tural transition in which ferromagnetic properties appear in the system.

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Magnetic phase transition from the antiferromagnetic to the paramagnetic state in $NaMnCl_3$

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The magnetic properties of a single crystal of antiferromagnetic $NaMnCl_3$ have been investigated with a vibrating-sample magnetometer in magnetic fields up to 65 kOe, over the temperature range 2 to 25 K. It is shown that in magnetic fields H = 15kOe a phase transition of the second kind, from the antiferromagnetic state to the paramagnetic, occurs in $NaMnCl_3$ at a temperature corresponding to the measurements. The features of this transition that are manifested on the magnetization curve of the single crystal are investigated.

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Sodium manganese chloride, $NaMnCl_3$, crystallizes in the hexagonal syngony, space group $R\bar{3}$. The parameters of the hexagonal cell at room temperature are: $a = 6.591$ Å, $c = 18.627$ Å.[1] The structure of NaMnCl consists of alternating layers of Mn^{++} and Na^+ ions separated by layers of Cl^- ions (Fig. 1). The distance between neighboring Mn^{++} ions located in a single plane is less by a factor 1.63 than the distance between Mn^{++} ions in neighboring planes. The elementary cell of $NaMnCl_3$ contains two magnetic ions.

It has been discovered[2,3] that $NaMnCl_3$ is antiferromagnetic, with Néel temperature $T_N = 7.1$K. Interest in further investigation of this crystal was determined by the possibility of studying a magnetic phase transition produced by application of a magnetic field, and due to destruction of the antiferromagnetic structure at a critical field, since from estimates obtained in Ref. 3 the exchange field is $H_E \sim 13$ kOe. According to Ref. 3, sodium manganese chloride belongs to the quasi-two-dimensional class of antiferromagnets.

$NaMnCl_3$ monocrystals for the investigations were grown by B. V. Beznosikov at the L. V. Kirenskiĭ Institute of Physics, Siberian Division, Academy of Sciences, USSR.2) X-ray investigations made on the $NaMnCl_3$ at room temperature showed agreement of the cell parameters with those given above and absence of paramagnetic impurities, within the limits of accuracy of the method. Investigations of the $NaMnCl_3$ monocrystals were made with a vibrating-sample magnetometer[4] over the magnetic-field range 0 to 65 kOe and the temperature range 2 to 20 K. Temperatures below $T = 4.2$ K were reached by evacuating helium from a cryostat. The orientation of the crystal axes of the

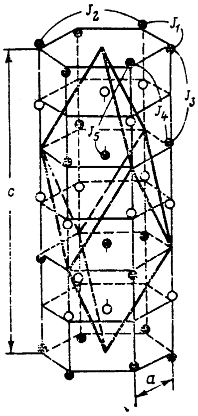

FIG. 1. Elementary cell of $NaMnCl_3$. Light points, Na^{1+}; dark points, Mn^{2+}.

monocrystals under investigation with respect to the applied magnetic field H and to the axes of the measuring coils was no worse than 2 – 3°.

RESULTS OF THE MEASUREMENTS

Figure 2 shows the variation of the magnetic moment M of a monocrystal of $NaMnCl_3$ with the applied magnetic field H at $T=4.2$ K. One curve represents the function $M(H)$ when the magnetic field H is applied perpendicular to the high-order c axis, $H\perp c$; the other curve represents the function $M(H)$ when the magnetic field is oriented along the c axis, $H \parallel c$. It is evident from Fig. 2 that at magnetic field $H=H_c=15$kOe, for both directions of the applied magnetic field H, there occurs in $NaMnCl_3$ a phase transition that is characterized by an abrupt change of form of the magnetization curve $M(H)$.

When the magnetic field is applied in the basal plane, $H\perp c$ (Fig. 2), in magnetic fields $0<H<8$ kOe the magnetic-moment dependence $M(H)$ is linear and is described by the expression

$$M(H)=\chi_\perp H,\quad \chi_\perp=1.1\pm0.5 \text{ cgs emu/mol.}$$

In magnetic fields $8<H<14$ kOe (near H_c), a nonlinear growth of the magnetic-moment dependence $M(H)$ occurs with increase of the magnetic moment. For magnetic field $H>H_c$, the magnetic-moment dependence asymptotically approaches the value of the saturation magnetic moment $2M_0=28\,000$ cgs emu/mol of the two magnetic Mn^{++} ions in the elementary cell. The form of the magnetization curves $M(H)$ does not change with change of the orientation of the applied magnetic field in the basal plane ($H\perp c$).

When the magnetic field is oriented along the c axis, $H\parallel c$, in magnetic fields $0<H<8$ kOe there is again a linear magnetic-moment dependence

$$M(H)=\chi_\perp^* H,\quad \chi_\perp^*=0.94\pm0.5 \text{ cgs emu/mol.}$$

in magnetic fields $8<H<H_c^*=15.6$ kOe (near H_c^*), a nonlinear growth of the $M(H)$ dependence again occurs; and for magnetic fields $H>H_c$, the $M(H)$ dependence again approaches asymptotically the value of the saturation magnetic moment $2M_0$ of the two magnetic Mn^{++} ions in the elementary cell.[3] The values of the magnetic susceptibilities $\chi_\perp$ and $\chi_\perp^*$ obtained in the present work agree to within 10% accuracy with the values obtained in Ref. 3.

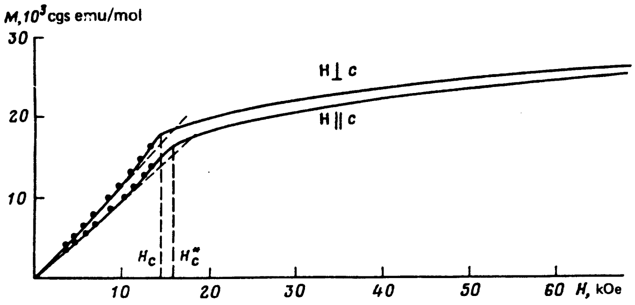

FIG. 2. Variation of magnetic moment with applied magnetic field for different orientations of H.

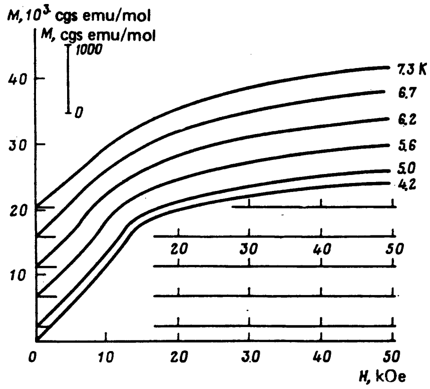

FIG. 3. Variation of magnetic moment with applied magnetic field at various temperatures.

Figure 3 shows the magnetic-moment relations $M(H)$ for the magnetic-field orientation $H\perp c$ at various temperatures. The investigations of the magnetization curves of $NaMnCl_3$ were made over the temperature range 2 to 20 K (Fig. 3 shows the $M(H)$ relations over the temperature range 4.2 to 7.3 K). It is evident from Fig. 3 that on change of temperature the general form of the magnetization curves $M(H)$ remains unchanged to temperatures of the order of 6 K; change occurs only in the values of the phase-transition magnetic field $H_c^*(T)$ and of the magnetic susceptibility $\chi_\perp^*(T)$. At temperatures greater than 7 K, the nonlinearity of the magnetic-moment dependence $M(H)$ in strong magnetic fields, $H>10$ kOe, remains, but the nonlinearity in $M(H)$ for $H<10$ kOe is not observed.

With the magnetic-field orientation $H\parallel c$, the magnetic-moment relations $M(H)$ at various temperatures are similar to the $M(H)$ relations of Fig. 3, with appropriate values of the phase-transition magnetic fields $H_c^*(T)$ and of the magnetic susceptibilities $\chi_\perp^*(T)$.

Figures 4 and 5 show the temperature dependences of the phase-transition magnetic field $H_c(T)$ (Fig. 4) and of the magnetic susceptibility $\chi_\perp(T)$ at $H\ll H_c$ (Fig. 5) for the different orientations of the applied magnetic

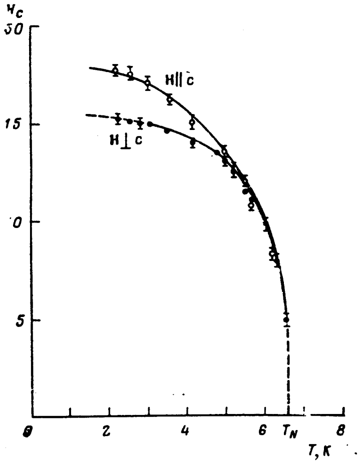

FIG. 4. Variation of phase-transition magnetic field with temperature.

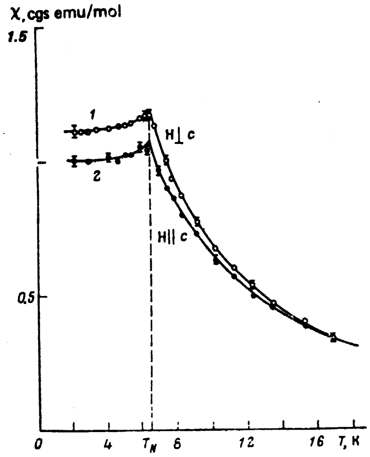

FIG. 5. Variation of magnetic susceptibility of $NaMnCl_3$ with temperature.

field H. Curves 1 in Figs. 4 and 5 represent the $H_c(T)$ and $\chi_\perp(T)$ relations for the magnetic-field orientation $H_\perp c$, curves 2 for $H \parallel c$.

The phase-transition magnetic field $H_c(T)$ at various temperatures $T < T_N$ was determined by the point of inflection on the magnetization curve $M(H)$, where a change of sign of the second derivative $\partial^2 M/\partial H^2$ occurs. The values of the magnetic susceptibility were determined from the linear section of the magnetic-moment relation $M(H)$ at magnetic field $H \ll H_c$. It is seen from Fig. 5 that the $\chi_\perp(T)$ and $\chi_\perp^*(T)$ relations have a small maximum near T_N but remain practically constant at temperatures $T < T_N$.

DISCUSSION OF RESULTS

From analysis of the temperature dependence of the magnetic susceptibility shown in Fig. 4, it follows that sodium manganese chloride is antiferromagnetic, with Néel temperature $T_N = 6.5 \pm 0.5$ K; this is in good agreement with the results of Ref. 3. In Ref. 3, somewhat different transition temperatures are given for different crystallographic directions. This is explained by the fact that the measurements of the susceptibility $\chi(T)$ were made at a single field $H = 5$ kOe, and in the processing of the results no account was taken of the nonlinearity of the magnetization curves $M(H)$ near the phase-transition point H_c.

From the experiments whose results are presented in Fig. 2, it may be concluded that when $T = 4.2$ K, $NaMnCl_3$ undergoes at magnetic field $H \sim 15$ kOe a phase transition of the second kind from the antiferromagnetic to the paramagnetic state. In the antiferromagnetic state, which occurs for magnetic field $H \ll H_c$, the antiferromagnetic vector L in $NaMnCl_3$ is oriented in the basal plane of the crystal, perpendicular to the c axis. This conclusion can be drawn from a comparison of the values of the magnetic susceptibility for magnetic field $H \perp c$, $\chi_\perp(T)$, and for magnetic field $H \parallel c$, $\chi_\perp^*(T)$. The difference in the values of susceptibility $\chi_\perp$ and $\chi_\perp^*$ is explained solely by the anisotropy of the transverse magnetic susceptibility. The antiferromagnetic vector L is perpendicular to H for both orientations of the magnetic field. On change of the orientation of the applied magnetic field H in the basal plane, $H \perp c$, there occurs a change of orientation of the antiferromagnetic vector $L \perp H$, since according to experiment (Fig. 2) anisotropy of the magnetic moment $M(H)$ on change of orientation of H⊥ cm is not observed.

In the antiferromagnetic state at $H \ll H_c$, the magnetic properties of a $NaMnCl_3$ monocrystal (the linear magnetic-moment dependence $M(H)$) are described by a thermodynamic potential Φ of the form

$$\Phi = {}^1/_2 B\mathbf{m}^2 + {}^1/_2 b m_z^2 + {}^1/_2 a\gamma_z^2 - \mathbf{mH}, \tag{1}$$

where the invariant $\frac{1}{2}B\mathbf{m}^2$ corresponds to the exchange interaction of the spins, the invariant $\frac{1}{2}bm_z^2$ is responsible for the anisotropy of the magnetic susceptibility $\chi_\perp$ for $H \mid c$ and for $H \parallel c$, and the invariant $\frac{1}{2}a\gamma_z^2$ is responsible for the orientation of the antiferromagnetic vector L with respect to the high-order axis of the crystal. By minimization of the thermodynamic potential (1) we get

$$M(H) = \chi_\perp H = H/B, \quad \mathbf{H} \perp c;$$
$$M(H) = \chi_\perp^* H = H/(B+b), \quad \mathbf{H} \parallel c.$$

The effective exchange-interaction field $H_E = M_0/\chi_\perp$ in $NaMnCl_3$, determined from experiment with the magnetic field oriented perpendicular to the axis, is found to be $H_E = 14 \pm 0.5$ kOe. The effective anisotropy field, determined from the anisotropy of the transverse magnetic susceptibility ($\chi_\perp$ and $\chi_\perp^*$), is found to be $H_A = 2.5 \pm 0.1$ kOe.

On increase of a magnetic field H applied either perpendicular to the high-order c axis or along it, a continuous rotation of the magnetic moments of the sublattices occurs up to magnetic field $H = H_c$, at which the antiferromagnetic state is destroyed and a phase transition occurs to the paramagnetic state at the given temperature. This transition is characterized by the following features.

1. According to molecular-field theory in the two-sublattice model, the transition of an antiferromagnet to the paramagnetic state on application of an external magnetic field should occur at a field[5]

$$H_c = 2H_E, \quad H_c^* = 2H_E + H_A, \tag{2}$$

where $H_E = M(T)/\chi_\perp$, $M(T)$ is the magnetization of a sublattice at the given temperature, and H_A is the anisotropy field. Then from the relations (2) and the values of H_E and H_A obtained from experiments at $H \ll H_c$, one can obtain the following values of the quantities H_c and H_c^*:

$$H_c = 24 \text{ kOe}, \; H_c^* = 27 \text{ kOe}.$$

From Fig. 5, however, it is evident that the experimental values of the critical fields H_c and H_c^* for the phase transition, obtained by extrapolation to temperature $T = 0$ K, are significantly smaller:

$$H_c(0) = 15.5 \pm 0.5 \text{ kOe}, \; H_c^*(0) = 18 \pm 0 \text{ kOe}.$$

2. The value of the effective exchange-interaction field calculated from the value of the Néel temperature T_N of this monocrystal,[6]

$$H_E^* = 3kT_N/(S+1)g\mu_B,$$

is found to be $H_E^* \approx 40$ kOe and considerably exceeds the value of the effective exchange-interaction field H_E determined from experiment in the antiferromagnetic state at $H \ll H_c$.

3. At magnetic fields $H \approx H_c$, $H < H_c$ (for both directions of the magnetic field $\mathbf{H}$), a nonlinear growth is observed on the magnetic-moment curve $M(H)$; that is, a deviation of the variation of magnetic moment with field from the linear relation predicted by molecular-field theory.

The difference between the exchange fields estimated from the magnetic susceptibility $\chi_\perp$ in the antiferromagnetic state at $H \ll H_c$ and from the value of the Néel temperature T_N of the given monocrystal can be explained by supposing that in $NaMnCl_3$, in addition to the antiferromagnetic interaction between the two sublattices, there is a stronger ferromagnetic interaction within the sublattices. The exchange field determined from the value of the magnetic susceptibility $\chi_\perp$ is caused by the antiferromagnetic interaction, whereas the Néel temperature is determined by the sum of the absolute values of the ferromagnetic and antiferromagnetic exchange interactions. Such a situation can arise if we suppose that the magnetic structure of $NaMnCl_3$ constitutes ferromagnetic planes coupled by weaker antiferromagnetic interaction. Then the magnetic properties of $NaMnCl_3$ should be described by two effective exchange fields, exchange interactions of different signs.

On approach of the magnetic field H to H_c, as has already been pointed out, a nonlinear growth of the magnetic moment $M(H)$ occurs. Such a form of the $M(H)$ relation near the point H_c of phase transition from the antiferromagnetic to the paramagnetic state can be explained on the basis of theoretical calculations presented in a paper of Jacobs and Siverstein,[7] with allowance for biquadratic exchange. Then the thermodynamic potential Φ describing the magnetic properties of the monocrystal near the phase-transition point is written in the form

$$\Phi = {}^1\!/_2 B(\mathbf{M}_1\mathbf{M}_2) + {}^1\!/_4 D(\mathbf{M}_1\mathbf{M}_2)^2 - (\mathbf{M}_1 + \mathbf{M}_2)\mathbf{H}, \tag{3}$$

where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations, B is the first-order exchange-interaction constant, and D is the biquadratic-exchange constant. For simplicity of the calculations, anisotropy is disregarded in this thermodynamic potential. We did this calculation for the case $\mathbf{H} \perp c$. By minimization of (3), one can obtain an equation for the motion of the magnetic moments of the sublattices on increase of the magnetic field H. The effect of biquadratic exchange also leads to a decrease of the value of the phase-transition magnetic field H_c as compared with the deductions of molecular-field theory. In Fig. 2, the points represent calculated $M(H)$ relations when the ratio of the biquadratic-exchange and first-order exchange constants is $\beta = D/B \approx 0.2$.

The nonlinearity of the $M(H)$ relation near the phase-transition point can also be explained on the basis of the theory of the effect of spin waves[8] on the magnetization near the phase-transition point H_c. But the temperature $T = 4.2$ K, at which the experiment was done whose data were compared with the theoretical curves, is still quite close to T_N, whereas the theoretical calculations were done for $T \ll T_N$; therefore for comparison of the experimental data with the deductions of theoretical calculations,[8] lower temperatures, $T < 2$ K, are needed hereafter.

To determine the temperature dependence of the phase-transition magnetic field $H_c(T)$ (Fig. 5), we plotted the variation of $\ln H_c(T)$ with $\ln(T)$. In Fig. 6, the points represent this relation for $\mathbf{H} \perp c$. It is seen from the curve that with lowering of the temperature, the phase-transition magnetic-field dependences $H_c(T)$ come out onto a dependence determined by the law $H_c \propto T^{3/2}$ (dotted curve), as was predicted in Ref. 8. But as has already been pointed out, for a more detailed treatment lower temperatures are needed.

For magnetic field $H \gg H_c$, the magnetization curves are described by Brillouin functions corresponding to the given temperature. The magnetic moment is expressed in the form

$$M(H) = Ng\beta S B_S(hS) = 2M_0 B_S(hS), \tag{4}$$

where

$$hS = \frac{g\mu_B S}{kT}\left[(H + H_E^{\text{eff}})\frac{M(T)}{2M_0}\right]. \tag{5}$$

Agreement of the experimental results for the $M(H)$ relation, with magnetic field $H \gg H_c$ and $\mathbf{H} \perp c$, with the theoretical calculation is observed when $H_E^{\text{eff}} = 10 \pm 1$ kOe. If we suppose that in the present case $H_E^{\text{eff}} = H_E^{\text{FM}} - H_E^{\text{AFM}}$, where H_E^{FM} is the effective field of ferromagnetic interaction (as was pointed out above, an effective interaction H_E^{FM} of ions in this crystal is possible within planes perpendicular to the c axis) and H_E^{AFM} is the effective field of antiferromagnetic interaction (as was pointed out, such interaction of ions is possible between ions of the planes perpendicular to c), then since $H_E^{\text{AFM}} = 14$ kOe, $H_E^{\text{FM}} = 23 \pm 1$ kOe.

If the magnetic field $\mathbf{H}$ is applied along the c axis, then a calculation of the theoretical $M(H)$ curve for $H_A = 3$ kOe and $H_E^{\text{AFM}} = 14$ kOe does not lead to agreement with experiment. By comparing the theoretical calculation of the Brillouin function at various values of the field H_{eff} with experiment, one can obtain the value of this field. But more detailed conclusions require additional experiments. Yet the experiments under discussion do not contradict the assumption that $NaMnCl_3$ is perhaps a quasi-two-dimensional antiferromagnet, in which the magnetic moments of ions in planes perpendicular to the c axis are parallel (that is, the ex-

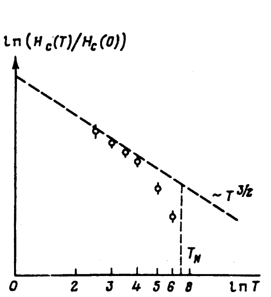

FIG. 6. Variation of $\ln H_c(T)/H_c(0)$ with $\ln T$.

change interaction of ions in the planes is ferromagnetic), whereas the magnetic moments of ions in different planes are antiparallel (that is, the exchange interaction of ions belonging to different planes is antiferromagnetic).

We consider the experimental data obtained at temperatures above the transition point T_N. As is seen from Fig. 3, above the phase-transition point the nonlinear magnetic-moment relation $M(H)$ in weak fields, $H<10$ kOe, disappears, and $M(H)$ is sufficiently well described by the expression $M(H) = \chi_\perp(T)H$. On increase of the magnetic field above 10 kOe, there appears a strong nonlinearity of the magnetic-moment dependence, characteristic of a "saturating" paramagnet; that is, when the magnetic moment $M(H)$ of the paramagnet is close to the magnetic moment of the magnetic ion at the prescribed temperature. The values of the magnetic susceptibility $\chi(T)$ above the phase-transition point T_N, presented in Fig. 5, were obtained by processing of the linear sections of the $M(H)$ relations of Fig. 3 at $H<10$ kOe. By processing the magnetic-susceptibility data above the phase-transition point, one can obtain a Curie-Weiss law with the value $\Theta = 3 \pm 1$ K. The fact that the value of Θ was found to be positive supports the assumption that the $NaMnCl_3$ monocrystal is perhaps a quasi-two-dimensional antiferromagnet, in which the value of the ferromagnetic exchange interaction in the basal plane, perpendicular to the c axis, exceeds the value of the antiferromagnetic exchange interaction between the planes.

Thus the results of the research show that in antiferromagnetic $NaMnCl_3$ at $T = 4.2$ K, a second-order phase transition from the antiferromagnetic to the paramagnetic state occurs in magnetic fields $H_c = 15$ kOe. In the research, the magnetic-moment dependence $M(H)$ was investigated with the magnetic field H perpendicular and parallel to the high-order axis of the crystal, at various temperatures.

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1) Staff member, L. V. Kirenskiĭ, Institute of Physics, Siberian Dvision, Academy of Sciences, USSR.

2) The authors thank B. V. Beznosikov for providing the monocrystals for the investigation.

3) It is seen from Fig. 2 that when $H = H_c$, for given magnetic fields the $M(H)$ relation approaches a value of the magnetic moment somewhat different from $2M_0$; but with increase of H, there occurs an increase of the magnetic moment $M(H)$, and this gives a basis for supposing that in strong magnetic fields $M(H)$ asymptotically approaches the value $2M_0$.

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Ferromagnetism of dilute PdNi alloys

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According to the experimental data, in dilute PdNi alloys there exist local moments associated with groups of two or three nearest-neighbor nickel atoms in the lattice. A theory which takes into account the polarization of the d-electrons on isolated nickel atoms due to the local moments, and the strong nonuniformity of the alloy, has made it possible to explain in a natural way the unique concentration dependences of the longitudinal susceptibility, magnetization, and Curie temperature of these alloys.

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INTRODUCTION

Alloys of palladium with nickel possess many distinctive properties, which make them an interesting object for study.

Unlike iron or cobalt atoms, a nickel atom in palladium does not possess a magnetic moment. Ferromagnetism has been reliably observed in alloys with a nickel concentration exceeding 2.3 at.%. Near this concentration the susceptibility of the alloy and also the elec-