

# Investigation of static magnetic properties of antiferromagnetic $\text{RbMnCl}_3$

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By use of a vibration magnetometer designed to measure the magnetic moment along three mutually perpendicular directions, the static magnetic properties of an antiferromagnetic single crystal of  $\text{RbMnCl}_3$  have been investigated at  $T = 4.2$  K, over the range of magnetic fields from 0–60 kOe. In the paramagnetic state,  $\text{RbMnCl}_3$  has the hexagonal symmetry  $D_{6h}$ <sup>4)</sup>. The temperature of the transition to the antiferromagnetic state is  $T_N = 94 \pm 1$  K. Static magnetic measurements at  $T = 4.2$  K show that at  $H = 0$ , the single crystal splits into six equivalent domains with the antiferromagnetic vector  $\mathbf{L}$  oriented in the basal plane of the crystal along its binary axes. If a magnetic field  $\mathbf{H}$  is applied along a binary axis, then on increase of  $H$  there occurs a rotation of the antiferromagnetic vector  $\mathbf{L}$  to a direction perpendicular to  $\mathbf{H}$  in those domain regions where  $\mathbf{L}$  makes an angle  $60^\circ$  with the applied magnetic field  $\mathbf{H}$ , and an abrupt flip of  $\mathbf{L}$  to a position perpendicular to  $\mathbf{H}$ , at field  $H = H_c = 6.3$  kOe, in those domain regions where  $\mathbf{L} \parallel \mathbf{H}$ . When the magnetic field is oriented in the basal plane at angle  $30^\circ$  to a binary axis, there occurs a gradual rotation of the antiferromagnetic vector  $\mathbf{L}$  toward a direction perpendicular to  $\mathbf{H}$ . An investigation was made of the weak ferromagnetism  $\sigma_z$  that occurs along the  $c$  axis of the crystal for certain positions of the antiferromagnetic vector  $\mathbf{L}$  in its basal plane. The orientation of the antiferromagnetic vector  $\mathbf{L}$  in the basal plane was prescribed by an applied magnetic field  $|\mathbf{H}| > H_c$  in this plane. The values of the effective fields responsible for the magnetic properties of the given single crystal,  $H_E$ ,  $H_{AE}$ , and  $H_{D\perp}$ , were determined.

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According to neutron-diffraction data,<sup>[1]</sup> a monocrystal of  $\text{RbMnCl}_3$ , which in the paramagnetic state at room temperature has hexagonal symmetry with six magnetic ions in the elementary cell (Fig. 1), transforms to an antiferromagnetic state at  $T_N = 94.2$  K. At  $T < T_N$  the antiferromagnetic vector  $\mathbf{L}$  is oriented in the plane perpendicular to the  $c$  axis of the crystal. As was shown in Ref. 1, it follows from the neutron-diffraction data that in the paramagnetic state at room temperature, the  $c$  axis of the crystal is a sixfold axis; but at  $T = 4.2$  K, in the antiferromagnetic state, the neutron-diffraction data are described on the assumption that the symmetry of the monocrystal is lowered to orthorhombic symmetry, with a twofold  $c$  axis of,  $Cm'm'$  and  $Cmc'm$ , and with the remaining binary axes

turned by  $30^\circ$  with respect to one another in the basal plane of the crystal. When the antiferromagnetic vector  $\mathbf{L}$  is oriented in the basal plane of the crystal,<sup>[1]</sup> the existence of weak ferromagnetism along the  $c$  axis of the crystal is possible in  $\text{RbMnCl}_3$  for certain positions of the vector  $\mathbf{L}$ . Such a form of weak ferromagnetism, occurring along the  $c$  axis of the crystal, whose magnitude and sign depend substantially on the position of the antiferromagnetic vector  $\mathbf{L}$  in the basal plane, is similar to the weak ferromagnetism detected and investigated earlier along a trigonal axis  $C_3$  in the rhombohedral crystals  $\text{CoCO}_3$  and  $\text{NiCO}$ <sup>[2,3]</sup> and in hematite,  $\alpha\text{-Fe}_2\text{O}_3$ .<sup>[4]</sup> In an investigation of the magnetic properties of  $\text{RbMnCl}_3$  near  $T_N$  a weak ferromagnetism of this crystal along the  $c$  axis was detected.<sup>[5]</sup>

The aim of the present research was to investigate the anisotropy of the static magnetic properties of antiferromagnetic  $\text{RbMnCl}_3$  at  $T = 4.2$  K and at various orientations of the applied magnetic field  $\mathbf{H}$  with respect to the axes of the monocrystal, and to investigate the existence of, and determine, the weak ferromagnetism in it. The investigation was made with a vibrating-sample magnetometer<sup>[6]</sup> designed to measure the magnetic moment of a monocrystal along three mutually perpendicular directions with respect to the applied magnetic field  $\mathbf{H}$  and the crystal axes:  $M_{||}(H)$ ,  $M_{\perp}(H)$ , and  $M_{\perp}(H)$ .

By measuring three mutually perpendicular components of the magnetic moment  $\mathbf{M}$  of the specimen, one can investigate its magnitude and orientation with respect to the crystallographic directions, and also the variation of the magnitude and orientation of  $\mathbf{M}$  upon variation of the magnitude and orientation of the applied magnetic field  $\mathbf{H}$ .

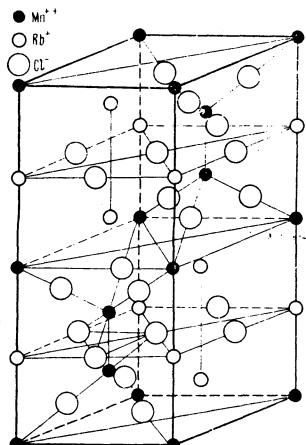


FIG. 1. Elementary cell of antiferromagnetic  $\text{RbMnCl}_3$ .

The experiments were performed on several monocrystals that were grown in the Physical Problems Institute, Academy of Sciences, USSR and in the L. V. Kirenskii Institute of Physics of the Siberian Branch, Academy of Sciences, USSR. The specimens were oriented with an accuracy of 1–2°.

## RESULTS OF THE MEASUREMENTS

Figure 2 shows the field dependence of the magnetic moment of the specimen measured along the applied magnetic field,  $M_{\parallel}(H)$ , when the magnetic field  $H$  was oriented in the basal plane of the crystal. Curve 1 in Fig. 2 represents the relation  $M_{\parallel}(H)$  obtained when  $H$  was oriented along one of the three binary axes of the crystal, Curve 2 the relation  $M_{\parallel}(H)$  when  $H$  was oriented at angle  $\psi = 30^\circ$  to binary axes of the crystal. From Fig. 2 (Curve 1) it is seen that when  $H$  is oriented along  $C_2$ , there is a quite abrupt jump on the  $M_{\parallel}(H)$  curve at magnetic field  $H = H_c = 6.3$  kOe. At  $H < H_c$ , on approach to  $H_c$  the  $M_{\parallel}(H)$  relation has a not very pronounced (of the order of 10%) nonlinear character, with increase of the slope of the magnetization curve; in weak fields  $H < 3$  to 4 kOe, the  $M_{\parallel}(H)$  relation is linear and is described by the expression

$$M_{\parallel}(H) = \chi^* H, \quad \chi^* = (0.8 \pm 0.1) \cdot 10^{-2} \text{ cgs emu/mol.}$$

At  $H > H_c$ , the relation is also linear and has the form

$$M_{\parallel}(H) = \chi_{\perp} H, \quad \chi_{\perp} = (1.6 \pm 0.1) \cdot 10^{-2} \text{ cgs emu/mol.}$$

When  $H$  is oriented at angle  $30^\circ$  to  $C_2$  (Curve 2), the  $M_{\parallel}(H)$  relation in the magnetic-field range  $0 < H < 15$  kOe is nonlinear, while for  $H > 15$  kOe it is linear and the  $M_{\parallel}(H)$  curve practically coincides with the  $M_{\parallel}(H)$  curve for  $H \parallel C_2$  (Curve 1). The  $M_{\parallel}(H)$  relations shown in Fig. 2 repeat, on change of the orientation of  $H$  in the plane perpendicular to the  $c$  axis, with period  $60^\circ$ .

Figure 3 shows the variation of the magnetic moment  $M_1^y(H)$  measured in the basal plane of the crystal perpendicular to an applied magnetic field in this plane. Curve 1 in Fig. 3a represents the relation  $M_1^y(H)$  when  $H$  is oriented along a binary axis  $C_2$ . It is seen that in this case  $M_1^y(H) \approx 0$  for all values of  $H$ . But if we change the orientation of  $H$  by an angle  $\psi = \pm 2^\circ$  with respect to

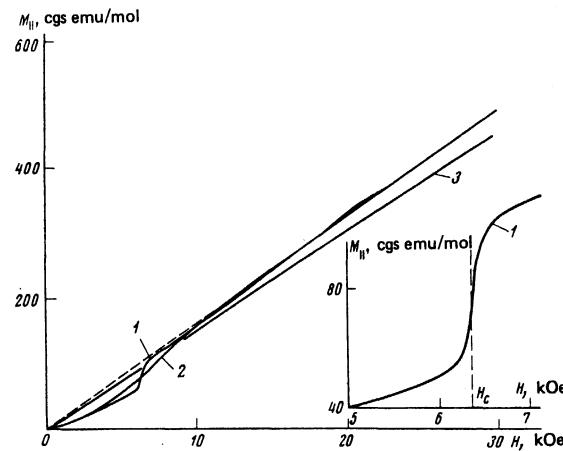


FIG. 2. Field dependence of the magnetic moment measured along the applied magnetic field  $H$ , for various orientations of  $H$ : 1,  $H \parallel C_2$ ; 2,  $\psi(H, C_2) = 30^\circ$ ; 3,  $H \parallel c$ .

the binary axis, then at  $H = H_c = 6.3$  kOe, when the  $M_{\parallel}(H)$  relation experiences a jump, there appears along the direction of the  $y$  axis a magnetic moment that thereafter again approaches zero: the  $M_1^y(H)$  relation takes the form represented by the curves  $\psi = \pm 2^\circ$  in Fig. 3a. On further change of the orientation of  $H$  (increase of  $|\psi|$ ) in the basal plane, the  $M_1^y(H)$  relation for the magnetic moment that appears changes as is shown in Fig. 3a by the curves  $\psi = \pm 15^\circ$ . Thus with increase of the angle  $\psi$  between the axis  $C_2$  and the magnetic field  $H$ , the appearance of a magnetic moment  $L_1(H)$  occurs over a wider interval of magnetic fields in the vicinity of  $H_c$ , while the value of  $M_1^y(H)$  at  $H = H_c$  decreases. On approach of  $\psi$  to  $30^\circ$ , the magnetic moment  $M_1^y$  appears starting with very weak magnetic fields (Fig. 3b) but decreases in magnitude; and at  $\psi = 30^\circ$ ,  $M_1^y(H) \approx 0$ . When  $H > 20$ – $25$  kOe,  $M_1^y(H) \approx 0$  at an arbitrary value of the angle  $\psi$ . The relations described repeat, on change of the direction of  $H$ , with period  $60^\circ$ .

Figure 4 gives the relations for  $M_1^z(H)$ , measured along the  $c$  axis of the crystal when the magnetic field  $H$  is oriented in the plane perpendicular to this axis. Curve 1 represents the  $M_1^z(H)$  relation in a magnetic field oriented along one of the three binary axes of the

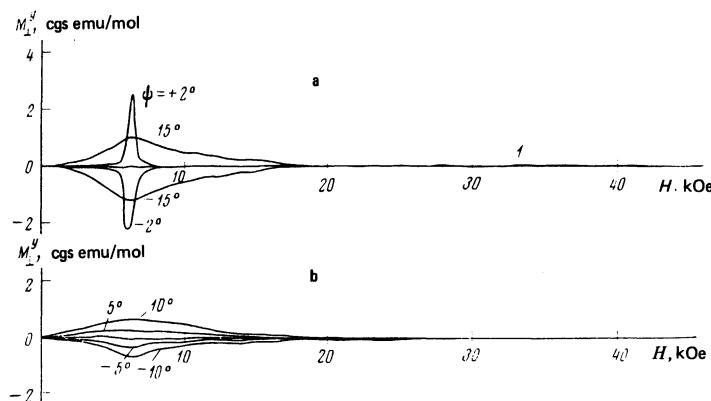


FIG. 3. Field dependence of the magnetic moment measured perpendicular to the applied magnetic field  $H$  in the plane perpendicular to the  $c$  axis of the crystal, for various orientations of  $H$ : a,  $H \parallel C_2 \pm \psi$ ; b,  $H \parallel C_2 + 30^\circ \pm \psi$ .

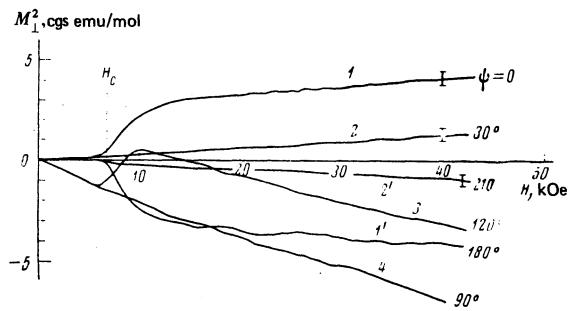


FIG. 4. Field dependence of the magnetic moment measured perpendicular to the applied magnetic field and along the  $c$  axis of the crystal, for various orientations of  $H$  in the basal plane.

crystal. Curve 2 represent the  $M_1^z(H)$  relation when  $H$  is oriented at angle  $30^\circ$  to these axes.

It is seen from the figure that when the magnetic field is oriented along a binary axis, the value of  $M_1^z(H)$  is nearly zero for  $0 < H < H_c = 6.3$  kOe. The presence of a small signal is due to inexact orientation of  $H$  with respect to the basal plane of the crystal. For  $H > H_c$  (Curve 1), there appears a nonvanishing magnetic moment  $M_1^z(H)$ , which has a nonlinear character in the field range  $H_c < H < 15$  kOe, but which for  $H > 15$  kOe can be described by the expression  $M_1^z(H) = \sigma_z + \chi' H$ . On change of the orientation of  $H$  by  $180^\circ$ , the signs of  $\sigma_z$  and of  $\chi' H$  reverse (Curve 1').

When  $H$  is oriented at angle  $30^\circ$  to a binary axis (Curve 2), the  $M_1^z(H)$  relation has the form  $M_1^z(H) = \chi' H$ . The appearance of a magnetic moment  $\chi' H$  is due to inexact orientation of  $H$  with respect to the basal plane of the crystal; there is a component  $h_z$  of the magnetic field  $H$  and consequently, also, a component  $\chi_z h_z$  of the magnetic moment along the measurement axis  $z$ , having  $180^\circ$  anisotropy. This relation can be easily taken into account, as was done in Ref. 3, by comparing the  $M_{||}(H)$  (Fig. 2) and  $M_1^z(H)$  relations for the same orientations of  $H$  in the basal plane of the crystal and estimating the amount of the inaccuracy in orientation of  $H$ .

At intermediate angles  $\psi$ , the  $M_1^z(H)$  relations have the form of Curves 3 and 4 of Fig. 4. The latter were obtained with an appreciable component  $h$  of the magnetic field  $H$  along the  $c$  axis.

Figure 5 shows the variation of the value of the magnetic moment  $\sigma_z$  with the angle  $\psi$  between the applied magnetic field  $H$  and a binary axis  $C_2$  of the crystal. The value of  $\sigma_z(\psi)$  was determined by extrapolation of the  $M_1^z(H)$  relation in strong fields to  $H = 0$ . Curve 1 in Fig. 5 was obtained by processing of the experimental curves shown in Fig. 4, with allowance for the orientation of the magnetic field  $H$  with respect to the basal plane.<sup>[3]</sup> It is seen from Fig. 5 that the experimental relation  $\sigma_z(\psi)$  is close to a function  $\sigma_z(\psi)$  of the form

$$\sigma_z(\psi) = \sigma_z^0 |\sin 3\psi|$$

for  $30^\circ < \psi < 210^\circ$  and

$$\sigma_z(\psi) = -\sigma_z^0 |\sin 3\psi|$$

for  $210^\circ < \psi < 390^\circ$ , where  $\sigma_z^0 = 2.6 \pm 0.2$  cgs emu/mol.

Figure 2 shows also (Curve 3) the variation of the magnetic moment measured along the magnetic field  $H$ , when the magnetic field  $H$  is oriented along the  $c$  axis (perpendicular to the basal plane). In magnetic fields  $0 < H < 30$  kOe, this relation is determined by the expression

$$M_{||}(H) = \chi_{||} H, \quad \chi_{||} = (1.5 \pm 0.1) \cdot 10^{-2} \text{ cgs emu/mol.}$$

In magnetic fields  $H > 30$  kOe, a change of slope of the magnetization curve  $M_{||}(H)$  is observed (it decreases), but this change amounts to a quantity of order 5%.

## DISCUSSION OF RESULTS

According to neutron-diffraction data, the antiferromagnetic vector  $L$  in  $\text{RbMnCl}_3$ , in the absence of a magnetic field, is oriented in the basal plane of the crystal. The experiments (see Figs. 2 and 3), in which a flip of  $L$  is observed when  $H$  is oriented in this same plane along one of the three binary axes, can be explained

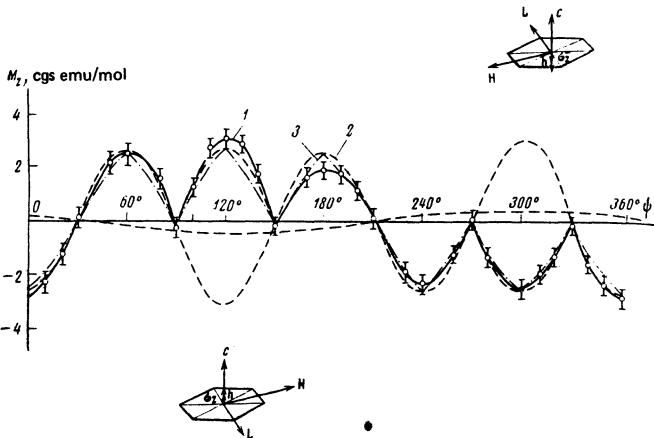


FIG. 5. Variation of the ferromagnetic moment, which appears along the  $c$  axis of the crystal at a magnetic field  $H > H_c$ , with orientation of the applied magnetic field in the basal plane.

qualitatively by supposing that in the absence of a magnetic field, the monocrystal splits into six equivalent domains, in which the antiferromagnetic vector  $\mathbf{L}$  is oriented in the basal plane of the crystal along the six directions of the binary axes of the crystal.

When a magnetic field  $H$  is applied along a binary axis, it is directed along the antiferromagnetic vector  $\mathbf{L}$  for one third of the crystal and at angle  $60^\circ$  to the vector  $\mathbf{L}$  in the rest of it. In a magnetic field  $H_c$  there occurs a flip of the antiferromagnetic vector  $\mathbf{L}$  from a position with  $\mathbf{L} \parallel \mathbf{H}$  to a position with  $\mathbf{L} \perp \mathbf{H}$ ; an abrupt jump occurs on the  $M_{\parallel}(H)$  curve, and at  $H = H_c$  a nonvanishing magnetic moment  $M_1^y(H)$  appears. In the part of the crystal in which  $\mathbf{L}$  is at angle  $60^\circ$  to  $\mathbf{H}$ , there occurs a continuous rotation of  $\mathbf{L}$  toward a direction perpendicular to the applied magnetic field  $\mathbf{H}$ . Thus there is nonlinearity of the  $M_{\parallel}(H)$  curve and a nonvanishing  $M_1^y(H)$ .

In regard to the results of measurement of  $M_1^y(H)$  represented in Fig. 3, it must be noted that for exact orientation of  $\mathbf{H}$  along a binary axis  $C_2$ ,  $M_1^y(H) \approx 0$  because of the fact that the crystal splits into two equivalent domains, with a symmetric distribution of the antiferromagnetic vector  $\mathbf{L}$  with respect to the applied magnetic field  $\mathbf{H}$ . The flip of the antiferromagnetic vector  $\mathbf{L}$  from a position  $\mathbf{L} \parallel \mathbf{H}$  to a position  $\mathbf{L} \perp \mathbf{H}$  at  $H = H_c$  occurs in two equivalent directions with respect to  $\mathbf{H}$ ; therefore in this case also,  $M_1^y(H) = 0$ . But if  $\mathbf{H}$  is oriented at angle  $\psi \approx 2^\circ$  to a binary axis, the crystal is in a single-domain state, and the flip of  $\mathbf{L}$  occurs in a distinct direction; then a signal  $M_1^y(H) \neq 0$  occurs.

The repetition of the magnetization curves  $M_1^y(H)$  and  $M_{\parallel}(H)$  after  $60^\circ$  confirms the conclusion that at  $H = 0$  the specimen splits into domains in which the antiferromagnetic vector  $\mathbf{L}$  is oriented along one of the six possible directions along binary axes of the crystal.

When  $\mathbf{H}$  is oriented at angle  $30^\circ$  to binary axes, the antiferromagnetic vector  $\mathbf{L}$  is perpendicular to  $\mathbf{H}$  in one third of the crystal volume; in the remaining part, where in weak fields  $\mathbf{L}$  is oriented at angle  $30^\circ$  to  $\mathbf{H}$ , a continuous rotation of  $\mathbf{L}$  occurs with increase of  $\mathbf{H}$ . There is a nonlinear  $M_{\parallel}(H)$  relation and a nonvanishing  $M_1^y(H)$  (if, as in the first case,  $\mathbf{H}$  is oriented at angle  $30^\circ \pm \psi$  to an axis  $C_2$ ).

In order to describe quantitatively the rotation of the anti-ferromagnetic vector  $\mathbf{L}$ , we must consider the form of the thermodynamic potential  $\Phi$ , which describes the magnetic properties of the monocrystal under investigation. For crystals of hexagonal symmetry we have

$$\begin{aligned} \Phi = & \frac{1}{2}Bm^2 + \frac{1}{2}D(\gamma_m)^2 + \frac{1}{2}\alpha\gamma_x^2 + \frac{1}{2}bm_x^2 \\ & - \frac{1}{2}d[(\gamma_x + i\gamma_y)^2 + (\gamma_x - i\gamma_y)^2]m_z \\ & + e[(\gamma_x + i\gamma_y)^6 + (\gamma_x - i\gamma_y)^6] - mH \end{aligned} \quad (1)$$

where the terms  $\frac{1}{2}B \cdot m^2$  and  $\frac{1}{2}D(\gamma \cdot m)^2$  correspond to exchange interaction, while  $\frac{1}{2}\alpha\gamma_x^2$  and  $\frac{1}{2}bm_x^2$  are relativistic interactions that determine the position of the antiferromagnetic vector  $\mathbf{L}$  with respect to the hexagonal crystal axis and the anisotropy of the magnetic

susceptibility for  $\mathbf{H} \perp c$  and  $\mathbf{H} \parallel c$ . The invariant

$$\frac{1}{2}d[(\gamma_x + i\gamma_y)^2 + (\gamma_x - i\gamma_y)^2]m_z$$

causes weak ferromagnetism along the hexagonal axis of the crystal for certain positions of the antiferromagnetic vector  $\mathbf{L}$  in the basal plane of the crystal; the invariant

$$\frac{1}{2}e[(\gamma_x + i\gamma_y)^6 + (\gamma_x - i\gamma_y)^6]$$

causes anisotropy of the magnetic properties in the basal plane of the crystal. By minimizing the thermodynamic potential (1), we can obtain the equations of rotation of the antiferromagnetic vector  $\mathbf{L}$  on application of a magnetic field  $\mathbf{H}$  and the expressions  $M_{\parallel}(H)$ ,  $M_1^y(H)$ , and  $M_1^x(H)$  for the magnetic domains.

But as was pointed out above, the neutron-diffraction experiments<sup>[1]</sup> in the antiferromagnetic phase can be explained by supposing that  $\text{RbMnCl}_3$  consists of three orthorhombic domains whose axes are spaced at angle  $60^\circ$ . The symmetry of these domains is either  $Cm'c'm$  or  $Cmcm$ . Weak ferromagnetism along the  $c$  axis of the crystal is allowed in the symmetry  $Cm'c'm$  but not in the symmetry  $Cmcm$ . In static magnetic measurements of the magnetic moment, what are measured are the magnetization curves  $M_{\parallel}(H)$  and  $M_1^y(H)$  of the whole crystal, which, just as in the case of hexagonal symmetry, have period  $60^\circ$ ; but the thermodynamic potential in the form  $\Phi$  is written for orthorhombic crystals of one of the domains.

In the present paper, in order to obtain the values of the effective fields that produce the magnetic properties of the given monocrystal, we shall consider magnetization curves for application of the magnetic field  $\mathbf{H}$  in rational directions: along a binary axis  $C_2$  in the basal plane of the crystal (along the antiferromagnetic vector  $\mathbf{L}$ ), at angle  $30^\circ$  to a binary axis in the basal plane of the crystal, and along the  $c$  axis perpendicular to the basal plane of the crystal (Figs. 2–4); that is, we shall consider the case of an easy-axis crystal, when in one of the three domains with different orientation of  $\mathbf{L}$  the antiferromagnetic vector  $\mathbf{L} \parallel \mathbf{H}$ . On change of the orientation of  $\mathbf{H}$ , this situation occurs in the crystal with period  $60^\circ$ .

The effective exchange-interaction field  $H_E = M_0/\chi_1$  for  $\text{RbMnCl}_3$  is determined from the experiments whose data are represented in Fig. 2 for  $H \gg H_c$ , when the magnetic moment  $M_{\parallel}(H)$  is described by the expression  $M_{\parallel}(H) = \chi_1 H$  for arbitrary orientation of  $\mathbf{H}$  in the basal plane of the crystal; that is, when the antiferromagnetic vector  $\mathbf{L}$  is perpendicular to  $\mathbf{H}$ : thus  $H_E = 860 \pm 40$  kOe.

The effective anisotropy field  $H_A = M_0 b$ , determined from the anisotropy of the magnetic susceptibility  $\chi_1$  for  $\mathbf{H} \perp c$  (Curve 1 of Fig. 2) and  $\chi_1^*$  for  $\mathbf{H} \parallel c$  (Curve 3 of Fig. 2), has the value  $H_A = 60 \pm 10$  kOe.

The effective anisotropy field in the basal plane of the crystal is determined from the experiment (Fig. 2, Curve 1) in which the magnetic field  $\mathbf{H} \parallel C_2$ ; that is, the applied magnetic field is parallel to the antiferro-

magnetic L in one of three domains of the crystal. A theoretical calculation, carried out by Borovik-Romanov<sup>[7]</sup> for a uniaxial crystal, gives an expression for the magnetic field  $H_c$  in the form  $H_c = \sqrt{Be}$ , where  $B$  is the exchange field in the crystal and  $e$  is the effective uniaxial-anisotropy field. Here  $H_c^2 = H_{AE}^2$ . The value obtained for the magnetic field  $H_c$  at which the antiferromagnetic vector L flips from a position with  $L \parallel H$  to a position with  $L \perp H$  is  $H_c = H_{AE} = 6.3 \pm 0.1$  kOe. The effective field responsible for the weak ferromagnetism  $\sigma_z$  along the  $c$  axis of the crystal, defined as  $H_D = \sigma_z / \chi_1$ , is  $H_D = 0.16 \pm 0.02$  kOe. A calculation carried out in Ref. 7 for the case in which, in a uniaxial crystal, there is a Dzyaloshinskii interaction  $H_{D\perp}$ , gives the relation  $H_c^2 = H_{AE}^2 - H_{D\perp}^2$ ; but in our case the numerical value of  $H_{D\perp}$  is small, and does not change the value of  $H_{AE}$  very much.

We turn now to a discussion of the experiment whose data are presented in Fig. 5. From this experiment it can be concluded that when the orientation of a magnetic field  $H > H_c$  in the basal plane is varied, so that a variation occurs in the orientation of  $L \perp H$  in this plane, for certain positions of L there occurs a weak ferromagnetism  $\sigma_z$  along the  $c$  axis of the crystal, whose variation can be described with sufficient accuracy by the expression  $\sigma_z = \sigma_z^0 |\sin 3\psi|$  (Curve 2 in Fig. 5).

The fact that the  $M_1^z(H)$  relation has a form close to  $\sigma_z = \sigma_z^0 |\sin 3\psi|$  (Curve 1 in Fig. 5) is explained by the fact that for an exact orientation  $H \perp c$  and for  $H > H_c$ , the monocrystal splits into two domains with ferromagnetic moments oriented along the  $c$  axis in opposite senses, since there is no distinguished direction of L in the plane. But if the magnetic field H is inclined at some angle  $\alpha$  to the basal plane, so that there is a component h of the magnetic field along  $c$ , there occurs a remagnetization of the crystal to a state with  $\sigma_z \parallel h$ . On variation of the orientation of H in the basal plane of the crystal, the magnetic field h varies according to the law  $h = h_0 \sin \psi$ ; in the experiment corresponding to Fig. 4, the angle  $\alpha$  was so chosen that H was oriented in the basal plane at angle  $\psi = 30^\circ$  and  $210^\circ$  to an axis  $C_2$ . Then in the angular range  $30^\circ < \psi < 210^\circ$  the monocrystal remagnetizes to a single-domain state with  $\sigma_z \parallel h \parallel c$ , whereas in the angular range  $210^\circ < \psi < 390^\circ$  the monocrystal remagnetizes to a single-domain state with  $-\sigma_z \parallel -h \parallel c$ .

In order to explain the variations, shown in Figs. 4 and 5, of the magnetic moment  $\sigma_z(\psi)$  that appears along the  $c$  axis, we must suppose that the magnetic moment  $\sigma_z$  that appears in the given magnetic fields is due to flipping of the antiferromagnetic vector L in that domain region of the crystal in which, for  $H < H_c$ ,  $L \parallel H$ . But in those domain regions where, for  $H \ll H_c$ , the vector L is at  $60^\circ$  to the applied magnetic field, and where increase of H leads to a continuous rotation, we must suppose that they rotate in opposite directions. Then the magnetic moments  $\sigma_z$  that appear along the axis are also oriented in opposite directions, and as a result they are mutually cancelled, that is, in these domain regions, no  $\sigma_z$  appears in the given magnetic

fields in the experiment described by Figs. 4 and 5. In order to remagnetize the specimen completely with  $\sigma_z \parallel c$ , it is necessary to apply a sufficiently strong magnetic field along the  $c$  axis. Thus in the experiments described by Figs. 4 and 5, remagnetization of the magnetic moment  $\sigma_z \parallel c$  occurs in one third of the crystal, by flip of L. The fact that the direction of flip of the vector L can be fixed is shown by the experiments on measurement of  $M_1^z(H)$  for  $\psi = \pm 2^\circ$  (Fig. 3). If  $\psi = 0$ , then  $M_1^z(H) \approx 0$ , and the flip of the vector L at  $H = H_c$  occurs in opposite directions; but if the magnetic field H is directed at angle  $\psi = \pm 2^\circ$  to the axis  $C_2$ , then the flip of L in a domain is fixed, and at  $H \approx H_c$  a magnetic moment  $M_1^z(H)$  appears. Orientation of the magnetic field H near  $C_2$  cannot fix the rotation of L in those domain regions where H makes a  $60^\circ$  angle with L. From the experimental data represented in Figs. 4 and 5 it may be concluded that when the orientation of the magnetic field in the basal plane is varied, and when H approaches one of the three  $C_2$ , creation of  $\sigma_z$  occurs in that domain region where there occurs a flip of the antiferromagnetic vector L associated with this axis; then a  $60^\circ$  period is observed in Fig. 5.

As a result of an investigation of AFMR in this material by Prozorova and Andrienko,<sup>[8]</sup> it was suggested that  $\text{RbMnCl}_3$  at helium temperatures consists of three crystallographic domains of orthorhombic symmetry, whose axes are arranged according to the neutron-diffraction investigations.<sup>[1]</sup> The ferromagnetic moment  $\sigma_z$  along the  $c$  axis then owes its origin to the second-order invariants in the expansion of the thermodynamic potential  $\Phi$  in the antiferromagnetic and ferromagnetic moments L and m. In this case the angular variation of the moment  $\sigma_z$  is expressed in the form

$$\sigma_z(\psi) = 2\sigma_z^0 \sin \psi$$

for  $0 + \frac{1}{3}\pi k < \psi < \frac{1}{6}\pi + \frac{1}{3}\pi k$  and

$$\sigma_z(\psi) = -2\sigma_z^0 \sin \psi$$

for  $\frac{1}{6}\pi + \frac{1}{3}\pi k < \psi < \frac{1}{3}\pi + \frac{1}{3}\pi k$  (Curve 3 of Fig. 5), as also follows from the discussion given above.

As is seen from Fig. 5, theoretical interpretation of the experiment on the determination of the angular variation of the ferromagnetic moment  $\sigma_z(\psi)$  is rendered difficult by the fact that the relations

$$\sigma_z(\psi) \approx \sigma_z^0 |\sin 3\psi|$$

in the case of hexagonal crystal symmetry and

$$\sigma_z(\psi) \approx 2\sigma_z^0 \sin \psi$$

in the case of three orthorhombic symmetries are close to each other. But there is no doubt about the occurrence of a ferromagnetic moment.

Thus it has been observed that in  $\text{RbMnCl}_3$  the antiferromagnetic vector L in the absence of a magnetic field is oriented along the binary axes in the basal plane of the crystal. When a magnetic field H is applied, increase of H leads to a discontinuous flip of L

at field  $H=H_c = 6.3$  kOe in a domain where, for  $H \ll H_c$ ,  $L \parallel H$ ; and to a continuous rotation of  $L$  in domains where  $L$  was oriented at angle  $60^\circ$  to  $H$ . In strong magnetic fields,  $H \gg H_c$ , independently of the orientation of  $H$  in the basal plane,  $L \perp H$ . On change of the orientation of the antiferromagnetic  $L$  in the basal plane of the crystal, for certain positions of  $L$  in this plane there appears a ferromagnetic moment  $\sigma_z$  along the  $c$  axis; its variation can be described with sufficient accuracy by the expression  $\sigma_z = \sigma_z^0 \sin 3\psi$ . In this work we have determined the values of the effective fields responsible for the magnetic properties of this monocrystal.

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## Spin-phonon mechanism of magnetic ordering in paramagnetic crystals

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The quantum field theory methods are applied to a phase transition in a system of paramagnetic ions interacting with acoustic phonons. It is shown that when a threshold condition, imposed on an external static magnetic field and paramagnetic ion density, is exceeded, a paramagnetic crystal has a critical temperature  $T_c$  below which it experiences spontaneous static deformation accompanied by a spontaneous magnetic ordering of the  $\langle S_x S_y \rangle \neq 0$  type. The resonant response of the spin-phonon system to an external rf field and to sound is determined in the range  $T \geq T_c$  and one of the resonance modes is found to be soft. The phase transition in question is not exhibited by a crystal with a macroscopically inhomogeneous distribution of paramagnetic ions.

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### INTRODUCTION

Investigations of a paramagnetic system interacting with a radiation field in a resonator have shown that a phase transition may occur when the density of paramagnetic ions and an external static magnetic field exceed certain critical values.<sup>[1]</sup> The ordered thermody-

namic phase is characterized by a spontaneous magnetic order directed at right-angles to the external field and by a static component of the radiation field.

It is of interest to consider an analogous problem of a phase transition in a spin-phonon system. The new thermodynamic phase should be manifested by magnetic