Spin-flip phase transitions in the quasi-1*D* hexagonal antiferromagnet $CsMnBr_3$

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(Submitted 11 October 1990)
Zh. Eksp. Teor. Fiz. **99**, 1254–1265 (April 1991)

A three-coordinate vibration magnetometer has been used to study the static magnetic properties of the easy-plane antiferromagnet $CsMnBr_3$ in fields up to 75 kOe over the temperature range 1.7–80 K. In a certain interval of fields $\mathbf{H}\perp\mathbf{c}$, a magnetization component $\mathbf{M}\|\mathbf{c}$ arises in the crystal. It is concluded from an analysis of the experimental data that the spins deviate from the basal plane of the crystal in this case. The static magnetic properties are calculated with the help of a thermodynamic potential Φ containing all the terms of second order in the vectors $\mathbf{l}$ and $\mathbf{m}$ allowed by the symmetry of the crystal. The results of these calculations show that the observed phase diagram could occur if the parameters of Φ describing the exchange interaction in the basal plane and describing the magnetic anisotropy were related in a specific way.

INTRODUCTION

The magnetic properties of quasi-1*D* hexagonal antiferromagnets with an ABX_3 structure, where the cations A and B are respectively an alkali metal ion and an ion of a 3*d* element, and X is a halide anion, have recently attracted considerable theoretical and experimental interest. The quasi-1*D* magnetic properties of such compounds are determined by the particular crystal structure of these compounds: The distances between the 3*d* cations along the principal axis are considerably smaller than in the perpendicular direction.[1]

Another characteristic feature of these compounds is that as they are cooled they go into an ordered state with a noncollinear magnetic structure, so they exhibit magnetic properties quite different from those of the well-studied collinear antiferromagnets. It is this combination of a quasi-1*D* nature and a noncollinear magnetic structure that is responsible for most of the research interest in these substances. One representative of this group of magnetic materials is $CsMnBr_3$, which furthermore has a nontrivial magnetic phase diagram.[2]

According to experimental data on elastic neutron scattering,[3] in a zero field ($H=0$) and below the temperature of 3*D* ordering, $T_N = 8.32$ K, the $CsMnBr_3$ crystal (of symmetry space group D_{6h}^4, with room-temperature lattice constants $a = 7.61$ Å and $c = 6.52$ Å; Ref. 4) has a triangular magnetic structure, with spins lying in the easy basal plane of the crystal, σ. The spins alternate in an antiferromagnetic fashion along the **c** axis (the principal axis), parallel to C_3, and neighboring spins in the basal plane make angles of 120° with each other (Fig. 1). The magnetic properties of crystals of the $CsMnBr_3$ type are usually described by a Hamiltonian which is quadratic in the spin operators,[1]

$$\mathcal{H}=2J_c\sum_i \mathbf{S}_i\mathbf{S}_{i+\Delta c}+2J_{ab}\sum_i \mathbf{S}_i\mathbf{S}_{i+\Delta a,\Delta b}$$
$$+D\sum_i (S_i^z)^2-g\mu_B\mathbf{H}\sum_i \mathbf{S}_i \qquad (1)$$

with the following parameter values as $T\to 0$: $J_c = 0.88$ meV, $J_{ab} = 0.0019$ meV, and $D = 0.014$ meV (Ref. 5). The antiferromagnetic exchange interaction is very anisotropic, differing in the directions along the principal axis (J_c) and in the basal plane (J_{ab}) ($J_c/J_{ab}\approx 460$). As a result, slightly above T_N (at $T>10$ K) the magnetic structure of $CsMnBr_3$ consists of a set of antiferromagnetic chains, and this compound exhibits properties characteristic of quasi-1*D* magnetic materials.[6]

In a classical calculation based on Hamiltonian (1), Chubukov[7] derived the magnetization M and the antiferromagnetic-resonance frequencies Ω_i of $CsMnBr_3$ as a function of the magnetic field **H**, directed either parallel to or perpendicular to the **c** axis. According to the results, in a field $\mathbf{H}\perp\mathbf{c}$ as $H\to 0$, one group of parallel spins (a magnetic sublattice; we call these spins $\mathbf{S}_1$) lie in the basal plane, perpendicular to the field **H**, while two other groups $\mathbf{S}_2$ and $\mathbf{S}_3$, make angles of $\pi/6$ and $5\pi/6$ with this field (Fig. 1b). With increasing H, the first of these sublattices rotates slightly toward the magnetic field, while the angle of 2θ between the two other sublattices decreases from its initial value of $2\pi/3$ in accordance with

$$\cos\theta=[2-(H/H_c)^2]^{-1}, \qquad (2)$$

where

$$H_c=(48J_cJ_{ab})^{1/2}S/g\mu_B, \qquad (3)$$

and vanishes at $H=H_c$. The sublattices partially collapse. If the condition $D>3J_{ab}$ holds, as it does in $CsMnBr_3$ according to the data of Ref. 5, the spins do not deviate from the basal plane at any value of H. The collapse of the sublattices consisting of the spins $\mathbf{S}_2$ and $\mathbf{S}_3$ corresponds to a second-order phase transition. This entire discussion of course also applies to the spins $\mathbf{S}_4$, $\mathbf{S}_5$, and $\mathbf{S}_6$ which lie in a neighboring basal plane, at a distance of $c/2$ from the plane under consideration ($\mathbf{S}_i\|\mathbf{S}_{i+3}$).

Using the parameter values given in Ref. 5, we find a value $H_c = 61$ kOe from (3). With a further increase in H, at $H_c<H<\tilde{H}_c\approx 8J_cS/g\mu_B$, there is a smooth rotation of the spins toward the **H** direction, as in the case $\mathbf{H}\|\mathbf{c}$, $H<\tilde{H}_c$. The angle α between the direction of the spins $\mathbf{S}_i$ and the field **H** under the conditions specified above is the same for all spins and is given by

$$\cos\alpha=g\mu_BH/8J_cS. \qquad (4)$$

Working from the calculations in Ref. 7, we find the following expressions for the magnetization of crystals with

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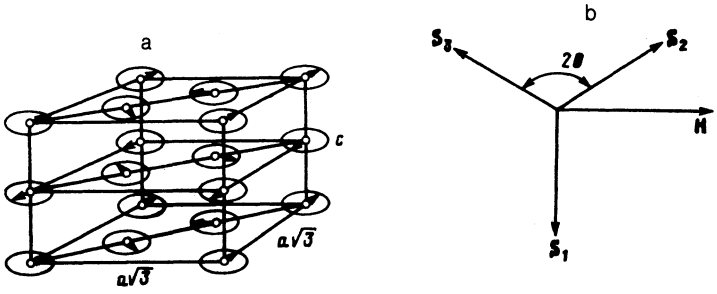

FIG. 1. a—Arrangement of the magnetic moments of the Mn^{2+} ions in the unit cell of the $CsMnBr_3$ crystal; b—magnetic moments of the sublattices in one of the basal planes of the crystal as $H \to 0$, brought to a common point.

a magnetic structure of the $CsMnBr_3$ type:

$$M = (g\mu_B)^2 N\rho H \left\{1+2\left[2-\left(\frac{H}{H_c}\right)^2\right]^{-2}\right\} \frac{1}{24 J_c \mu} \qquad (5)$$

for $H \leqslant H_c$, $\mathbf{H} \perp \mathbf{c}$ or

$$M = (g\mu_B)^2 N\rho H/8J_c\mu, \qquad (6)$$

for $H_c \leqslant H < \tilde{H}_c$, $\mathbf{H} \perp \mathbf{c}$ and also for $H < \tilde{H}_c$, $\mathbf{H} \| \mathbf{c}$, where N is Avogadro's number, μ is the molecular weight, and ρ is the density (the x-ray density of $CsMnBr_3$ is[4] $\rho_x = 4.30\,\text{g/cm}^3$).

In experiments on elastic neutron scattering, Gaulin *et al.*[2] studied the magnetic phase diagram (H,T) in fields H up to 65 kOe. The fields were applied in the basal plane of the crystal. Gaulin *et al.* confirmed the conclusions reached by Chubukov[7] regarding the existence, between the triangular and paramagnetic phases, of an intermediate phase with partially collapsed sublattices (in contrast with the phase at $H > \tilde{H}_c$, with totally collapsed sublattices). Gaulin *et al.* found a value $H_c\,(T = 2\text{ K}) = 64$ kOe. [In saying that this intermediate phase lies "between" the other phases, we mean that T is being varied at constant $H < H_c\,(0\text{ K})$.]

Results of a study[8] of the static magnetic properties of $CsMnBr_3$ agree qualitatively with the theoretical predictions of Ref. 7, but they indicate that the spins deviate from the basal plane of the crystal at fields $H \approx H_c$, $\mathbf{H} \perp \mathbf{c}$. Chubukov's theory[7] does not describe such a deviation.

Our purpose in the present study was to pursue the research on the static magnetic properties of the $CsMnBr_3$, calculating these properties with the help of a thermodynamic potential Φ containing all the terms of second order in the magnetic moments which are allowed by the symmetry of the crystal.

TEST SAMPLES AND EXPERIMENTAL PROCEDURE

The magnetization was measured on a vibration magnetometer similar to that described in Ref. 9. A horizontal magnetic field up to 75 kOe was produced by a slotted superconducting magnet consisting of two windings. With three pairs of measurement coils it was possible to simultaneously measure the three mutually perpendicular components of the magnetic moment of the sample, one of which, $\mathcal{M}_x$, was parallel to the magnetic field **H**. The vibration axis of the sample was perpendicular to **H**. The absolute error in the measurements of $\mathcal{M}$ was $\sim 7\%$, while the relative changes in $\mathcal{M}$ within a single experiment could be measured more precisely, within 3%.

Measurements were carried out over the temperature range 1.7–80 K. Above 4.2 K, the sample temperature was measured by an (iron + 0.03% gold)/chromel thermocouple within an error $\sim 3\%$; below 4.2 K the sample temperature was determined to within ~ 0.1 K from the saturation vapor pressure of helium.

The test samples were single crystals with dimensions $\sim 2\times 2\times 2$ mm. The $CsMnBr_3$ crystals are very hygroscopic; in air, they quickly decompose, converting into a white substance having a large paramagnetic susceptibility. Before each experiment, a sample was accordingly cleaved from the interior of a single-crystal boule held in a desiccator. The $CsMnBr_3$ single crystals were grown by S. V. Petrov at the Institute of Physical Problems, Academy of Sciences of the USSR.

EXPERIMENTAL RESULTS

Figure 2 shows the results of measurements of the magnetization components parallel to the field **H**: $\mathbf{M}_{x\|}$ for $\mathbf{H} \| \mathbf{c}$ and $\mathbf{M}_{x\perp}$ for $\mathbf{H} \perp \mathbf{c}$. We see that the $M_x(H)$ curve is generally described well by expressions (5) and (6), but there are two deviations. First, for $H \geqslant H_c$ the $M_{x\perp}(H)$ curve runs under $M_{x\|}(H)$; second, for $H \geqslant H_c$ the $M_{x\perp}(H)$ curve is steeper than the theoretical curve. In other words, it deviates from the behavior $M \propto H$ which would follow from (6). The first of these features can be attributed to a possible anisotropy of the g-factor ($g_\| > g_\perp$); such an anisotropy has been seen previously[1] in several quasi-1D antiferromagnetic chlorides which have different magnetic 3d ions and which are isomor-

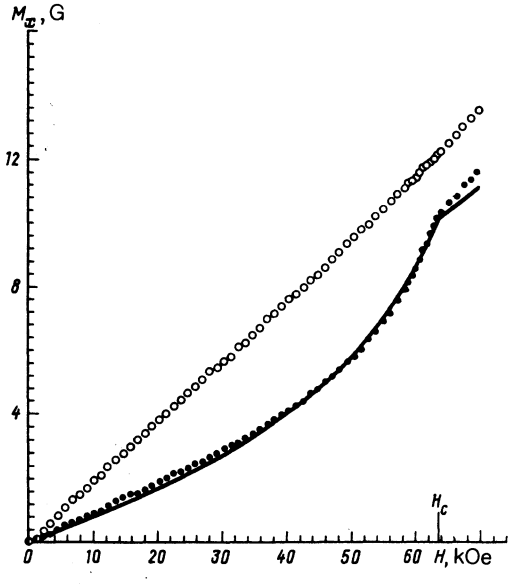

FIG. 2. Field dependence of the magnetizations (○) $M_{x\|}$ and (●) $M_{x\perp}$ at $T = 1.7$ K. The solid line is a curve of $M(H)$ calculated from the theory of Ref. 7.

phic with $CsMnBr_3$. The second of these features can be explained under the assumption that the collapse of the sublattices does not terminate at the field H_c. The latter possibility could of course stem from an imperfect stacking of the crystal and a related deviation of the field **H** from the basal plane. In an effort to rule out the possibility of this trivial explanation, we rotated **H** in a plane perpendicular to the basal plane at steps of 1°. However, we were not able to eliminate the effect.

Figure 3 shows the temperature dependence of the magnetic susceptibility in a field $H = 22.5$ kOe, at which the $M_{x\perp}(H)$ dependence is still essentially linear. It follows from the results in Fig. 3 that the magnetic susceptibility remains anisotropic up to $T \sim 80\ \text{K} \gg T_N$. The results of these measurements are approximately the same as the $\chi(T)$ dependence found previously[3] in a field $H = 15.3$ Oe. The value found from (6) with allowance for the $M_{\parallel}(H)$ dependence as $T \to 0$ is $J_c = 0.89$ meV $\pm$ 7% and agrees, to within the experimental error, with the data of Refs. 3 and 5.

The most important result, in our opinion, is that when the field **H** is parallel to the basal plane of the crystal a signal appears in the z coils in a certain magnetic-field interval. This signal falls off with increasing temperature, vanishing at $T \approx T_N$. The presence of this signal indicates the appearance of a magnetization component $\mathbf{M}_z \| \mathbf{c} \perp \mathbf{H}$ in this field interval. Figure 4 illustrates the situation with an $M_z(H)$ dependence measured at $T = 1.7$ K. Because of the small value of M_z, in analyzing the experimental data we subtracted from the signal induced in the z coils a parasitic signal which stemmed primarily from the paramagnetic moment induced in the heater (constantan wire wound around a sapphire holder). This signal turned out to be significant in cases in which a high sensitivity was required in the M_z measurements. The maximum value of M_z was reached in a field $H \approx H_c$; at the lowest temperature, $T = 1.7$ K, it was less than $\sim 3\%$ of M_x in this field. It can be concluded from this result that the collapse of the magnetic moments of the sublattices is accompanied by a deviation of these moments from the basal plane of the crystal.

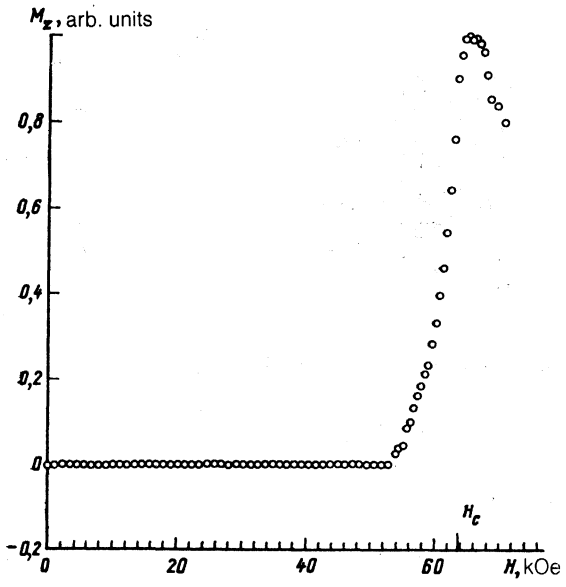

FIG. 4. Field dependence of the magnetization $\mathbf{M}_z \| \mathbf{c}$ ($\mathbf{H} \perp \mathbf{c}$, $T = 1.7$ K).

Figure 5 shows the temperature dependence of the field H_c, which we measured as the field at which M_z reached its maximum. This dependence agrees well with the temperature dependence measured for the critical field H_c in Ref. 2. That field was interpreted by Gaulin *et al.*[2] as the field at which the sublattices collapsed. Figure 6 shows the temperature dependence of the maximum value $(M_z)_{\max} = M_z(H_c)$ and of the width ΔH of the field interval in which this component of **M** is observed.

Taken together, these results seem to indicate that a transition to an intermediate phase with a component $M_z \neq 0$ occurs in $CsMnBr_3$ at a certain field H_{c1} slightly below H_c, instead of the transition, at H_c, from the triangular phase to the partially collapsed phase which is predicted by the theory of Ref. 7 with the parameter values J_{ab} and D given above. Extrapolation of the experimental data in Fig. 4 to higher fields, with allowance for the dependence $M_x(H)$

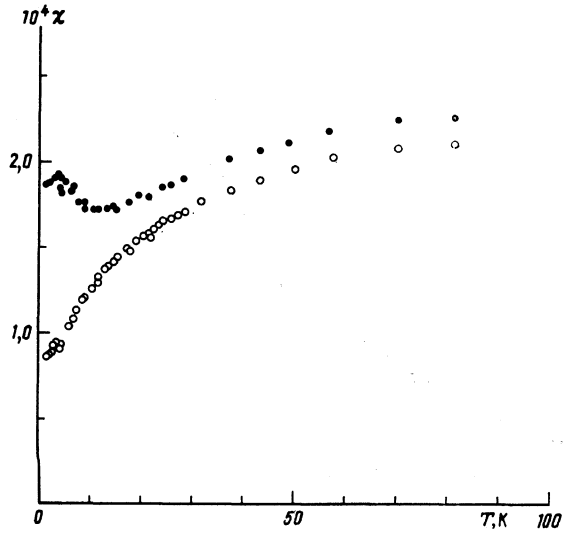

FIG. 3. Temperature dependence of the magnetic susceptibility χ of a single crystal for the orientations (●) $\mathbf{H} \| \mathbf{c}$ and (○) $\mathbf{H} \perp \mathbf{c}$ ($H = 22.5$ kOe).

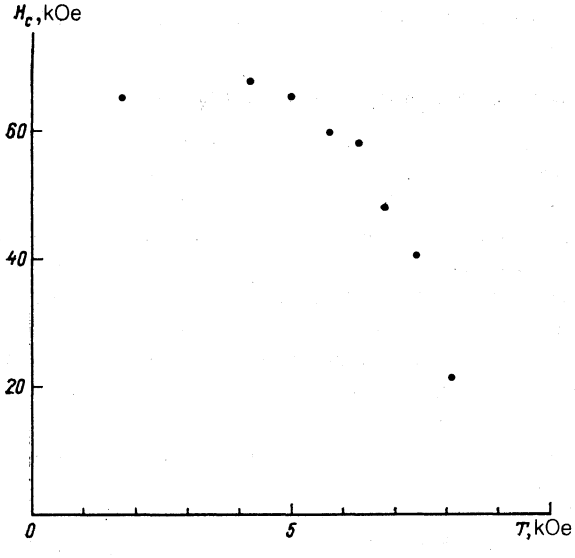

FIG. 5. Temperature dependence of the critical magnetic field H_c found from the $M_z(H)$ dependence.

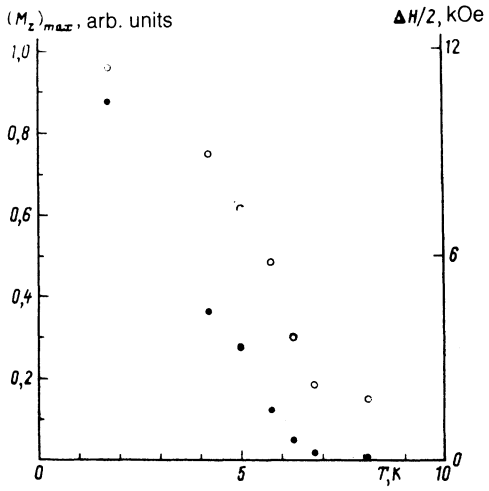

FIG. 6. Temperature dependence of (●) the maximum value $(M_z)_{\max} = M_z(H_c)$ and (○) the width ΔH of the field interval in which M_z is observed.

(Fig. 2), suggests that at a certain field $H_{c2} > H_c$ there is a transition from a phase with $M_z \neq 0$ to a partially collapsed phase or to a phase with similar properties with $M_z = 0$ and $M_x \propto H$. Consequently, between the triangular phase and the partially collapsed phase which was predicted in Ref. 7 and which has been observed previously,[2,8] or a similar phase, there is an intermediate "angular" phase with spins making an angle with the basal plane.

THEORY

It is simple to show that there exist no relations among the parameters of the Hamiltonian (1) which enable that Hamiltonian to explain the existence of a phase with a component $M_z \neq 0$ in $CsMnBr_3$ when the field **H** is applied in the basal plane of the crystal. The reason that the phase diagram generated from (1) depends on only the ratio of the parameters D and J_{ab} ($J_c \gg J_{ab} > 0$). For $D > 3J_{ab}$, as was shown above, the spins remain in the basal plane for any value of a field $\mathbf{H} \perp \mathbf{c}$, so we have $M_z = 0$.

For $D < 0$ the crystal becomes an easy-axis crystal. As $H \to 0$, the spins form the same triangular lattice, with angles of 120° between the directions of neighboring spins, as in the case with $D > 3J_{ab}$, which we have already discussed. The only distinction is that now these triangles lie in a plane perpendicular to the basal plane and to the field $\mathbf{H} \perp \mathbf{c}$. As the magnetic field is increased at $H < \tilde{H}_c$, the angle α between the directions of the spins $\mathbf{S}_i$ and **H**, as in the case with $D > 3J_{ab}$ but $\mathbf{H} \| \mathbf{c}$, is the same for all spins and varies in accordance with (2). Correspondingly, the magnetization $\mathbf{M} \| \mathbf{H}$ in this case is determined by (6) ($M_x \propto H$), and again we have $M_z = 0$ for any value of H.

In the case $0 < D < 3J_{ab}$, which we have not yet discussed, a phase transition occurs from a phase analogous to the phase in the case $D > 3J_{ab}$ to a phase corresponding to the condition $D < 0$. This transition occurs at a field $H_c^* < H_c$, whose value is determined below. In this case, of course, the magnetization $\mathbf{M} \| \mathbf{H}$ of the crystal is given by expression (5) in fields $H < H_c^*$, while in fields $H > H_c^*$ it is given by expression (6). It is simple to see that in this case we should observe a jump in the magnetization M_x at the field H_c^*, but we should have a component $M_z = 0$ at either $H < H_c^*$ or $H > H_c^*$.

On the other hand, it is clear that the Hamiltonian (1) does not contain all possible second-order terms. In particular, it lacks the obvious term $\Sigma_i S_i^z S_j^z$, which describes the magnetic anisotropy. In particular, this term stems from a dipole–dipole interaction. To generate a theoretical description of the results, we accordingly write a thermodynamic potential which contains all terms of second order in the sublattice magnetizations $\mathbf{M}_i$ that are allowed by the symmetry of the crystal. For this purpose we follow the customary procedure of introducing vectors **L** and **M** which are linear combinations of the sublattice magnetizations $\mathbf{M}_i = N\rho g\mu_B \mathbf{S}_i/6\mu$ and which are the bases of irreducible representations of the symmetry space group of the crystal:

$$\begin{aligned} \mathbf{L}_1&=\mathbf{M}_1+\mathbf{M}_2+\mathbf{M}_3-\mathbf{M}_4-\mathbf{M}_5-\mathbf{M}_6,\\ \mathbf{L}_2&=2\mathbf{M}_1-\mathbf{M}_2-\mathbf{M}_3-2\mathbf{M}_4+\mathbf{M}_5+\mathbf{M}_6,\\ \mathbf{L}_3&=3^{1/2}(\mathbf{M}_2-\mathbf{M}_3-\mathbf{M}_5+\mathbf{M}_6), \end{aligned} \tag{7}$$

$$\begin{aligned} \mathbf{L}_4&=2\mathbf{M}_1-\mathbf{M}_2-\mathbf{M}_3+2\mathbf{M}_4-\mathbf{M}_5-\mathbf{M}_6,\\ \mathbf{L}_5&=3^{1/2}(\mathbf{M}_2-\mathbf{M}_3+\mathbf{M}_5-\mathbf{M}_6),\\ \mathbf{M}&=\mathbf{M}_1+\mathbf{M}_2+\mathbf{M}_3+\mathbf{M}_4+\mathbf{M}_5+\mathbf{M}_6. \end{aligned}$$

In choosing bases, we took into account the circumstance that the magnetic unit cell does not coincide with the crystallographic unit cell ($a_m = 3a, b_m = 3b, c_m = c$): The vectors $\mathbf{L}_1$ and **M** realize two 1*D* irreducible representations of the group of translations with a star vector $\mathbf{k} = 0$, while ($\mathbf{L}_2, \mathbf{L}_3$) and ($\mathbf{L}_4, \mathbf{L}_5$) realize two 2*D* representations with vectors $\mathbf{K} = \pm [1/3, 1/3, 0]$.

As a result, following the general rules,[10] we find the thermodynamic potential density

$$\Phi = \frac{A}{2}\mathbf{L}_1^2 + \frac{B}{2}\mathbf{M}^2 + \frac{C}{2}(\mathbf{L}_2^2+\mathbf{L}_3^2) + \frac{D}{2}(\mathbf{L}_4^2+\mathbf{L}_5^2) + \frac{a}{2}L_{1z}^2 + \frac{b}{2}M_z^2 + \frac{c}{2}(L_{2z}^2+L_{3z}^2) + \frac{d}{2}(L_{4z}^2+L_{5z}^2) - \mathbf{MH}. \tag{8}$$

A potential similar to (8) has been written out by Vitebskiĭ *et al.*,[11] but they were interested in the antiferromagnetic-resonance spectrum of $CsMnBr_3$ in a field $H = 0$, and they omitted several relativistic terms.

For our problem, we can simplify the potential (8). In the first place, since we are interested in static properties, and since a state with $\mathbf{L}_4 = \mathbf{L}_5 = 0$ is realized in $CsMnBr_3$ according to neutron data,[3,5] we can omit these terms from (8). Second, since the exchange along the **z** axis is much stronger than all other interactions, we can conveniently single out pairs of nearest neighbors along this axis and introduce the new vectors

$$\mathbf{l}_i = \mathbf{M}_i - \mathbf{M}_{i+3}, \quad \mathbf{m}_i = \mathbf{M}_i + \mathbf{M}_{i+3}, \tag{9}$$

where $i = 1,2,3$. Using the conditions $\mathbf{m}_i \ll \mathbf{l}_i$, $(\mathbf{m}_i \mathbf{l}_i) = 0$, and $l_i^2 = l_0^2 - m_i^2$, where $l_0 = 2M_i$, retaining terms through second order in m_i/l_i, and considering the exchange interaction only between nearest neighbors, we can write potential (8) as follows, with an accuracy to within terms on the order of J_{ab}/J_c and $(H/J_c)^2$:

$$\Phi = \frac{F}{2}(m_1^2+m_2^2+m_3^2)$$
$$+Bl_0^2(-\cos\varphi_1\cos\varphi_2\cos\theta-\cos\varphi_1\cos\varphi_3\cos\theta$$
$$+\cos\varphi_2\cos\varphi_3\cos 2\theta+\sin\varphi_1\sin\varphi_2+\sin\varphi_1\sin\varphi_3$$
$$+\sin\varphi_2\sin\varphi_3)$$
$$+\frac{(a+4c)}{2}l_0^2(\sin\varphi_1^2+\sin\varphi_2^2+\sin\varphi_3^2)$$
$$+(a-2c)l_0^2(\sin\varphi_1\sin\varphi_2$$
$$+\sin\varphi_1\sin\varphi_3+\sin\varphi_2\sin\varphi_3)+\frac{b}{2}(m_1^2\sin\varphi_{1m}^2+m_2^2\sin\varphi_{2m}^2$$
$$+m_3^2\sin\varphi_{3m}^2)-H_\perp(m_1\cos\varphi_{1m}\sin\theta_{1m}+m_2\cos\varphi_{2m}\sin\theta_{2m}$$
$$+m_3\cos\varphi_{3m}\sin\theta_{3m})-H_\parallel(m_1\sin\varphi_{1m}+m_2\sin\varphi_{2m}+m_3\sin\varphi_{3m}).$$
(10)

In writing (10) we made use of the assertion (easily proved) that for one pair of sublattices ($\mathbf{M}_1$ and $\mathbf{M}_4$) the conditions $\mathbf{l}_1\perp\mathbf{H}$ and $\mathbf{l}_1\perp\mathbf{c}$ hold at arbitrary H. We also introduced the angles $\varphi_i=\varphi_{il}$, φ_{im}, which are azimuthal angles reckoned from the basal plane, and θ_{im}, which are tangential angles in the plane, reckoned from $\mathbf{l}_1$. The angle 2θ, between $\mathbf{l}_2$ and $\mathbf{l}_3$, is the same as the angle 2θ between $\mathbf{S}_2$ and $\mathbf{S}_3$, which was introduced above.

For a later comparison of our results with the conclusions of Ref. 7, we write equations relating the parameters of Φ and $\mathscr{H}$. The exchange parameters from (8), (10), and (1) are related by

$$F=-A+B-4C=\frac{24\mu J_c}{(g\mu_B)^2\rho N},$$
$$B=A-2C=\frac{18\mu J_{ab}}{(g\mu_B)^2\rho N}. \quad (11)$$

The incorporation of only the term $D\Sigma_i(S_i^z)^2$ which describes the magnetic anisotropy, in (1) is equivalent to imposing the following constraints on the parameters a, b, and c:

$$a=\frac{2\mu D}{3(g\mu_B)^2\rho N},\quad b=\frac{6\mu D}{(g\mu_B)^2\rho N},\quad c=\frac{\mu D}{(g\mu_B)^2\rho N}. \quad (12)$$

Working from (10), minimizing with respect to $\mathbf{m}_i$, we find

$$m_i=(H_\perp\cos\varphi_{im}\sin\theta_{im}+H_\parallel\sin\varphi_{im})/(F+b\sin^2\varphi_{im}). \quad (13)$$

The analysis below is limited to the case of interest here, $\mathbf{H}\perp\mathbf{c}$. The potential (10) can be simplified substantially by transforming to a coordinate system which is fixed in the plane passing through the triad of vectors $\mathbf{l}_i$ [within the accuracy of the derivation of (10), the vectors $\mathbf{l}_i$ lie in a common plane] and which makes an angle γ with the basal plane (Fig. 7). In this notation, the angular part of the potential Φ is

$$\Phi=-\frac{H^2}{2F}\left[1+2(\cos^2\theta+\sin^2\theta\sin^2\gamma)-\frac{b}{2F}\sin^4\theta\sin^2 2\gamma\right]$$
$$+Bl_0^2(\cos 2\theta-2\cos\theta)+6cl_0^2\sin^2\theta\sin^2\gamma. \quad (14)$$

From (14), minimizing Φ with respect to θ and γ, we easily find the phase diagram that we need. In the case $|b|\ll F$ and $b<0$ (this is the case in $CsMnBr_3$; the second of these conditions follows from $g_\perp<g_\parallel$), with $\mathbf{H}\perp\mathbf{c}$ and $H<\tilde{H}_c=Fl_0$, we obtain the following three phases (Fig. 8):

$$\gamma=0,\quad \cos\theta=[2-(H/H_c^2)]^{-1} \quad (15)$$

at $H<H_{c1}$, where $H_c^2=BFl_0^2$;

$$\cos 2\gamma=\left[1-\left(\frac{H}{H_c^*}\right)^2\right]\left[1-\left(2-\frac{6c}{B}\right)^{-2}\right]^{-1}\frac{F}{|b|},$$
$$\cos\theta=\left(2-\frac{6c}{B}\right)^{-1} \quad (16)$$

for $H_{c1}<H<H_{c2}$, where $H_c^*=(6cF)^{1/2}l_0$; and

$$H_{c1},\ H_{c2}=H_c^*\left\{1\mp\left[1-\left(2-\frac{6c}{B}\right)^{-2}\right]\frac{|b|}{F}\right\}^{1/2},$$
$$\gamma=\frac{\pi}{2},\quad \cos\theta=\left(2-\frac{6c}{B}\right)^{-1} \quad (17)$$

for $H_{c2}<H<\tilde{H}_c$. At the points of the phase transitions, H_{c1} and H_{c2}, the angles θ and γ vary continuously.

The magnetization of the crystal is given by

$$M_x=\frac{H}{F}\left(3-2\sin^2\theta\cos^2\gamma+\frac{|b|}{F}\sin^4\theta\sin^2\gamma\right). \quad (18)$$

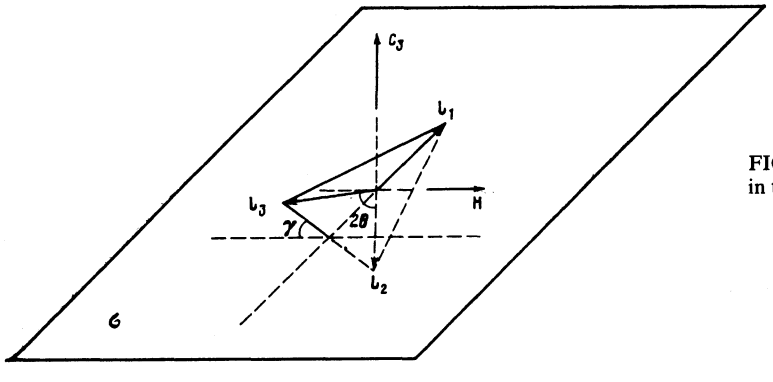

FIG. 7. Schematic diagram of the orientations of the spins in the angular phase.

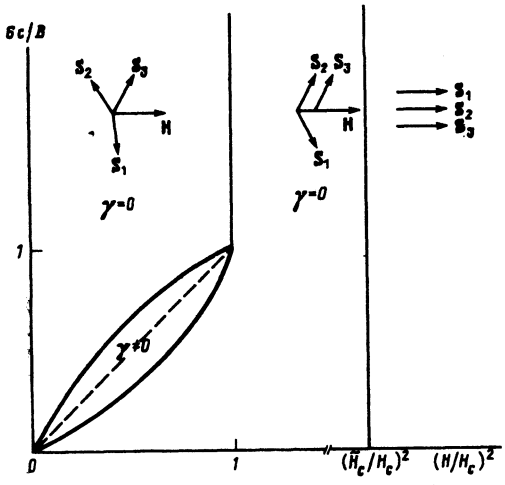

FIG. 8. Phase diagram of crystals of the $CsMnBr_3$ type in a field $\mathbf{H} \perp \mathbf{c}$ as calculated from the thermodynamic potential (8).

$$M_z = -\frac{H}{F}\sin^2\theta \sin 2\gamma \times \left[1 + \frac{|b|}{4F}\sin^4\theta \sin^2 2\gamma (\cos^2\theta + \sin^2\theta \sin^2\gamma)\right]. \quad (19)$$

These results hold at $T = 0$. As the temperature is raised, the critical fields change, and M_z decreases. This decrease can be taken into account by multiplying expressions (19) by $(1 - \chi_\parallel/\chi_\perp)$: $M_z \to 0$ as $T \to T_N$.

DISCUSSION OF RESULTS

As we mentioned earlier, the magnetization curves $M_x(H)$ which we measured are approximately the same as those predicted by Chubukov.[7] On the other hand, these experimental results suggest that in a certain interval of fields H the state of the magnetic system of the crystal corresponds to an angular phase with $M_z \neq 0$. The calculations above show that (first) the values of the critical fields H_c and $\tilde{H}_c$ and the condition $6c > B$, under which a state with $\gamma = 0$ persists in strong fields, which we calculated are the same as those calculated by Chubukov,[7] if we make use of the conversion coefficients in (11) and (12). Second, in the case $6c > B$, the calculated phase diagrams are also the same.

The results of the calculations are different for $0 < 6c < B$. In this case, one should observe two second-order phase transitions with increasing H, instead of the single first-order phase transition from a state with $\gamma = 0$ to a state with $\gamma = \pi/2$ which was predicted by Chubukov.[7] The reason is that our data reveal an angular phase with $0 < \gamma < \pi/2$ between these states. The existence of an angular phase is determined by the presence of a term bM_z^2 in Φ. The field interval in which this phase should be observed is

$$\Delta H = H_{c2} - H_{c1} = H_c \cdot \left[1 - \left(2 - \frac{6c}{B}\right)^{-2}\right] \frac{|b|}{F}. \quad (20)$$

In turn, the coefficient b describes an anisotropy of the magnetic susceptibility according to (12). This anisotropy also prevails in the paramagnetic region and has an anomalously large value in $CsMnBr_3$: $|b|$ amounts to $\sim 10\%$ of F. This property appears to be a general property of quasi-1D antiferromagnets.[1]

If we set $b = 0$, the angular phase disappears, according to our calculations, and a single phase transition should be observed at $H = H_c^*$. However, to second order the accuracy, with which the potential (8) is written, the potential Φ does not depend on the angle γ in a field $H \neq H_c^*$. In order to determine the order of this transition, we thus need to examine the higher-order terms.

If we wish to explain the presence of an angular phase in $CsMnBr_3$ we are thus obliged to assume that the condition $0 < 6c/B < 1$ holds in this crystal. The approximate agreement of the experimental dependence $M_x(H)$ with that calculated in Ref. 7, which we mentioned earlier, would then mean that $6c/B$ is approximately unity. In this case, at fields $H \geqslant H_{c1}$ the angle 2θ, between the spins $\mathbf{S}_2$ and $\mathbf{S}_3$, would be small, and the directions of the spins in the case $\gamma \neq 0$ would be slightly different from those which would prevail in the case $\gamma = 0$. This may be why an angular phase is not observed in neutron experiments.[2]

Unfortunately, this explanation of the experimental data does not answer the question of whether the relations among the anisotropy parameters and the exchange in the basal plane which have been found previously in neutron experiments[5] and from the antiferromagnetic-resonance spectrum[12] ($D > 3J_{ab}$) correspond to the relations which would be necessary, according to our calculations, for the existence of an angular phase ($6c < B$). The reason is that in the analysis of the experimental data in those earlier studies the expressions which were used for the magnon spectrum were calculated from the Hamiltonian (1). The most important step to take in order to resolve this question is thus to calculate the magnon spectrum in $CsMnBr_3$ on the basis of thermodynamic potential (8), containing all the second-order terms.

I am deeply indebted to L. A. Prozorova, A. V. Chubukov, and I. A. Zaliznyak for valuable discussions; D. V. Nikiforov for assistance in the measurements; and S. V. Petrov for furnishing the $CsMnBr_3$ single crystals.

[1] N. Achiva, J. Phys. Soc. Jpn. **27**, 561 (1969).
[2] B. D. Gaulin, T. E. Mason, and M. F. Collins, Phys. Rev. Lett. **62**, 1380 (1989).
[3] M. Eibschutz, R. C. Sherwood, F. S. L. Hsu, and D. E. Cox, *Proceedings of the Eighteenth Annual Conference on Magnetism and Magnetic Materials* (Denver, 1972), AIP Conf. Proc. No. 10, AIP, New York, 1973, Pt. 1, p. 684.
[4] J. Goodyear and D. J. Kennedy, Acta Crystallogr. Sect. B **28**, 1640 (1974).
[5] B. D. Gaulin, M. F. Collins, and W. J. L. Buyers, J. Appl. Phys. **61**, 3409 (1987).
[6] B. D. Gaulin and M. F. Collins, Can. J. Phys. **62**, 1132 (1984).
[7] A. V. Chubukov, J. Phys. C. **21**, L441 (1988).
[8] B. Ya. Kotyuzhanskiĭ and D. V. Nikiforov, J. Phys. Condensed Matter **3**, (1991).
[9] A.N. Bazhan, A. S. Borovik-Romanov, and N. M. Kreĭnes, Prib. Tekh. Eksp. No. 1, 213 (1973).
[10] G. Y. Lyubarskii, *Application of Group Theory in Physics*, Fizmatgiz, Moscow, 1958 (Pergamon, New York, 1960).
[11] I. M. Vitebskiĭ, V. L. Sobolev, and A. A. Chabanov, Fiz. Nizk. Temp. **16**, 1428 (1990) [Sov. J. Low Temp. Phys. **16**, 808 (1990)].
[12] I. A. Zaliznyak, L. A. Prozorova, and S. V. Petrov, Zh. Eksp. Teor. Fiz. **97**, 359 (1990) [Sov. Phys. JETP **70**, 203 (1990)].

Translated by D. Parsons