutron source (moderator) are monochromatized by a mechanical chopper synchronized with the repetition of neutron pulses, and then come into the sample. The energy transfer is determined from the time-of-flight of the detected neutrons. In the present experiment, we used detectors located at scattering angles (ϕ) from 5.48° to 11.55°. We measured the inelastic scattering from a single-crystal sample of $CsVBr_3$ at temperature points between 40 K and 200 K. The mosaic spread was measured to be 1.7° at the full width at half maximum (FWHM). The sample was mounted with [001] and [110] in the scattering plane. In the scan geometry with ϕ and ψ (ψ is the crystal angle between k_i and [110]), q is given by

$$q = k_i \sin\psi + k_f \sin(\phi - \psi). \tag{2.1}$$

The crystal angle was chosen to be $\psi = 9.73°$ in order to realize the constant-q scan at the detector with $\phi = 9.73°$. The incident neutron energy was chosen to be $E_i = 80$ meV, in order to cover all magnetic excitations as well as to realize the constant-q scan at the magnetic zone center ($q = 1.05$ Å^{-1} at $T = 25$K) in the present ϕ-range. The observed inelastic spectrum can be expressed as $(k_f/k_i)[n(\omega)+1]S(q,\omega)$ adding a background, where $[n(\omega)+1]$is the temperature factor, $[\exp(-\omega/T)-1]^{-1}$, with the energy transfer ω. By correcting the ω dependent prefactor $(k_f/k_i)[n(\omega)+1]$, $S(q,\omega)$ was deduced, and then $S(q)$ was obtained by integrating $S(q,\omega)$ in the energy range from $\omega = 0$ to 75 meV. The upper boundary of the integration is over all excitations even at around the zone boundary at the present T-range.[1] The value of q for each detector was determined as the crossing point between the scan locus in eq. (2.1) and the previously-reported dispersion relation at $T = 25$ K.[1] Since the scan locus is steep and the magnetic excitations are localized on the dispersion curve within the energy widths of several meV, q can be regarded to be almost unchanged at around the magnetic excitations on the scan locus. The obtained $S(q)$ is shown in Fig.1.

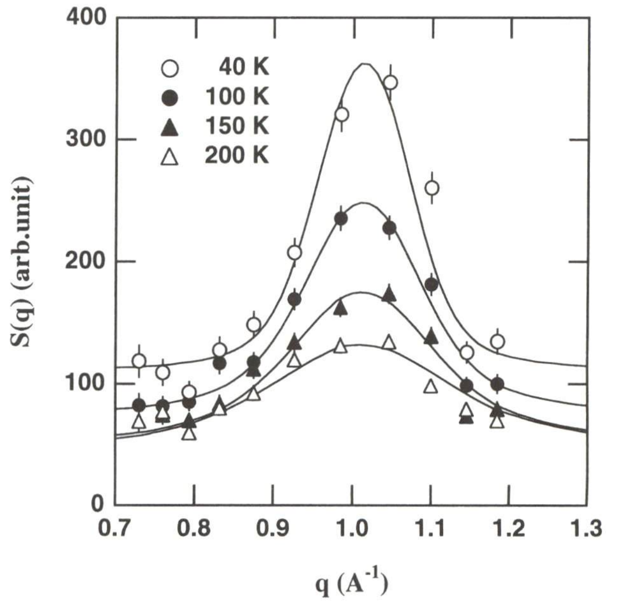

Fig. 1. The temperature dependence of $S(q)$ in $CsVBr_3$ measured on the chopper spectrometer, INC. The solid lines are the fitted curves with a Lorentzian function convoluted with the instrumental resolution adding a constant background.

§3. Result and Discussion

First, the q resolution (FWHM) was evaluated from eq. (2.1), considering k_i and k_f to be functions of E_i for a given time-of-flight, as follows:

$$(\Delta q)^2 = \left(\frac{\partial q}{\partial E_i}\right)^2 (\Delta E_i)^2 + \left(\frac{\partial q}{\partial \phi}\right)^2 (\Delta\phi)^2 + \left(\frac{\partial q}{\partial \psi}\right)^2 (\Delta\psi)^2. \tag{3.1}$$

Each derivative can be derived as follows:

$$\frac{\partial q}{\partial E_i} = \frac{k_i}{2E_i}\left(\sin\psi - \frac{L_1}{L_2}\frac{E_f}{E_i}\sin(\phi-\psi)\right), \tag{3.2}$$

$$\frac{\partial q}{\partial \phi} = k_f \cos(\phi - \psi), \tag{3.3}$$

$$\frac{\partial q}{\partial \psi} = k_i \cos\psi - k_f \cos(\phi-\psi), \tag{3.4}$$

where E_f is the final neutron energy, L_1 and L_2 are the flight paths between the moderator and the sample and between the sample and the detector, respectively. Similarly, one can derive the relation, $\Delta\omega/\Delta E_i = 1 + L_1/L_2$, where $\Delta\omega$ is the resolution in the energy transfer at $\omega = 0$. At present, $\Delta\omega$ was determined to be 4.1 meV (FWHM) by measuring the width of the elastic incoherent scattering. On INC, $L_1 = 8.2$ m and $L_2 = 2.5$ m, thus, ΔE_i was obtained to be 1.0 meV. Since ϕ is small, the angular resolution can be given in the same manner as that in the small angle neutron scattering instrument, as follows:[11]

$$(\Delta\phi)^2 = \frac{8\ln 2}{12}\left\{\left(\frac{w_m}{L_1}\right)^2 + \left(\frac{w_s}{L'}\right)^2 + \left(\frac{w_d}{L_2}\right)^2\right\}, \tag{3.5}$$

with the widths of the moderator, sample and detector, w_m, w_s and w_d, respectively, where $1/L' = 1/L_1 + 1/L_2$. Since $w_m = 10$ cm, $w_s = 2$ cm and $w_d = 2.5$ cm, $\Delta\phi$ was obtained to be 0.013 (=0.7°). Finally, $\Delta\psi$ is the measured mosaic spread. Δq can be calculated as a function of q, as listed in Table I.

Assuming the resolution function to be Gaussian, the observed $S(q)$ was fitted to the following scattering func-

Table I. The q resolution, Δq, calculated for each detector with the scattering angle, ϕ, for $E_i = 80$ meV and $\psi = 9.73°$. The values of q for individual detector is taken from that of the crossing point in the (q, ω) space between the scan locus and the dispersion curve determined at $T = 25$ K,[1] and the value of ω is also listed.

ϕ (°)	q (Å^{-1})	ω (meV)	Δq (Å^{-1})
5.48	0.73	46.7	0.082
6.09	0.76	42.7	0.079
6.69	0.79	37.8	0.076
7.30	0.83	31.7	0.074
7.91	0.88	24.2	0.073
8.52	0.93	15.2	0.074
9.12	0.99	5.9	0.077
9.73	1.05	10.2	0.075
10.34	1.01	19.6	0.073
10.95	1.15	27.7	0.073
11.55	1.18	34.2	0.074

tion:

$$S(q) = \int \frac{A}{\sqrt{2\pi}\sigma} \exp\left(-\frac{(q-q')^2}{2\sigma^2}\right) \frac{\kappa/\pi}{(q'-q_0)^2+\kappa^2} dq' + B, \quad (3.6)$$

where $\sigma = \Delta q/\sqrt{8\ln 2}$, and A, q_0, κ and B are adjustable parameters. In the fit, Δq was given as a function of q, as listed in Table I. As shown in Fig.1, the observed spectrum was well fitted to eq. (3.6). It is noted that the intensity at the data point at $q = 1.18$ Å^{-1} was, first, larger than the fitted curve at any T, because the observed inelastic spectrum exhibits excitations at around $\omega = 30$ meV, which should be of phononic origin. We plotted $S(q)$ in Fig.1 with this contribution subtracted. First, parameterizing all the adjustable parameters, the error bars for parameters were very large. Then, A was fixed to be the averaged value, because integrated intensity over (q, ω)-space should be constant independent of T, and, $\kappa(T)$ was obtained as shown in Fig.2.

In classical theory, $\kappa(T)$ is expressed by $\kappa(T) = T/(2JS2a)$, where a is the lattice constant.[12] The solid line in Fig.2 is $\kappa(T)$ calculated from this formula with $J = 99$ K. The measured $\kappa(T)$ is in good agreement with the theory, κ can be extrapolated to vanish at $T = 0$ K. This result is consistent with current theory: at $T = 0$ K, half integer spin systems show power-law decay in the spin correlation and integer spin systems show exponential decay.[8] The presently-obtained linear T-dependence of κ at the observed T-range is also consistent with the result from the numerical study for an S=3/2 system.[9] The present result in $CsVBr_3$ is consistent with our previous result in $CsVCl_3$.[1]

§4. Conclusion

We measured the static magnetic correlation function, $S(q)$, in an S=3/2, 1D Heisenberg antiferromagnet, $CsVBr_3$, at temperatures between 40 K and 200 K above T_N, on the chopper spectrometer, INC. In order to discuss one-dimensionality in the present system, magnetic properties were investigated at T higher than T_N, where the properties in the 3D ordered state should be negligible. The observed $S(q)$ was well fitted to a Lorentzian function convoluted with the instrumental resolution function which was given by the geometrical configuration of the spectrometer as well as the observed energy resolution. The determined $\kappa(T)$ showed a linear T-dependence in the observed T-range, and qualitatively agreed with classical theory. This is consistent with current theory, also with our previous result in $CsVCl_3$.

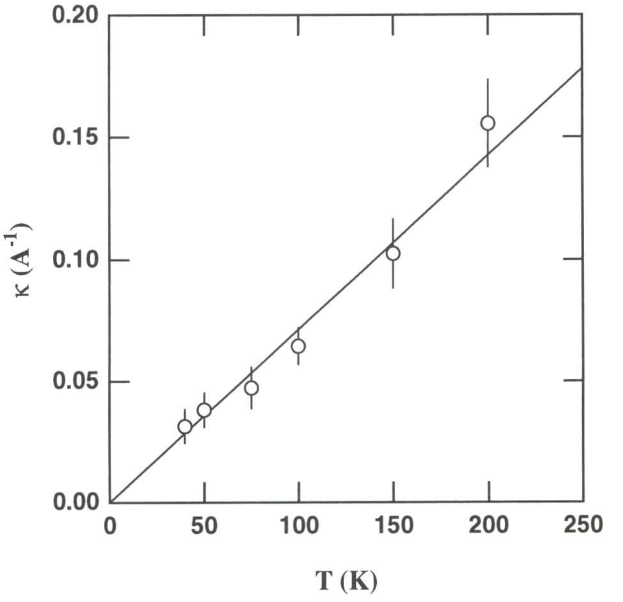

Fig. 2. Temperature dependence of the inverse magnetic correlation length $\kappa(T)$ in $CsVBr_3$ measured on INC (open circles). The solid line is the classical prediction for $J = 99$ K.

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