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LETTER TO THE EDITOR

Static magnetic properties of the quasi-one-dimensional hexagonal antiferromagnet $CsMnBr_3$

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Abstract. The static magnetic properties of the easy plane antiferromagnet $CsMnBr_3$ were studied by means of a three-coordinate vibrating sample magnetometer in fields up to 75 kOe and in the temperature range 1.7–80 K. It was revealed that in some field interval, $\boldsymbol{H} \perp \boldsymbol{c}$, a magnetization component $\boldsymbol{M} \parallel \boldsymbol{c}$ appears in the crystal. The results obtained indicate that spins deviate from the basal plane. The phase diagram corresponding to this deviation is discussed.

Recently, a considerable number of theoretical and experimental studies have been devoted to the magnetic properties of the quasi-one-dimensional hexagonal antiferromagnet $CsMnBr_3$ (space group D^4_{6h} with lattice parameters $a = 7.61$ Å and $c = 6.52$ Å, density $D = 4.30\ \mathrm{g\ cm^{-3}}$ [1]). This interest is generated, in part, by a combination of the quasi-one-dimensionality and non-collinear magnetic structure of this antiferromagnet, which result in a non-trivial magnetic phase diagram [2].

In accordance with data obtained in elastic neutron diffraction experiments [3], $CsMnBr_3$ has, for $H = 0$ and below the three-dimensional magnetic ordering temperature $T_N = 8.32$ K, a stacked triangular structure with spins lying in the basal easy plane of a crystal, aligned antiferromagnetically along the main c-axis with angles of 120° between the neighbouring spins in the basal plane. The magnetic properties of $CsMnBr_3$ are usually described by the quadratic Hamiltonian

$$\mathcal{H} = 2J \sum_i \boldsymbol{S}_i \boldsymbol{S}_{i+\Delta c} + 2J' \sum_i \boldsymbol{S}_i \boldsymbol{S}_{i+\Delta a,b} + D \sum_i (S^z_i)^2 - g\mu_B \boldsymbol{H} \sum_i \boldsymbol{S}_i \tag{1}$$

with $J = 0.88$ meV, $J' = 0.0019$ meV, $D = 0.014$ meV (at $T \Rightarrow 0$ K) [4].

Using the classical approach to the Hamiltonian (1), Chubukov [5] calculated the dependences of the magnetization $\boldsymbol{M}$ and antiferromagnetic resonance frequencies ω_i in $CsMnBr_3$ on the magnetic field directed along and perpendicular to $\boldsymbol{c}$. According to the results of his calculations, in the field $\boldsymbol{H} \perp \boldsymbol{c}$, one of the magnetic sublattices in the basal plane, $\boldsymbol{S}_1$, is directed perpendicularly to $\boldsymbol{H}$, while two others, $\boldsymbol{S}_2$ and $\boldsymbol{S}_3$, are directed at angles of $\pm\pi/6$ to $\boldsymbol{H}$.

As the field increases, the first sublattice cants towards the field, while the angle between the two others, 2Θ, decreases from its initial value $\pi/3$ according to the law

$$\cos\Theta = [2 - (H/H_c)^2]^{-1} \tag{2}$$

and reaches zero at $H = H_c = (48JJ')^{1/2}S/g\mu_B$. In fact, under the condition $D > 3J'$, which, according to [4], occurs in $CsMnBr_3$, the spins do not leave the basal plane in any field H. Obviously, the above also refers to the spins, S_4, S_5 and S_6, lying in the neighbouring plane at a distance of $c/2$ from the plane considered above. A phase transition of the second order corresponds to the convergence of sublattices already described. Calculations based on the data in [4] yield $H_c = 61$ kOe.

Further magnetic field increase at $H_c < H < \tilde{H}_c \approx 8JS/g\mu_B$ as well as at $\boldsymbol{H} \parallel \boldsymbol{c}$, $H < H_c$ results in a smooth turn of the spins towards the field direction: the angle, α, between the $\boldsymbol{S}_i$ and $\boldsymbol{H}$ is determined by the equation

$$\cos \alpha = g\mu_B H/8JS. \tag{3}$$

As a result it is possible to derive the following equations for the magnetization $\boldsymbol{M}$, which is parallel to $\boldsymbol{H}$,

$$\mathrm{M} = (g\mu_B)^2 NDH\{1 + 2/[2 - (H/H_c)^2]^2\}/24J\mathrm{M} \tag{4}$$

at $H \leqslant H_c$, $\boldsymbol{H} \perp \boldsymbol{c}$ and

$$\mathrm{M} = (g\mu_B)^2 NDH/8J\mathrm{M} \tag{5}$$

at $H_c \leqslant H < \tilde{H}_c$, $\boldsymbol{H} \perp \boldsymbol{c}$ and $H < \tilde{H}_c$, $\boldsymbol{H} \parallel \boldsymbol{c}$, where N is Avogadro's number, M is the molecular weight. Gaulin *et al* [2], in experiments with elastic neutron diffraction, investigated the H, T magnetic phase diagram in fields up to 65 kOe directed in the basal plane of the crystal. They confirmed the conclusion of Chubukov [5] regarding the existence of an intermediate phase with convergent sublattices between triangular and paramagnetic phases and obtained the value $H_c(T = 2\ \mathrm{K}) = 64$ kOe.

The aim of the present work is to investigate the static magnetic properties of $CsMnBr_3$ and to subject the conclusions of Chubukov [5] to experimental verification.

The measurements of the magnetic moment were carried out by means of a vibrating sample magnetometer with a superconducting magnet and three pairs of measuring coils. This device permitted simultaneous measurements of the three orthogonal components of the magnetic moment of the sample, one of which, M_x, was parallel to the magnetic field. The axis of vibration of the sample was perpendicular to $\boldsymbol{H}$. The absolute precision of the measurement of M was about 7%, while relative deviation of M in one experiment was measured with greater precision: about 3%. The temperature of the sample above 4.2 K was measured by a thermocouple of Au + Fe-chromel with a precision of about 3%, and below 4.2 K, according to the saturation pressure of helium vapour, with a precision of about 0.1 K. The measurements were carried out on single crystals with dimensions around $2 \times 2 \times 2\ \mathrm{mm}^3$.

The results of the measurements of the component of magnetization parallel to $\boldsymbol{H}$, for $\boldsymbol{H} \parallel \boldsymbol{c}$, $M_{x\parallel}$, and for $\boldsymbol{H} \perp \boldsymbol{c}$, $M_{x\perp}$, are shown in figure 1. It is evident that the dependences $M_x(H)$, on the whole, are adequately described by equations (4) and (5), excepting two deviations. Firstly, at $H > H_c$ the curve $M_{x\perp}(H)$ remains below $M_{x\parallel}(H)$ and secondly, this curve is steeper than the theoretical (i.e. differs from the dependence $M \propto H$, which follows from (5)). The first of the observed deviations can be explained by the anisotropy of the g-factor ($g_\parallel > g_\perp$); the second, by the assumption that the convergence of the sublattices is not completed in the field H_c. The latter, obviously, could be linked to the deviation of $\boldsymbol{H}$ from the basal plane. In order to exclude this trivial possibility, we rotated $\boldsymbol{H}$ in the plane perpendicular to the basal one at intervals of 1°. Nevertheless, we were unable to avoid the observed phenomenon.

The temperature dependences of specific magnetic susceptibilities, measured at $H =$ 22.5 kOe, in which the dependence $M_x(H)$ is still practically linear, are shown in figure

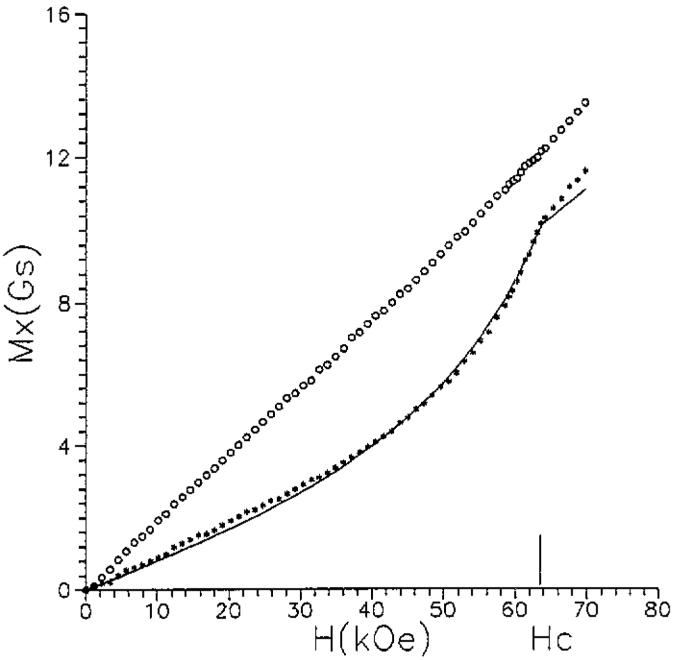

Figure 1. Field dependences of the magnetizations $M_{x\parallel}$(O) and $M_{x\perp}$(*); $T = 1.7$ K. The full curve is calculated using theory [5].

2. The results of these measurements are close to the dependencies $\chi(T)$ measured earlier [3] in the field $H = 15.3$ Oe. From this, it follows that the anisotropy of susceptibility is preserved up to $T \sim 80\,\mathrm{K} \gg T_N$. The value of $J = 0.89$ meV, derived by means of (5) from the dependence $M_{\parallel}(H)$, coincides within the range of experimental error with the data in [4].

The most substantial result, in our view, is illustrated in figure 3. This figure shows the field dependence of the magnetization component $M_z \parallel c$, measured at $\boldsymbol{H} \perp \boldsymbol{c}$. Note that due to the smallness of M_z ($M_z < 0.03\, M_{x\perp}$), the background signal was subtracted from the signal induced in the z-coils. This background signal was mainly the result of a paramagnetic moment induced in the heating coil, wound from constantan wire on a sapphire sample holder, and evident due to the high sensitivity of the measurements. From figure 3, it can be observed that the magnetization M_z appears in the sample in the vicinity of H_c. The maximum value of M_z is reached for $H \approx H_c$. The result obtained indicates that the convergence of the sublattice magnetizations is accompanied by spin deviations from the basal plane. The temperature dependence of the field H_c, in which M_z is maximal, is shown in figure 4. This dependence is in good agreement with the temperature dependence of the critical field H_c, measured in [2], and interpreted as a field in which the sublattices converge.

The sum of the results obtained indicates that instead of the transition to the convergent phase, predicted by the theory [5], for the aforementioned values of the parameters J' and D, the resultant phenomenon is a transition to the intermediate phase, with the component $M_z \neq 0$. The description of the static magnetic properties of $CsMnBr_3$, on the basis of the thermodynamical potential with all second-order terms in

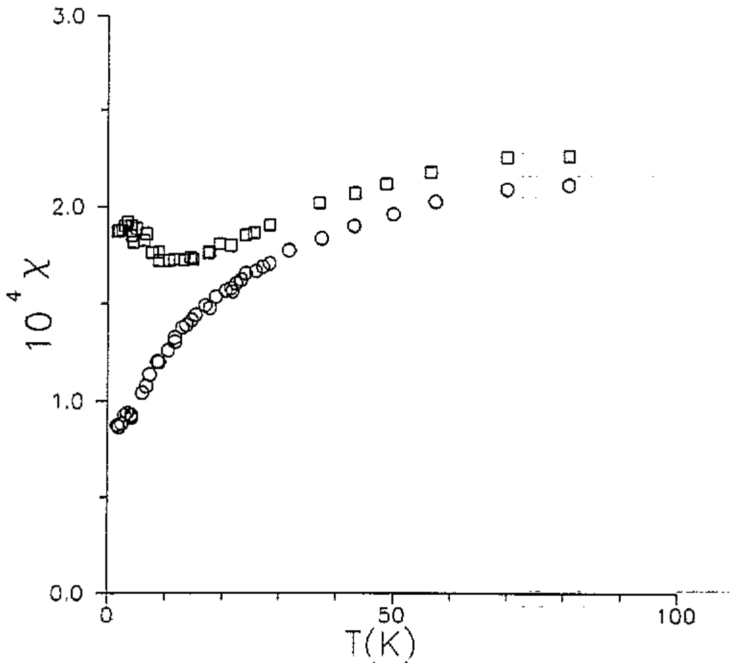

Figure 2. Temperature dependences of the single-crystal magnetic susceptibility χ for $\boldsymbol{H} \parallel c$ (□) and $\boldsymbol{H} \perp c$ (○); $H = 22.5$ kOe.

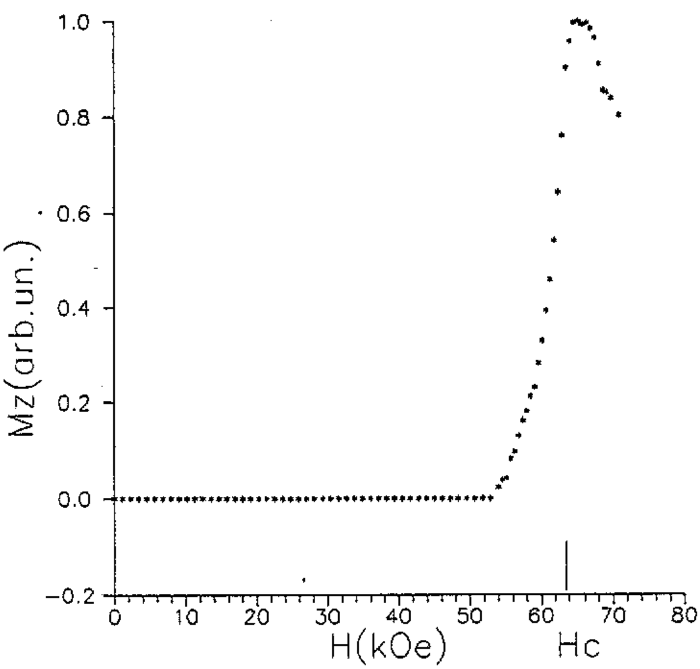

Figure 3. Field dependence of the magnetization $\boldsymbol{M}_z \parallel c$; $\boldsymbol{H} \perp c$, $T = 1.7$ K.

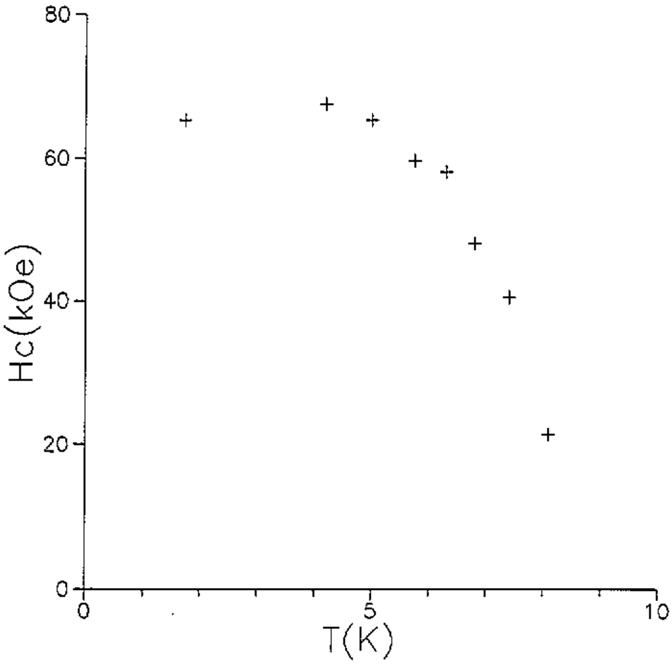

Figure 4. Temperature dependence of the critical magnetic field H_c, obtained from the $M_z(H)$ dependence.

l_i and $\boldsymbol{m}$ allowed by crystal symmetry (l_i and $\boldsymbol{m}$, $i = 1$–3, are linear combinations of $\boldsymbol{S}_j$, $j = 1$–6, which realize the irreproducible representations of the space group of the crystal), shows that, at the definite ratios of one of the anisotropy constants and J', the angle 2Θ between sublattices S_2 and S_3 (see above) decreases with the field $\boldsymbol{H} \perp \boldsymbol{c}$ in accordance with (2). At some value of the field $H_{c1} < H_c$ the sublattices S_2 and S_3 leave the basal plane. In fact, all three vectors S_i lie in the same plane σ and S_1 belongs to both σ and basal planes.

The angle γ between these planes increases from $\gamma = 0$ at $H = H_{c1} < H_c$ to $\gamma = \pi/2$ at $H = H_{c2} > H_c$. Under the intermediate values of γ, $M_z \neq 0$. Hence there is the intermediate 'angle' phase, and reorientations occur via two phase transitions of the second order. It is important that the 'angle' phase exists due to the anisotropy of susceptibility χ, and the field width $H_{c1} < H < H_{c2}$ of this phase is proportional to this anisotropy. A detailed description of this new phase will be given elsewhere.

Unfortunately, this proposed explanation of the experimental data leaves unsettled the problem of accordance of ratios between anisotropy and exchange in the plane parameters used in our theory, and determined in neutron experiments [4] and in the study of antiferromagnetic resonance spectra [6]. In the above mentioned papers the fitting with experimental data was established on the basis of a Hamiltonian (1), which contains fewer parameters than allowed by the crystal symmetry. For example, the obvious term $\Sigma_{i,j} S_i^z S_j^z$ also defining the crystal anisotropy is absent in (1). Thus, to solve this problem it is necessary to calculate magnon spectra in $CsMnBr_3$, using the complete Hamiltonian.

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