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Spin reorientation in the easy-plane hexagonal antiferromagnet under a canted magnetic field

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Abstract. Spin reorientation process in the quasi-one-dimensional easy-plane antiferromagnet on a stacked triangular lattice has been considered theoretically and studied in an experiment on CsMnBr_3 . The second-order phase transition associated with the flip of two pairs of sublattices has been shown to persist if the magnetic field is canted at an angle φ from the basal plane. The field and angular dependences of the magnetic torques measured at $T = 1.8$ K, although in qualitative agreement with the classical theory, demonstrate a strong effect of quantum fluctuations.

1. Introduction

The ground-state magnetic properties of the quasi-one-dimensional antiferromagnets on a stacked triangular lattice are currently of great theoretical and experimental interest. Initially this was stimulated by Haldane's [1] conjecture about the singlet ground state of the weakly anisotropic 1D antiferromagnetic array of integer spins, but now much attention has been attracted also by the peculiarities associated with the non-collinear spin arrangement occurring in these systems at $T < T_N$ owing to the influence of weak interchain exchange.

The spin systems of most hexagonal halide compounds ABX_3 (A being an alkaline metal, and B a metal of the 3d group) with space symmetry group D_{6h}^4 can be treated as a set of weakly coupled linear antiferromagnetic chains disposed on a triangular lattice in the basal plane. Numerous experiments (ENS and NMR) indicate that the majority of such compounds (CsNiCl_3 , RbNiCl_3 , CsMnBr_3 , CsVI_3 , etc) experience, at $T < T_N$, triangular magnetic ordering; adjacent spins in the basal plane form an angle close to 120° with each other while spins in each chain are antiparallel. Anisotropic interactions of relativistic origin usually choose the chain direction to be easy (as in CsNiCl_3 or RbNiCl_3) or hard (CsVI_3 and CsMnBr_3) axis. The magnetic properties of such compounds are believed to be well described by the Hamiltonian (the Z' axis being along C_6 (figures 1 and 2))

$$\mathcal{H} = J \sum_{ij} \mathcal{S}_i \mathcal{S}_j + J' \sum_{ij} \mathcal{S}_i \mathcal{S}_j + D \sum_i (\mathcal{S}_i^z)^2 - \gamma H \sum_i \mathcal{S}_i. \quad (1)$$

The first term here describes the intrachain exchange interaction, the second the exchange in the basal plane, and the third and fourth the anisotropy energy and Zeeman

energy of the spins, respectively, in the external field H . In the present paper we shall consider the case of the easy-plane antiferromagnet: $J > 0$, $J' > 0$, $D > 0$. Quasi-one-dimensionality implies the smallness of the ratio J'/J which, as well as D/J , is usually about 10^{-2} – 10^{-3} .

Recent classical calculations [2, 3] of the reorientation process in such a system starting from the Hamiltonian (1) have shown that the principal parameter to define the course of reorientation is $d = D/3J'$. If the increasing magnetic field is applied in the basal (easy) plane of the compound with $d > 1$ (strong planar anisotropy outweighs weak interchain exchange J'), then spins remain in the basal plane and at $H = H_c = (48S^2JJ')^{1/2}$ the second-order phase transition associated with the flip of two pairs of sublattices occurs. If under the same conditions $d < 1$ is the case, then at $H = H_c^{sf} = (16S^2JD)^{1/2} < H_c$ the planar arrangement becomes unstable and the first-order spin-flop transition takes place; spins tilt from the basal plane to become almost perpendicular to the field. Then, as the field increases, at H_c' (close to the saturation field H_c^{sat} if d is not very close to unity), the second-order spin-flip transition like that in the case $d > 1$ happens. Subsequent reorientation in both cases proceeds in a similar way; all spins smoothly cant to the field direction saturating at $H = H_c^{\text{sat},1} = 8SJ + 18SJ'$. If the magnetic field is applied along the hard axis C_6 , the spin reorientation for any value of d is not accompanied by any phase transition until the full saturation of $H = H_c^{\text{sat},2} = 8SJ + 18SJ' + 2SD$.

If in the case $d < 1$ the magnetic field is applied at some non-zero angle φ to the basal plane, the instability of spin arrangement at H_c^{sf} disappears and the first-order transition is absent (just as in the case of the spin-flop in the easy-axis compound CsNiCl_3 [4]). The main purpose of the present investigation was to study the spin reorientation process in the case $d > 1$ (it is believed to correspond to CsMnBr_3 , in which the fit of the classical expressions to the measured AFMR frequencies yields $J = 214$ GHz, $J' \approx 0.5$ GHz and $D = 1.95$ GHz [5]) when the magnetic field is applied at an auxiliary angle φ to the basal plane.

2. Classical theory

A classical ground-state spin configuration is obtained through the minimization of (1) with the spin operators \mathcal{S} (where $\mathcal{S}^2 = S(S+1)$) substituted by the classical vectors of size S . First we consider the situation far from saturation: $H \ll H_c^{\text{sat}}(\varphi) \approx H_c = 8SJ$.

Because of the non-zero field component in the hard-axis direction, spins leave the basal plane from the very start of the reorientation. In contrast with the case of the spin-flop transition at $d < 1$, in our case spin flip as described above takes place in the canted field but at some higher $H = H_c(\varphi)$. In the leading order in D/J , J'/J and H/H_c we have

$$H_c^2(\varphi) = H_c^2(d-1)/(d \cos^2 \varphi - 1) \quad H_c^2 = 48S^2JJ' \quad (2)$$

and the angles designated in figures 1(a) and 1(b) are the following:

$$\alpha = \pi/2 - (H/H_c) \cos \varphi \quad (\beta + \gamma)/2 = \pi/2 - (H \cos \varphi \cos \zeta)/(H_c \cos \eta) \\ \varphi_1 = \pi/2 - (H/H_c) \sin \varphi \quad (3)$$

$$(\varphi_2 + \varphi_3)/2 = \pi/2 - (H/H_c)(\sin \varphi \cos \eta - \cos \varphi \sin \eta \sin \zeta)$$

where $(\beta - \gamma)/2 = \zeta$ and $(\varphi_2 - \varphi_3)/2 = \eta$ are given by the equations

$$\cos \zeta = [(1 + A^2)/(1 + B^2)]^{1/2} \quad \cos \eta = (A/B)[(1 + B^2)/(1 + A^2)]^{1/2} \quad \text{at } H \ll H_c(\varphi) \quad (4a)$$

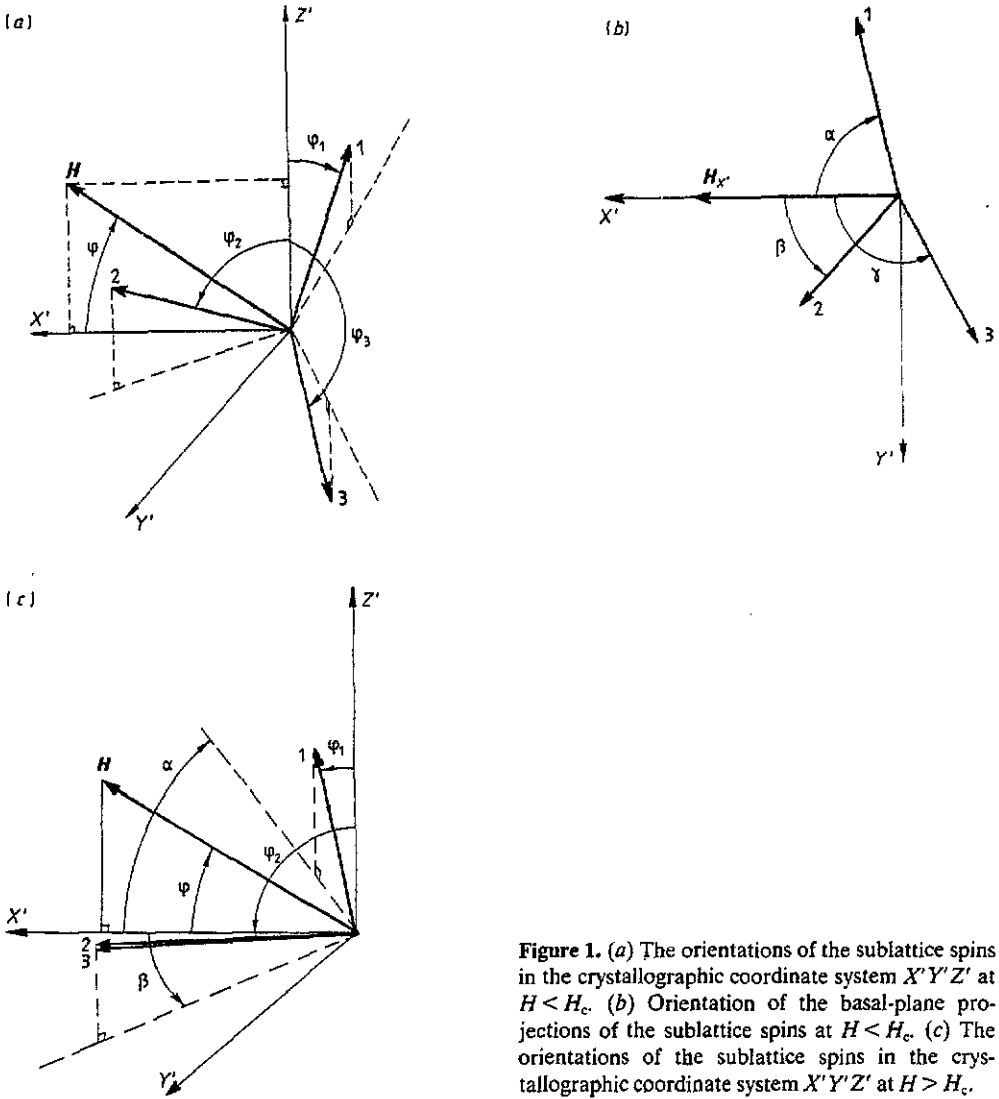


Figure 1. (a) The orientations of the sublattice spins in the crystallographic coordinate system $X'Y'Z'$ at $H < H_c$. (b) Orientation of the basal-plane projections of the sublattice spins at $H < H_c$. (c) The orientations of the sublattice spins in the crystallographic coordinate system $X'Y'Z'$ at $H > H_c$.

and

$$\cos \zeta = 1 \quad \cos \eta = 1 \quad \text{at } H \geq H_c(\varphi). \tag{4b}$$

Here $H_e = 8SJ$ as introduced above, and A and B are defined by

$$\begin{aligned} A &= Y - (1 + Y^2)^{1/2} & B &= A(2 - z \cos^2 \varphi) - z \sin \varphi \cos \varphi \\ Y &= [z \cos(2\varphi) - d]/[z \sin(2\varphi)] & z &= (H/H_c)^2. \end{aligned} \tag{5}$$

As the field increases (but at $H \ll H_e$), the angles $\alpha, (\beta + \gamma)/2, \varphi_1$ and $(\varphi_2 + \varphi_3)/2$ almost do not change and are close to $\pi/2$, while the angles $\beta - \gamma$ and $\varphi_2 - \varphi_3$ (in fact, the angle between sublattices 2 and 3 as well as between $2'$ and $3'$ in figure 1(a)) undergo

a quick change and in our case $d > 1$ turns into zero at the critical field. Equation (2) for the critical field $H_c(\varphi)$ corresponds to the condition $B^2 \geq A^2$, that is essential for equations (4a) to be valid. When φ is increased, $H_c(\varphi)$ increases, rising to infinity at $\varphi = \varphi_c = \cos(1/\sqrt{d})$, which means that our approximation fails and H_c becomes approximately equal to H_c instead of being much smaller as was presupposed. To investigate the correct behaviour of $H_c(\varphi)$ if it is not small in comparison with H_c , we have performed another expansion of the equations minimizing (1). In the vicinity of the flip transition keeping terms to the first non-vanishing order in ζ , $\eta \rightarrow 0$ we derived the critical field for the transition to be given by the real root of

$$\begin{aligned} (H^2/H_c^2)[1 - (3J'/4J)(t+1)][(c+d\cos^2\varphi)/2 - 1 + (D/8J)(2-t)\sin^2\varphi] \\ = (3J'/8J)(2-t)(t-2+d) \end{aligned} \quad (6)$$

$$t(t-1)[1 - (H^2/H_c^2)(1 - (D/2J)\sin^2\varphi)] = (3J'/2J)(H^2/H_c^2)(2-t^3).$$

The parameter t in these equations denotes the ratio

$$t = [(\sin\varphi_1)/(\sin\varphi_2)][(\sin\alpha)/(\sin\beta)]. \quad (7)$$

If d is not very close to unity, one can obtain the following: if $d\cos\varphi \geq 1 + 3J'/2J$, then $t = 1$ and $H_c(\varphi)$ is given by (2); if $d\cos\varphi \leq 1 - 3J'/2J$, then $t = 2 - d\cos^2\varphi$, and

$$H_c(\varphi) = H_c\{1 - (D/2J)\sin^2\varphi + (3J'/2J)[2 - t^3/t(t-1)]\}^{-1/2}. \quad (8)$$

Note that the last expression is also correct in the case $d < 1$. The subsequent reorientation up to saturation proceeds in the same way as in [2, 3] (see figure 1(c)) with

$$\begin{aligned} \cos\alpha &= (H/H_c)[(\cos\varphi)/(\sin\varphi_1)]\{1 - (3J'/2J)[(1+t)/t]\} \\ \cos\beta &= (H/H_c)(\cos\varphi)/(\sin\varphi_2)\{1 - (3J'/2J)[(1+t)/2]\} \\ \cos\varphi_1 &= (H/H_c)\sin\varphi\{1 - (3J'/2J)[(1+t)/t] - D/4J\} \\ \cos\varphi_2 &= (H/H_c)\sin\varphi\{1 - (3J'/2J)[(1+t)/2] - D/4J\} \end{aligned} \quad (9)$$

where t is given by (7). Implying $\alpha, \beta \rightarrow 0$ and $\varphi_1, \varphi_2 \rightarrow \varphi$ one obtains $t = 2$ and the saturation field to be $H_c^{\text{sat}}(\varphi) = 8JS + 18J'S + 2DS\sin^2\varphi$.

Considering figures 1(a) and 1(b), one can easily derive the following expressions for the basal plane M'_x and the hexagonal axis M'_z components of the net magnetic moment per mole:

$$\begin{aligned} M'_x &= g\mu_B N_A S (H/H_c) \frac{1}{3} [(3 - 2\sin^2\zeta \cos^2\eta) \cos\varphi - \sin\zeta \sin(2\eta) \sin\varphi] \\ M'_z &= g\mu_B N_A S (H/H_c) \frac{1}{3} [(1 + 2\cos^2\eta) \sin\varphi - \sin\zeta \sin(2\eta) \cos\varphi] \end{aligned} \quad (10)$$

where μ_B is the Bohr magneton and N_A the Avogadro number. In experiment we measure the longitudinal (along the field) M_x and the transverse M_y magnetic torques (figure 2) which are given by

$$M_x = M'_x \cos\varphi + M'_z \sin\varphi \quad M_y = -M'_x \sin\varphi + M'_z \cos\varphi. \quad (11)$$

Curves obtained through the substitution of (4)–(6) into (11) are drawn in figures 3 and 5 as broken curves. At $H = H_c(\varphi)$ they demonstrate a break inherent to the second-order phase transition. When the field is approaching its critical value, the transverse magnetization $M_y(H)$ drops rapidly to become zero at $H \geq H_c$ while the relative changes in $M_x(H)$ are not so pronounced.

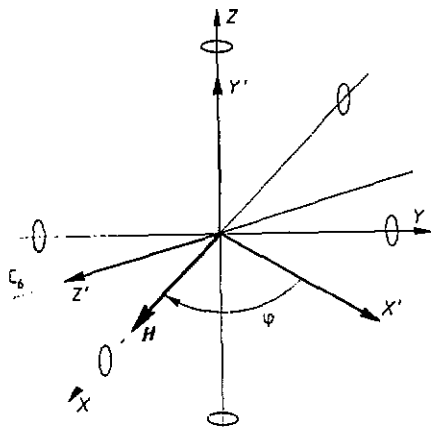


Figure 2. The mutual orientation of the sample $X'Y'Z'$ and experimental (tied to the pick-up coils which are denoted by the open circles) XYZ coordinate systems.

3. Experimental procedure and results

To verify the results of our calculations we have undertaken a set of magnetostatic measurements on CsMnBr_3 samples. The large single crystals of CsMnBr_3 were grown from a melt by the Bridgman techniques with subsequent annealing as described in [5]. The crystals cleaved excellently down the binary planes and were quite hygroscopic and so had to be handled with caution. The samples to be investigated were orthogonal prisms with the linear dimensions 1–2 mm. They were cleaved from the inside of the bulk single crystals just before the experiment and stuck with polystyrene glue to avoid hydration. However, in late spring, when the air became mild and damp, the intensive moisture condensation made it almost impossible to obtain a good unhydrated sample with such a straightforward procedure. Here we shall report the experiments performed in winter and early spring with good samples.

The magnetic torque measurements were taken using a triple-axis vibrating-sample magnetometer with a 75 kOe superconducting magnet [6]. The specimen was attached with its binary plane to the platform which vibrated along the vertical Z axis and which could be rotated around this axis. The magnetic field was applied along the horizontal X axis. In this set-up the hexagonal C_6 axis of the crystal always lies in the horizontal X – Y plane and can be directed at an angle in the range $\pm 135^\circ$ with respect to the field (figure 2). Three pairs of pick-up coils permitted the simultaneous measurement of the three orthogonal components of the magnetic moment, but use was made of only X and Y coils to measure the component M_x along the magnetic field and the component M_y perpendicular to it and lying in the plane containing the field and the C_6 axis which we believed to be given by (11).

The magnetization curves $M_x(H)$ and $M_y(H)$ obtained at $T \approx 1.8 \text{ K} \ll T_N$ at some angles φ between H and C_6 are shown in figures 3 and 4. The break corresponding to the second-order transition at $H_c(\varphi)$ was very pronounced on all $M_y(H)$ curves until $H_c(\varphi)$ remained inside the range of our experiment. Since the magnitude of the transverse magnetization is small ($M_y(H) \leq 0.1M_x(H)$ at $\varphi \leq 15^\circ$), the experimental error in its measurement was about 10 times larger than that of the longitudinal magnetization which was 1% or less. The installation was calibrated using a superconducting lead cylinder of $m = 0.45 \pm 0.02 \text{ mg}$ with a ratio of length to diameter of about 30 at $T =$

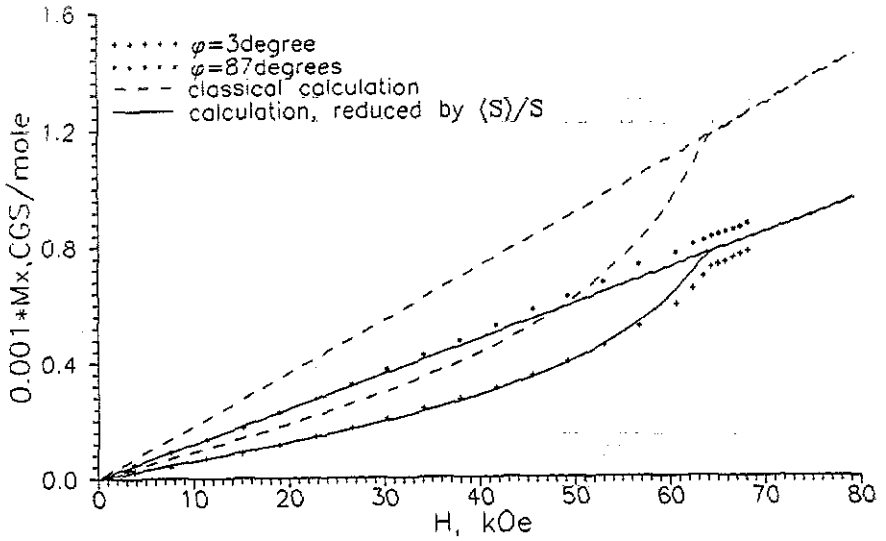


Figure 3. The longitudinal magnetizations $M_x(H)$ at $T = 1.8$ K.

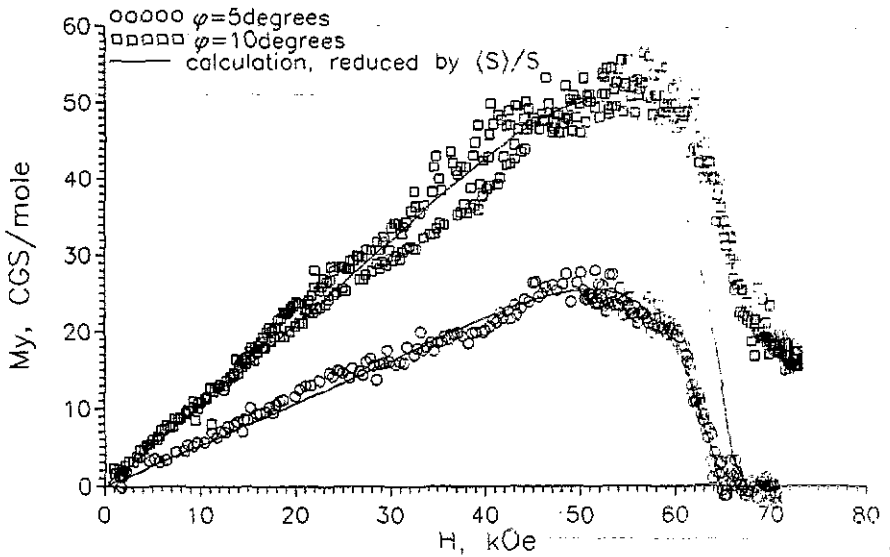


Figure 4. The transverse magnetizations $M_y(H)$ at $T = 1.8$ K.

4.25 K with an absolute accuracy of about 6%. Hence, the resultant accuracy of the determination of the absolute values of molar susceptibilities in our experiment was about 6% for $M_x(H)$ and about 11% for $M_y(H)$.

Another way to investigate the spin-flip transition at $H_c(\varphi)$ is to measure the transverse magnetization $M_y(\varphi)$ in the constant applied field $H > H_c$ when the angle φ is varied. To perform such experiment, we equipped our magnetometer with a special circuit to produce a voltage proportional to the angle φ . The resultant curves for some

different values of H are presented in figure 5. In the flipped phase (at small angles $\varphi \leq \varphi_c(H)$), $M_y(\varphi)$ grows linearly and slowly with φ . At $\varphi = \varphi_c(H)$ it exhibits a break and then a steep increase to become approximately sinusoidal at $\varphi \geq 45^\circ$.

4. Discussion

As is evident from figures 3–5 the classical theory developed in section 2 adequately describes the general features of the spin reorientation process in the quasi-one-dimensional hexagonal antiferromagnet CsMnBr_3 in the arbitrarily oriented magnetic field. The angular dependence of the critical field $H_c(\varphi)$ (figure 6) is within the range of our experiment in good agreement with equation (2) if we take $d = 1.8$ (full curve in figure 6), although there are important divergencies between the classical theory and experiment that need to be considered.

First, the measured magnitudes of the molar magnetic torques are about 1.5 times smaller than those obtained from the classical theory if we use the same parameter H_c as in the resonance measurements [5] and $d = 1.8$, which is far beyond our experimental error. It should be mentioned that the absolute values obtained in our experiment agree within 6% with the results of the measurement of $M_x(H)$ when $H \parallel C_6$ by Goto *et al* [7], although Kotyuzhanskii and Nikiforov [8] reported different values. In our opinion, such a large discrepancy between the classical calculations and experiment has to be attributed to the influence of the quantum fluctuations which were ignored in section 2. It was shown earlier on the basis of the $1/S$ expansion [2] that the quantum corrections to the classical frequencies of the antiferromagnetic resonance in such system cause only the parameters J , J' and D of the Hamiltonian (1) in the resultant formulae to be substituted by their renormalized values \tilde{J} , \tilde{J}' and \tilde{D} which are obtained from the fit to the experiment. In fact, AFMR measurements [5] gave the renormalized value $\tilde{d} = \tilde{D}/3\tilde{J}' = 1.3$ (somewhat lower than 1.8), but the classical value to be used in classical approximation can roughly be estimated through $d \approx \tilde{d}(S/\langle S \rangle)(1 - 1/2S)^{-1} \approx 2.46$ [1]. Therefore, if we use the classical expressions to fit the magnetic torque measurements, we also must expect some renormalized value of d , which may differ from that derived from the resonance. We believe that $d = 1.8$ is the 'corrected' value for d that should be involved instead of the classical value in the expressions for magnetizations given in (3)–(11)†. Direct calculation of the quantum corrections to the ground-state configuration of the quasi-one-dimensional triangular antiferromagnet in the magnetic field is a highly complicated and to date unresolved problem, but its most appreciable effect is known to be the sublattice spin contraction. As was established by the neutron diffraction [9] the magnitude of the Mn^{2+} ($S = \frac{5}{2}$) magnetic moment in CsMnBr_3 in zero external field extrapolated to $T = 0$ K (and at $T = 1.8$ K already) is only about $3.3\mu_B$ instead of $5\mu_B$ demonstrating substantial zero-point spin deviation ($\langle S \rangle \approx 1.65$). Our results indicate that we can roughly take quantum corrections into account if we multiply our classical expressions for the magnetizations with the renormalized H_c as derived from the AFMR and INS experiments and $d = 1.8$, by the relative spin reduction $\langle S \rangle/S$ (figures 3–5).

† It should be mentioned that the magnitude of the magnetization if the classical expressions are used is sensitive mainly to the renormalization of H_c which is only about 8% for $S = \frac{5}{2}$, being equal $(\pi - 2)/2\pi S$ in the first order in $1/S$ [2].

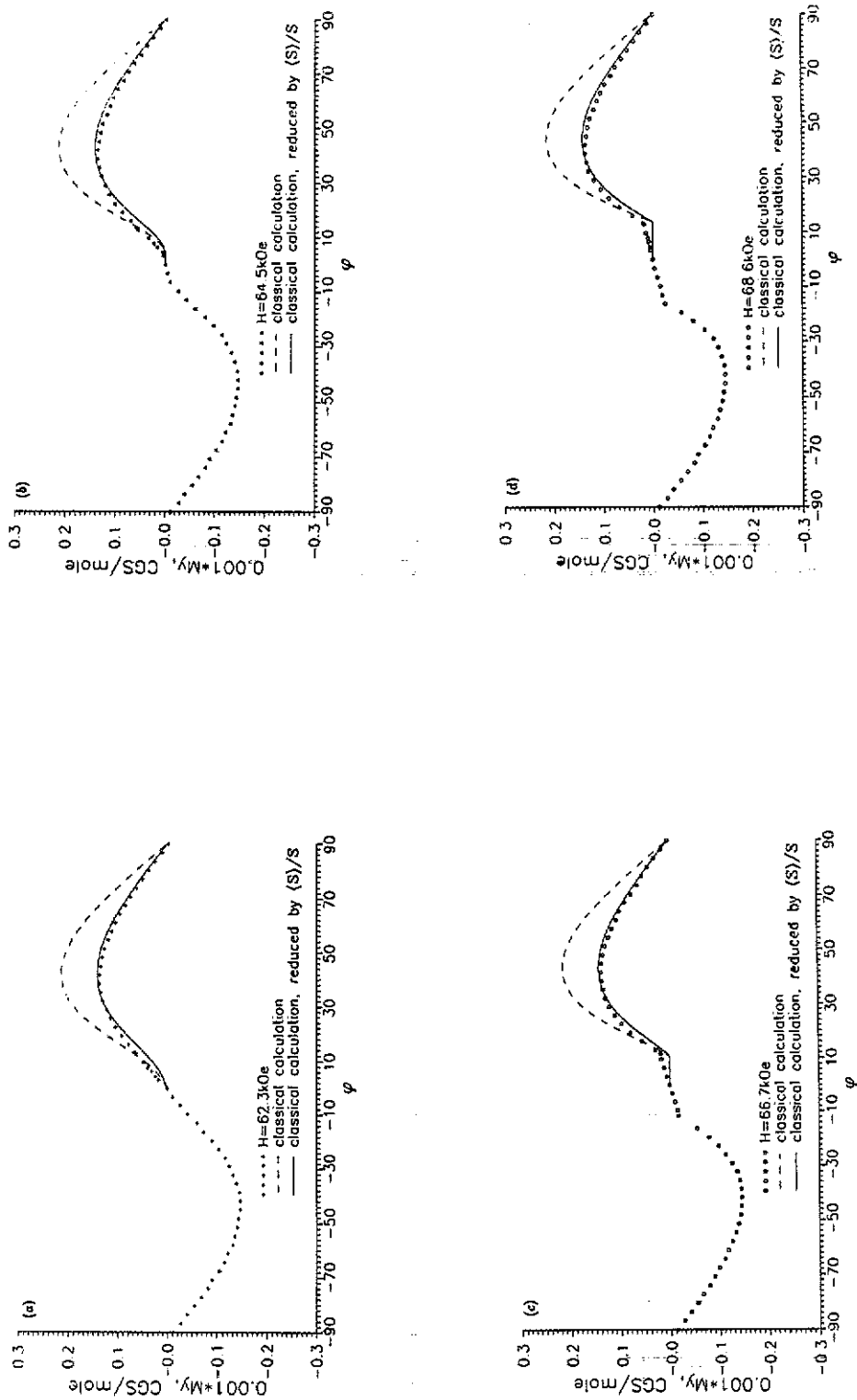


Figure 5. (a)–(d). The angular dependences of the transverse magnetization $M_y(\varphi)$ at $T = 1.8$ K.

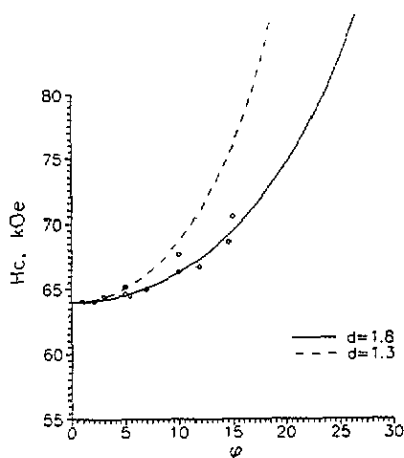


Figure 6. The angular dependence of the critical field $H_c(\varphi)$.

The other discrepancy which cannot be accounted for through the simple multiplication by $\langle S \rangle / S$ is the visible deviation of the functional dependences of the longitudinal magnetization $M_x(H)$ from the classical formulae giving the 10% anisotropy between M_x^\perp (when H is nearly perpendicular to the basal plane, the upper curve in figure 3) and M_x^\parallel (when H almost lies in the basal plane, the lower curve) at $H \geq H_c$. This anisotropy naturally reveals itself also in the transverse magnetization dependences, causing $M_y(\varphi)$ at $H \geq H_c$ to have a finite slope at $\varphi \leq \varphi_c$ instead of being equal to zero (figure 5) and $M_y(H)$ at $\varphi > 0$ to have a non-zero value at $H \geq H_c$ (figure 4). To describe these peculiarities of the magnetization curves, accurate consideration of the anisotropy of quantum fluctuations and their dependences on the magnetic field is necessary.

Acknowledgments

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