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LETTER TO THE EDITOR

^{81}Br-NMR in the triangular-lattice *XY* antiferromagnetic $CsMnBr_3$ below 1 K

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Abstract. The ^{81}Br-NMR of the triangular-lattice XY antiferromagnetic $CsMnBr_3$ is measured below 1.0 K. As temperature decreases starting from 350 mK, the spectrum splits up into several peaks; five sharp peaks are observed at 90 mK. The spin–lattice relaxation rate, $1/T_1$, decreases as a function of the temperature between 1.0 K and 300 mK, and then increases rapidly below 300 mK. These results suggest that a phase transition occurs below 300 mK.

A number of theoretical studies examining the magnetic phase transitions in an antiferromagnetic planar (XY) model of a triangular lattice have recently been published. Lee *et al* [1] claimed that the phase transition in this system occurs due to the symmetry breaking of helicity or chirality. Kosterlitz and Thouless [2] have explained why an order–disorder phase transition occurs by introducing the vortex and antivortex as the missing elementary excitations. At low temperatures, the vortex and antivortex form bound pairs, and these bound vortex pairs, together with spin waves, destroy the long-range spin order, producing an algebraically decaying spin–spin correlation function. The phase transition occurs when the vortex pairs are unbound [1]. Miyashita and Shiba [3] and Kawamura [4] have shown that, if the temperature tends to zero kelvin, all such phenomena will vanish, and the system then takes a uniform ground state.

$CsMnBr_3$ crystallizes into a hexagonal lattice, space group $P6_3/mmc$, with lattice parameters $a = 7.61$ Å and $c = 6.52$ Å [5]. The antiferromagnetic super-exchange interaction ($J_0 = 0.88$ meV) between the neighbouring Mn spins along the c-axis is stronger than that ($J = 0.0019$ meV) in the ab-plane [6, 7]. At low temperatures, an in-plane anisotropy ($D = 0.014$ meV), due mainly to dipolar interaction, restricts the magnetic moments to the ab-plane. Therefore, the system of the Mn spins in this material is approximately stacked into a triangular antiferromagnet XY lattice. In our previous work [8], we have pointed out that the Br-NMR spectrum, observed at helium temperature, in zero external field, is extremely wide. We have found that this is due to the distribution of the internal field. We have shown further that $CsMnBr_3$ exhibits a disordered magnetic structure property in two dimensions [8]. That is, the spins are coupled antiferromagnetically along the c-axis and point to various directions in the c-plane, and this disordered state may vanish near the ground state. In order to determine whether the disorder–order phase transition occurs, we have measured the ^{81}Br-NMR spectrum at lower temperature without an external field.

Single crystals of $CsMnBr_3$ were grown by the Bridgman method [9]. The NMR spectra were taken with an incoherent pulsed NMR apparatus with operating frequency ranging from

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30 MHz to 50 MHz. The resonance spectrum of the Br-NMR in a single crystal of $CsMnBr_3$ was measured by the spin-echo method. The spin–lattice relaxation times were obtained by measuring the spin-echo intensity as a function of the time interval between the two searching pulses (the pulse lengths are about 1 μs) and the saturation comb pulses, which break the resonance over a very wide frequency range.

We have performed an NMR experiment using a dilution refrigerator from 90 mK to 1.0 K in zero field. To measure the NMR spectra, we need to apply a radio-frequency magnetic field (rf field) to the sample in the mixing chamber. In order to avoid heating by an eddy current of the rf field, we built the mixing chamber from a plastic material (STYCAST 1266).

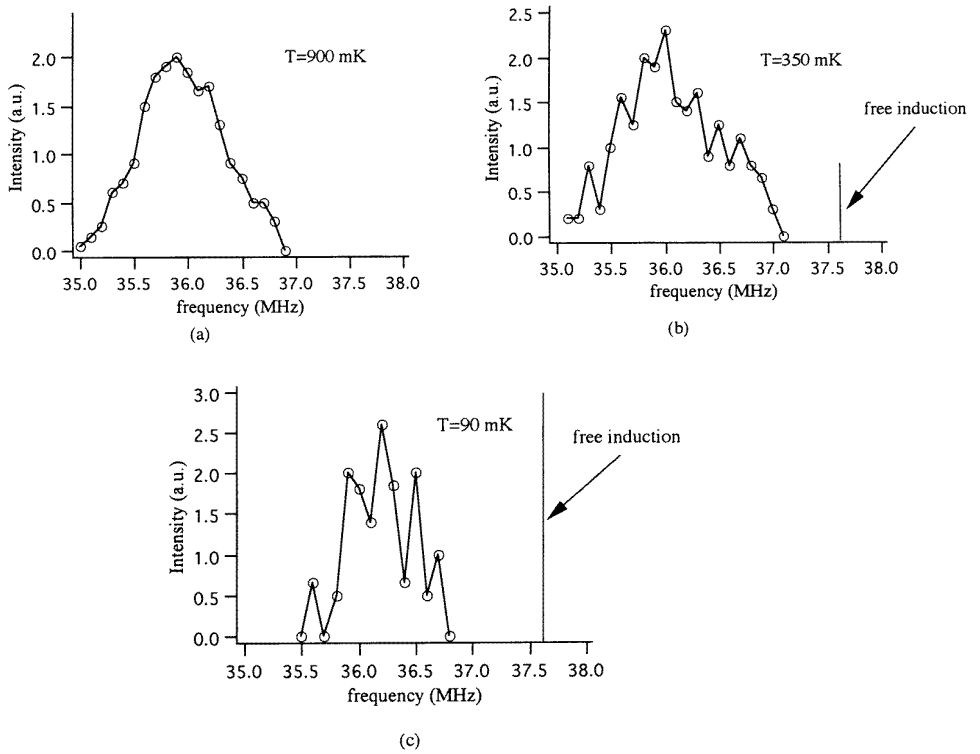

Figure 1. The spin-echo spectrum of ^{81}Br-NMR in zero external field: (a) at 900 mK; (b) at 350 mK; (c) at 90 mK. Circles represent the experimental data, and lines, connecting the circles, are drawn to make clear the spectrum form. The free-induction decay signal (very narrow, FWHM $\simeq$ 50 kHz) can only be represented by lines.

The results of the ^{81}Br-NMR measurement are shown in figure 1. In this figure (a) shows the spectrum at 900 mK; the spectrum form is similar to the one at 4.2 K, which is shown in figure 2. These results show that the spin system is still in the disordered state [8] above 900 mK. In this disordered state, the entire spin system in the c-plane can be separated into: region A containing trains of spins; and region B including the Y-like combinations of spins. We can see that the two types of small ordered-state area exist [8], as shown in figure 3. As the temperature decreases, the spectrum splits up into several peaks at 350 mK, as shown in figure 1(b); and five sharp peaks were observed at 90 mK, as shown in figure 1(c). The results on the temperature dependence of the spin-echo intensity at 35.84 MHz

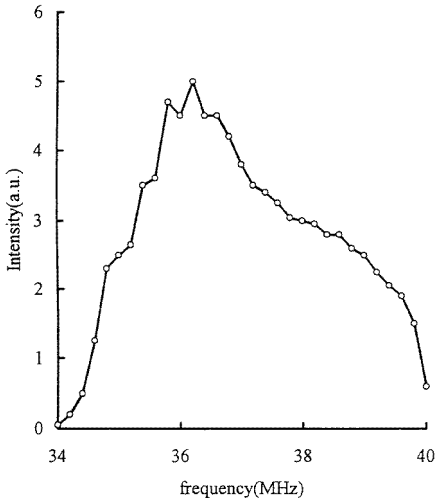

Figure 2. The spin-echo spectrum of ^{81}Br-NMR at 4.2 K in zero external field. Circles represent the experimental data, and lines, connecting the circles, are drawn to make clear the spectrum form.

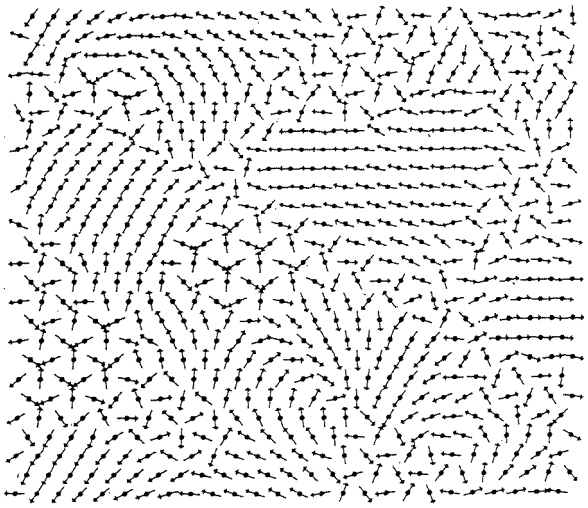

Figure 3. The Monte Carlo simulation result, obtained using a configuration of the actual crystal, a stacked triangular-lattice antiferromagnet with a finite size (26×26 spins and ten planes).

are shown in figure 4(a); the intensity is inversely proportional to the temperature between 200 mK and 1.0 K, and a dip is observed at 170 mK. A free-induction-decay (FID) signal (very narrow resonance line: FWHM $\simeq$ 50 kHz) is also observed below 350 mK at 37.52 MHz. The temperature dependence of the intensity of the FID at 37.52 MHz is shown in figure 4(b); the intensity increases rapidly with decreasing temperature. We can assume that, below 350 mK, the small ordered-state regions enlarge more and more, and the disordered state tends to the uniform state.

As we can observe in figures 1(a) to 1(c), the centre frequency of the spin-echo spectrum

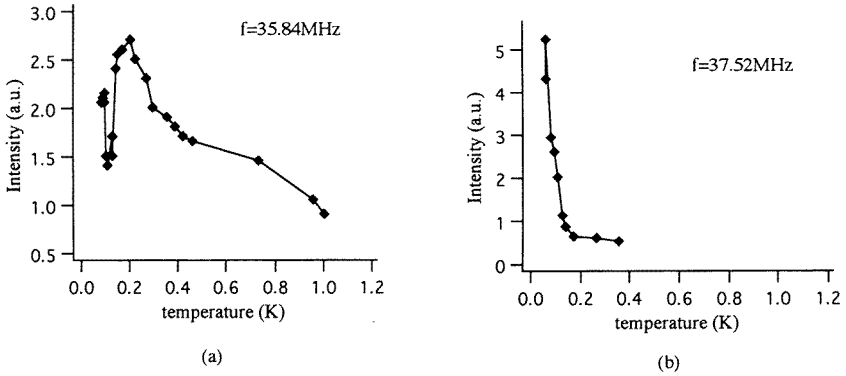

Figure 4. (a) The temperature dependence of the spin-echo intensity at 35.84 MHz. (b) The temperature dependence of the free-induction-decay signal intensity at 37.52 MHz. Circles represent the experimental data, and lines, connecting the circles, are drawn to make clear the intensity change.

increases slightly with decreasing temperature, as shown. This effect might be a consequence of a frequency-pulling effect as reported in reference [10]. However, the FID frequency is temperature independent. We can suppose that the FID signal is the pure quadrupole resonance (PQR), which is determined by an electric field gradient at a nuclear site. When the temperature tends to a phase transition point, the increasing spin fluctuation cancels the internal field, and the PQR can be observed.

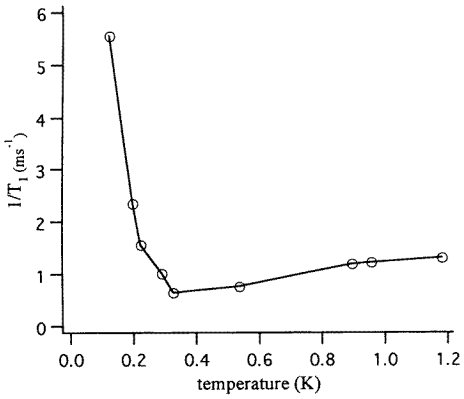

Figure 5. The temperature dependence of the spin–lattice relaxation time rate $1/T_1$. Circles represent the experimental data, and lines, connecting the circles, are drawn to make clear the rate change.

The spin–lattice relaxation time (T_1) of the ^{81}Br-NMR was measured from 90 mK to 1.0 K. We have measured the frequency dependence of T_1 at helium temperature, and found that T_1 (over the spectrum) is frequency independent. Therefore, we assume that T_1 also does not vary over the spectrum at lower temperature and measure T_1 at the defined frequency 35.84 MHz. The results for the relaxation rate ($1/T_1$) are shown in figure 5; $1/T_1$ decreases with the temperature decrease between 1.0 K and 300 mK, and increases rapidly below about 300 mK. It is well known that the relaxation rate ($1/T_1$) is proportional

to the low-frequency component of the spin–spin correlation function [11]. Furthermore, in the vicinity of a phase transition temperature, the fluctuation of the spins and the relaxation rate $(1/T_1)$ increase rapidly. From figure 5, the relaxation rate $(1/T_1)$ is still increasing with decreasing temperature at 100 mK. After reaching a maximum, at a possible transition point, it should decrease again. Therefore, we can suppose that this spin system approaches a new phase transition point below 300 mK.

In conclusion, it is probable that a phase transition has occurred below 300 mK. We have shown [8] that $CsMnBr_3$ exhibits a disordered magnetic structure property in two dimensions at helium temperature. This disordered state may vanish close to the uniform ground state when temperature tends to zero kelvin in accordance with the theory of Miyashita and Shiba [3] and Kawamura [4]. We can suggest that a disorder–order phase transition occurs below 300 mK in $CsMnBr_3$ on the *c*-plane. In order to demonstrate conclusively that this phase transition occurs in our system, we need more complete theory to analyse the NMR spectra.

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