Investigation of NMR in a quasi-one-dimensional antiferromagnet CsMnBr₃


ᵃP. Kapitza Institute for Physical Problems, RAS, 117334 Moscow, Russia,
bInstitute of Spectroscopy, RAS, 142090 Troitk, Moscow Region, Russia

Abstract

The NMR spectrum of CsMnBr₃ has been studied. It splits into 3 branches in agreement with the theory of an triangular antiferromagnet. All branches are affected by a dynamic interaction with the electronic AFMR mode.

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CsMnBr₃ is a hexagonal crystal, which becomes antiferromagnetic at Tₙ = 8.3 K. Neutron scattering data [1] have shown that CsMnBr₃ is an easy plane antiferromagnet with a quasi-one-dimensional triangular spin structure. Two features established in [2,3,4] are important for understanding our NMR results: 1. The magnetic field \( \mathbf{H} \) applied in the easy plane adjusts one pair of the 6 sublattices perpendicular to \( \mathbf{H} \) and rotates the other two pairs towards one another until the triangular structure is transformed into a quasi-collinear structure above \( H_c = 64 \) kOe.

2. The AFMR spectrum contains one low frequency mode at \( \nu_1 = \gamma D/|H|^2 \).

We have studied the NMR spectrum of CsMnBr₃ in a wide range of frequencies (200 - 500 MHz) and in magnetic fields up to 80 kOe. The static and the RF fields were applied in the basal plane. The measurements were performed in the temperature range between 1.3 K and 30 K. Preliminary results of these measurements have been published in [5].

The resonance frequency \( \nu_{\omega_0} \) of the nuclear spins of Mn²⁺ is mainly defined by the hyperfine field \( H_{HF} = -A(S)/\gamma_n h \), where \( A \) is the hyperfine interaction constant and \( S \) the average spin of the magnetic ion. Using the relations for the magnetic field dependence of the angles \( \theta_i \) between the sublattice magnetizations and the magnetic field given in [2] we have calculated the field dependence of the NMR frequencies \( \nu_{\omega_0} \) for two sublattice magnetizations oriented perpendicular to \( \mathbf{H} \) and \( \nu_{\omega_0} \) for two other pairs of sublattices. They are represented in Fig. 1 by dotted lines 1', 2, 3.

The real NMR spectrum of CsMnBr₃, which we have observed is indicated in Fig.1 and 2 by dots. It was found to split into 3 branches which agrees well with a theoretical prediction for triangular antiferromagnets. It also clearly demonstrates

![NMR spectrum of CsMnBr₃ (high frequencies).](image)

Fig. 1. NMR spectrum of CsMnBr₃ (high frequencies).
the deformation of the spin triangles at the transition to the collinear structure. All three NMR branches are affected by a strong dynamic interaction with the low frequency electronic (AFMR) mode. This interaction gives rise to a frequency shift ("pulling").

Above 45 kOe the effect of pulling becomes very small. In Fig.1 the experimental points for the upper - 3 and the lowest - 2 branches are in satisfactory agreement with the corresponding calculated curves. But for the middle branch - 1 we have observed an unexpected behavior. We can not give an explanation for it but propose the following empirical relationship for the field dependence of the frequency $\nu_{\omega,0}$ in the absence of a dynamic interaction: $\nu_{\omega,0} = \nu_{\omega,0}^0 (1 + cH')$, where $c = 1.92 \times 10^{-5} \text{kHz}^{-2}$. In Fig.1 this relation is represented by the dotted line 1.

In previous publications [6,7,8] the pulling of the NMR and AFMR spectra was calculated only for 2- or 4-sublattice collinear antiferromagnets. Recently Zaliznyak and co-authors [9] have calculated the frequencies of coupled oscillations in CsMnBr$_3$ for the lowest AFMR mode and for all three NMR branches. But his relations can be used only below $\approx 30$ kOe when the magnetic structure is practically not distorted by the applied field. Here we are presenting the result of more complicated calculations.

The equation derived for the frequencies of all 4 modes (three quasi-nuclear and one quasi-electron) is the following:

$$\nu_i^2 - \nu_0^2 = \frac{1}{3} \sum_{\omega,0} \frac{(\nu_i^2 + \nu_{\omega,0}^2 - \nu_{\omega,0}^2)\nu_{\omega,0}^2}{(\nu_{\omega,0}^2 - \nu_i^2)}.$$  \hspace{1cm} (1)

where $\nu_{\omega,0}$ is given by (1) (curve 1) and $\nu_{\omega,0}$ for $i = 2, 3$ are represented by curves 2,3 calculated using the relations from [2]. The equations for $\nu_i$ follow:

$$\nu_i^2 = \nu_{\omega,0}^2 \left( 1 - \frac{\nu_i H}{\nu_{\omega,0}} \cos(\theta_i) \right);$$ \hspace{1cm} (2)

$\theta_i$ are the angles between the sublattice magnetizations and the applied field. In the equations for the coupled oscillations (1) there are two parameters: $\nu_{\omega,0}$ and $\nu_{\omega,0}$. The value of the first corresponds to the gap in the AFMR mode; we used for it the value obtained in the experiments of Zaliznyak et al. [9]. The second parameter - $\nu_{\omega,0}$ is equal to the NMR frequency at $H = 0$. Its value was derived from the best fit of the calculated NMR spectrum to our experimental data. This best fit is represented by solid lines in Fig.1 and 2. All the experimental data below $H_c = 64$ kOe agree well with the calculated curves. From the obtained value $\nu_{\omega,0} = 415$ MHz (using $A = -1.53 \times 10^{-14}$ erg [10]) we have calculated the average spin of Mn$^{2+}$ in CsMnBr$_3$ to be $\langle S \rangle = 1.8$.

This corresponds to the following reduction of the spin momentum due to magnetic quantum fluctuations: $\delta S/S = (S - \langle S \rangle)/S = 28\%$. This value is in a good agreement with the data received by other methods [1-4].

It is surprising that above $H_c$, where a first approximation CsMnBr$_3$ should behave like a quasi-two-sublattice antiferromagnet with one NMR frequency, we have observed two branches of NMR.

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References


Fig. 2. NMR spectrum of CsMnBr$_3$. 