Propagating Domain Walls in $CsCoBr_3$

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Neutron scattering from the quasi one-dimensional Ising-like antiferromagnet $CsCoBr_3$ has revealed that a well-defined inelastic peak occurs at elevated temperatures. The peak is associated with the Villain mode arising from the propagation of thermally activated domain walls.

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The spin dynamics of the antiferromagnetic Ising-like chain is dominated by the motion of domain walls, or solitons. In 1975 Villain[1] predicted that the presence of thermally excited domain walls should lead to a low-frequency component of the longitudinal neutron scattering, bounded by a peak whose frequency obeys a sinusoidal dispersion law. In recent experiments of Yoshizawa *et al.*[2] on the quasi one-dimensional material $CsCoCl_3$, a non-Lorentzian inelastic shoulder appeared at elevated temperatures, but no peaks corresponding to a Villain mode were observed. In this Letter we report on neutron-scattering experiments on the one-dimensional antiferromagnet $CsCoBr_3$, in which a well-defined mode has been observed directly for the first time.

The $S=\frac{1}{2}$ Ising-like antiferromagnetic chain has an exchange Hamiltonian

$$H=2J\sum_i \{S_i^z S_{i+1}^z+\epsilon[S_i^x S_{i+1}^x + S_i^y S_{i+1}^y]\}$$
$$=H^{zz}+H^{xy}. \qquad (1)$$

To lowest order the ground state is the Néel state where adjacent spins are aligned in opposite directions. The first excited states at $\omega\approx 2J$ are superpositions of states with antiferromagnetic domains bounded by domain walls. The transverse term H^{xy}, which couples a state with a domain wall at the site n to states with walls at $n\pm 2$ (Fig. 1) is responsible for the propagation of the walls.

In neutron scattering the spin-wave response appearing at $\omega\approx 2J$ is due to transitions from the ground state to the band of excited states; the domain-wall or soliton response appearing near $\omega\approx 0$ arises from transitions within the band of excited states. The soliton scattering has appreciable intensity only when the excited states are thermally populated.

Using perturbation theory to first order in ϵ, Ishimura and Shiba (IS)[3] have calculated the $T=0$ spin-wave response. When this approach is extended to finite temperatures[4] we obtain in addition the soliton response. To first order in ϵ for $|\omega|<\omega_Q$, where Q is the wave vector along the chain direction in units of the inverse distance between spins, the response is given by

$$S^{zz}(Q,\omega)=\frac{F(\beta)I_0(2\beta\epsilon J)e^{\beta\omega/2}\cosh[\beta/2\cot(Q)(\omega_Q^2-\omega^2)^{1/2}]}{\cos^2(Q/2)(\omega_Q^2-\omega^2)^{1/2}}$$

$$S^{xx}(Q,\omega)=2F(\beta)I_0(2\beta\epsilon J)e^{\beta\omega/2}\frac{\cosh[\beta/2\cot(Q)(\omega_Q^2-\omega^2)^{1/2}]}{(\omega_Q^2-\omega^2)^{1/2}}-\frac{\sinh\left[\beta/2\frac{\cos(Q)}{|\sin(Q)|}(\omega_Q^2-\omega^2)^{1/2}\right]}{|\omega_Q|}, \qquad (2)$$

where the Villain-mode frequency is $\omega_Q=4\epsilon J\times\sin(Q)$, $F(\beta)$ is a function of temperature showing a characteristic thermal activation, and $I_0(x)$ is the modified Bessel function. For $|\omega|>\omega_Q$, $S^{xx}(Q,\omega)=S^{zz}(Q,\omega)=0$.

The result for S^{zz} gives the same spectral response as that first derived by Villain using another method. Both the longitudinal and transverse responses have a square-root singularity at the cutoff frequency ω_Q. As shown below, this mode has been observed for the first time in $CsCoBr_3$.

$CsCoBr_3$ has a hexagonal structure, $c=6.261$ Å,

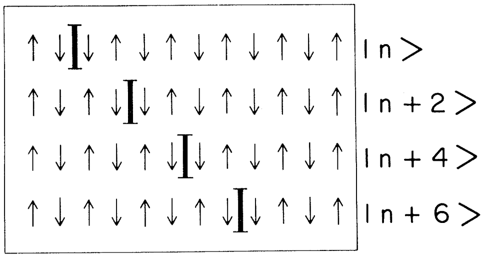

FIG. 1. Antiferromagnetic domain walls. The state $|n\rangle$ with a wall at position n is coupled to the states $|n\pm 2\rangle$.

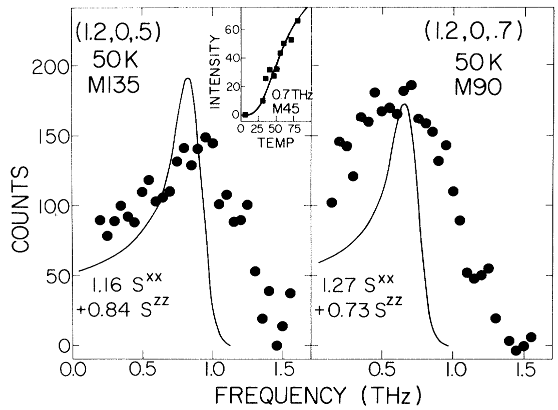

FIG. 2. Observed scattering above background at $Q=\pi/2$ (left) and $Q=7\pi/10$ (right) at a temperature of 50 K. The monitor M is a measurement of the number of incident neutrons. The theoretical curves are described in the text. Inset: Scattering above background vs temperature for $Q=\pi/2$, and frequency of 0.7 THz. The solid line is a guide to the eye and shows that the scattering is thermally activated.

$a = 7.445$ Å, isomorphous to that of $CsCoCl_3$, with chains of magnetic Co^{++} ions (two per unit cell) along the c axis. Above the three-dimensional ordering temperature ($T_{N_1} = 28.3$ K) the Co^{++} spins lie along the c axis.[5] The sample of $CsCoBr_3$ was prepared from a stoichiometric ratio of 48.69 g CsBr and 50 g $CoBr_2$. The single crystal 17 mm in diameter and 40 mm high was aligned so that its $(h,0,l)$ plane lay in the scattering plane of the N5 triple-axis spectrometer at the NRU reactor, Chalk River. A neutron beam reflected from a (111) Si monochromator and collimated to 0.6° was scattered by the specimen through a 0.7° collimator for analysis by a (111) Si analyzer operating at fixed scattered-neutron energy $E_\perp$. For most scans $E_\perp$ was 3 THz.

When the scattering vector $\vec{\kappa}$ makes an angle φ with the $z(c)$ axis, the magnetic scattering intensity is given by

$$I(\vec{\kappa}\omega) \propto V_{\vec{\kappa}}^{\,\mathrm{eff}} f(\vec{\kappa})^2 \{\sin^2\varphi S^{zz}(Q,\omega) + (1+\cos^2\varphi) S^{xx}(Q,\omega)\}, \tag{3}$$

where $V_{\vec{\kappa}}^{\,\mathrm{eff}}$ is an effective volume correcting for self-absorption of neutrons, $f(\vec{\kappa})$ is the magnetic form factor, and $Q = (\vec{\kappa}\cdot\hat{z})c/2$.

The strategy for observing the low-frequency magnetic scattering was as follows. Constant-$\vec{\kappa}$ scans were performed at various wave vectors at a temperature of 5 K, where the scattering is dominated by the elastic nuclear incoherent scattering (NIS) centered at $\omega = 0$, plus a fast-neutron background extending to higher frequencies. The intensity of the NIS at a given wave vector $\vec{\kappa}$ was taken as a measure of $V_{\vec{\kappa}}^{\,\mathrm{eff}}$. The same scan was repeated at higher temperatures (35, 50, and 80 K) and the smoothed 5-K background was subtracted from the data. The strong intensity of the NIS at $\omega = 0$ restricted the measurements of the magnetic scattering to frequencies greater than $\omega = 0.15$ THz.

Figure 2 shows data at 50 K for two wave vectors. The data show well-defined peaks which we interpret as the Villain modes arising from thermally excited propagating domain walls. The background subtracted is typically ≈45 counts at (1.2,0,0.7) and ≈90 counts at (1.2,0,0.5). The solid lines represent the theoretical line shapes for $J = 1.62$ THz, $\epsilon = 0.137$. The parameters are independently determined from spin-wave measurements, using an extended version of the IS theory.[4] The theoretical lines include $V_{\vec{\kappa}}^{\,\mathrm{eff}}$, $f(\vec{\kappa})$, monitor, polarization factor, and convolution with a Gaussian resolution function of full width at half maximum 0.18 THz. The peak at the zone boundary (1.2,0,0.5) occurs at 0.89 ± 0.07 THz. At (0.3,0,1.5), where the response is 98% transverse, a peak is observed at the same frequency in agreement with Eq. (2).

The inset of Fig. 2 shows the variation of the intensity above background with temperature. The results show that the scattering is thermally activated as expected for domain walls.

As seen in Fig. 2, the observed scattering is much broader than predicted by theory. Villain noted that broadening can be expected on the basis of collisions between domain walls. The collisional effects are of course included in the exact calculations of IS of $S^{zz}(Q,\omega)$ for rings of 10 spins. These show a broadened distribution of scattering

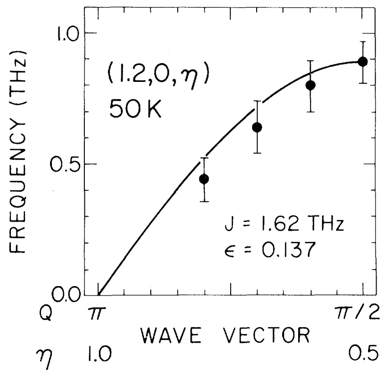

FIG. 3. Dispersion of the Villain mode. The solid line shows the Villain prediction, $\omega_Q = 4\epsilon J \sin(Q)$, for the ϵ and J which describe the spin-wave response in $CsCoBr_3$.

in qualitative agreement with the present measurements. Results at $Q=\pi/2$ (not presented here) show that at 35 K the peak is sharper while at 80 K only a squared-off shoulder remains; this confirms that the collisional effects increase with temperature.

Yoshizawa *et al.*[2] did not observe a well-defined peak in the low-frequency response of $CsCoCl_3$. The conditions of the present experiment were probably more favorable because the incoherent scattering in $CsCoBr_3$ is less than that of $CsCoCl_3$, and the single crystal used was of exceptionally high quality. It is also possible that collisional effects were somewhat greater in the chloride.

Figure 3 shows the dispersion of the peak observed in the neutron scattering. The theoretical line is that for the Villain mode with J and ϵ as above. The frequency of the mode is consistent with the expected sin(Q) dispersion. As discussed by Villain,[1] one does not expect the theory to be valid in the region $|Q-\pi| \ll \kappa_c$, the inverse correlation length. At 50 K $\kappa_c \approx 0.14\pi$ so that collisional effects are important but clearly not sufficient to damp out the peak.

A preliminary estimate of the ratio of the intensity of the transverse to longitudinal response is in reasonable agreement with theory. A complete description of these experiments and the new theoretical calculations will be presented elsewhere.

The results obtained show that at elevated temperatures a well-defined peak occurs in the low-frequency longitudinal and transverse spin response of the Ising-like antiferromagnet $CsCoBr_3$. From its dispersion, spectral line shape, and temperature dependence, the peak can be identified as arising from the Villain mode due to thermally activated propagating domain walls.

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