research, V. F. Dergachev and Z. A. Magomedov for technical help in preparation of the samples, and F. N. Sultanov for participation in a number of the experiments.

[1)]In a cubic semiconductor with nondegenerate bands.
[2)]We of course assume that the length of the ultrasonic pulse is sufficiently great: $t_i \gg t_f$.

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Phase diagram of a uniaxial antiferromagnet

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The approximation of molecular-field theory is used to investigate the temperature dependence of the transition field and of the boundaries of the region of metastable states in an easy-axis antiferromagnet for a phase transition in which the magnetic moments of the sublattices turn over (a "spin-flop" transition). It is shown that at high temperatures a spin-flop transition always occurs as a first-order phase transition.

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For orientational phase transitions induced by an external magnetic field and of the type in which the magnetic moments of the sublattices turn over ["spin-flop" (SF) transitions], in easy-axis antiferromagnets, a characteristic property is the presence of various types of phase transitions and of critical points. This fact makes an antiferromagnet a convenient object for study of critical phenomena and of phase transitions. Furthermore, a magnetic field is an easily controlled means of acting on the object of investigation, and this considerably simplifies the experimental technique; on the other hand, the external magnetic field enters in a simple manner in the equation of state, and this substantially facilitates theoretical investigations. All of this explains the large number of experimental and theoretical papers devoted to the study of the SF transition in an antiferromagnet.[1-8 1)]

A detailed theoretical investigation of the nature of phase transitions in easy-axis antiferromagnets at $T=0$ was carried out in Refs. 2 and 6. For temperatures different from zero, the theoretical investigations have been limited either to the low-temperature range ($T \ll T_N$, where T_N is the Néel temperature)[3] or to temperatures close to the triple-point temperature T_t.[4]

In the present paper, the approximation of molecular-field theory is used to study the temperature variation of the SF transition field H_{tr} and of the boundaries of the region of metastable states, in the case in which the magnetic field is directed along the easy axis. In Ref. 2 it was shown that at $T=0$, depending on the character of the anisotropy, the SF transition occurs either as two phase transitions of the second kind or as a phase transition of the first kind. Calculations made in the present paper show that at finite temperatures, the transition field and the instability fields of the individual phases depend little on the anisotropy and are primarily determined by the values of the exchange constants. At high temperatures, the transition from the antiferromagnetic (AF) state to the SF phase always

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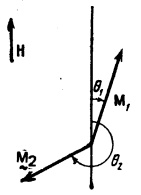

FIG. 1.

occurs as a first-order phase transition.

1. As is well known, the free-energy density of a two-sublattice antiferromagnet in an external magnetic field H, directed along the easy axis, can be written, in the approximation of molecular-field theory,

$$F=\lambda\sigma_1\sigma_2\cos(\theta_1-\theta_2)+{}^1/_2\delta(\sigma_1^2+\sigma_2^2)-\beta_1\sigma_1\sigma_2\cos\theta_1\cos\theta_2 -{}^1/_2\beta(\sigma_1^2\cos^2\theta_1+\sigma_2^2\cos^2\theta_2)-h(\sigma_1\cos\theta_1+\sigma_2\cos\theta_2)-T[S(\sigma_1)+S(\sigma_2)]. \quad (1)$$

Here the following notation has been introduced: $\sigma_1 = M_1(T)/M_0$ and $\sigma_2 = M_2(T)/M_0$ are the relative magnetizations of the sublattices; θ_1 and θ_2 are the angles between the magnetization vectors $\mathbf{M}_1$, $\mathbf{M}_2$ of the sublattices and the easy axis (Fig. 1); $h = HM_0$; λ and δ are the exchange constants corresponding to the intersublattice and to the intrasublattice interactions; β and β_1 intrasublattice and intersublattice anisotropy constants; T is the temperature; and S is the sublattice entropy $[dS(\sigma_i)/d\sigma_i = -B_s^{-1}(\sigma_i)$, where $B_s^{-1}(\sigma_i)$ is the inverse Brillouin function and s is the spin of the magnetic moment of the sublattice].

For systems with spin $s \geq 1$, it is necessary to introduce into the expression for the free energy the energy of single-ion anisotropy. But in the low-temperature range ($\sigma_1, \sigma_2 \to 1$) and at temperatures close to the triple-point temperature $\sigma_1, \sigma_2 \ll 1$), allowance for single-ion anisotropy reduces to a suitable transformation of the constant β in (1). Then β will have different values in the low- and high-temperature ranges.[5]

The possible configurations of the system are determined by solution of the system of equations $\partial F/\partial x_i = 0$; the region of stability of the solutions of this system is determined from the condition of nonnegativity of the principal minors of the matrix A_{ik} formed from the second derivatives of the free energy with respect to the parameters x_i of the system ($x_1 = \sigma_1, x_2 = \sigma_2, x_3 = \theta_1, x_4 = \theta_2$).

Investigation of the solutions of the system $\partial F/\partial x_i = 0$ shows[2,6] that, depending on the value of the external field and of the temperature, the following phases correspond to a stable state of the antiferromagnet.

1) The AF phase: $\theta_1 = 0, \theta_2 = \pi$; σ_1 and σ_2 are determined by the system of equations

$$\begin{aligned} -(\lambda-\beta_1)\sigma_2+(\delta-\beta)\sigma_1-h-TdS(\sigma_1)/d\sigma_1=0,\\ -(\lambda-\beta_1)\sigma_1+(\delta-\beta)\sigma_2+h-TdS(\sigma_2)/d\sigma_2=0. \end{aligned} \quad (2)$$

2) The SF phase: $\theta_1 = -\theta_2 = \theta, \sigma_1 = \sigma_2 = \sigma$; σ and θ are determined by the equations

$$\cos\theta = h/(2\lambda-\beta-\beta_1)\sigma(T), \quad (\lambda-\delta)\sigma = -TdS/d\sigma. \quad (3)$$

3) The paramagnetic (PM) phase: $\theta_1 = \theta_2, \sigma_1 = \sigma_2$.

4) The intermediate phase (IP) (in a certain field and temperature range, an antiferromagnet in which $\beta < 0$ is in this state).[2,6]

We shall now write the relations that determine the boundaries between individual phases.

Doing this is simplest for the boundary between the SF phase and the PM phase. For this purpose, it is sufficient to substitute $\theta = 0$ in the system of equations (3)[9]:

$$h_{\text{SF-PM}}(T) = (2\lambda-\beta-\beta_1)\sigma(T). \quad (4)$$

By use of the system of equations (2), the free energy of the system in the AF state near the line of transition to the PM state can be written

$$F_{\text{AF}} = (\lambda+\delta-\beta-\beta_1)\sigma_{\text{tr}}^2 - 2\sigma_{\text{tr}}h - [(\lambda-\delta+\beta-\beta_1)+T(d^2S/d\sigma^2)_{\sigma_{\text{tr}}}]\eta^2+\ldots, \quad (5)$$

where σ_{tr} is the relative magnetization of the sublattices on the AF-PM phase-transition line, and where $\eta = (\Delta\sigma_1 + \Delta\sigma_2)$ is the order parameter.

On equating the coefficient of η^2 to zero and using the equations for $\sigma_{\text{tr}}(T)$, we get the system of equations that determines $h_{\text{AF-PM}}(T)$ and $\sigma_{\text{tr}}(T)$,[5]

$$\begin{aligned} &(\lambda-\delta+\beta-\beta_1)+T(d^2S/d\sigma^2)_{\sigma_{\text{tr}}}=0,\\ &h_{\text{AF-PM}}(T) = (\lambda+\delta-\beta-\beta_1)\sigma_{\text{tr}} - T(dS/d\sigma)_{\sigma_{\text{tr}}}. \end{aligned} \quad (6)$$

For $d^2S/d\sigma^2$, the following equation can be derived:

$$\frac{d^2S}{d\sigma^2} = \left\{\sigma^2 + \frac{\sigma}{s}\operatorname{cth}\left[\frac{B_s^{-1}(\sigma)}{2s}\right] - \frac{s+1}{s}\right\}^{-1}. \quad (7)$$

The system of transcendental equations (6) takes a simple form when $s = \frac{1}{2}$:

$$\sigma_{\text{tr}} = (1-T/T_N)^{1/2}, \quad h_{\text{AF-PM}} = (\lambda+\delta-\beta-\beta_1)\sigma_{\text{tr}} - T\operatorname{arcth}\sigma_{\text{tr}}. \quad (8)$$

The Néel temperature $T_N = (\lambda - \delta - \beta - \beta_1)$ can be obtained from the system (2) for $\sigma_1, \sigma_2 \to 0$.

Simultaneous solution of the system of equations (3), (4), and (8) determines the parameters of the triple point (T_t, h_t, σ_t).

In particular, for $s = \frac{1}{2}$ we get from equations (3), (4), and (8)

$$\sigma_t = (1-T_t/T_N)^{1/2}, \quad (9)$$

$$h_t = (2\lambda-\beta-\beta_1)(1-T_t/T_N)^{1/2}, \quad (10)$$

and T_t is determined from the equation $(\lambda - \delta)\sigma_t = T_t \times \operatorname{arcth}\sigma_t$ with use of (9).

It is convenient here to introduce the dimensionless parameter $\varepsilon = (\beta - \beta_1)/(\lambda - \delta)$, which determines the relation between the exchange and the anisotropy properties of the antiferromagnet. By use of equations (2) and (4), one can write, in terms of T_N and of the temperature T_3 obtained by extrapolation of the SF-PM phase-transition line to $h = 0$,

$$\varepsilon = (T_N - T_3)/T_3. \quad (11)$$

Usually the value of ε is of the order of magnitude 10^{-2}. For example, for $CuCl_2 \cdot 2H_2O$, $\varepsilon = 7.19 \cdot 10^{-3}$; for MnF_2, $\varepsilon = 2.49 \cdot 10^{-2}$; for $GdAlO_3$, $\varepsilon = 1.16 \cdot 10^{-2}$ (according to data of Refs. 5, 7, and 10). By carrying out in equations (3) and (6) an expansion with respect to the small parameter ε, one can obtain the following expressions for the triple-point parameters:[2)]

$$\sigma_t = \left[\frac{5}{3}\frac{(s+1)^2}{(s+1)^2+s^2}\varepsilon\right]^{1/2}, \quad (12)$$

$$T_t=(\lambda-\delta)\frac{(s+1)}{3s}\left[1-\frac{\varepsilon}{2}\right], \tag{13}$$

$$h_t=(2\lambda-\beta-\beta_1)\left[\frac{5}{3}\frac{(s+1)^2}{(s+1)^2+s^2}\right]^{1/2}. \tag{14}$$

It should be noted that whereas the transition field to the SF phase at $T=0$, $h_{tr}^0=[(\beta-\beta_1)(2\lambda-\beta-\beta_1)]^{1/2}$, is determined by the value of the intersublattice exchange λ, the triple-point field h_t depends substantially on the intrasublattice exchange constant δ. The ratio of the fields h_t and h_{tr}^0 is determined by the relation between the values of λ and of δ:

$$\frac{h_t}{h_{tr}^0}=\left[\frac{10(s+1)^2}{3[(s+1)^2+s^2]}\frac{\lambda}{\lambda-\delta}\right]^{1/2}. \tag{15}$$

Analysis of the relation (15) enables us to draw an interesting conclusion: with increase of the spin number of the magnetic moment of the sublattice, there is an increase of the effect of intrasublattice exchange on the temperature variation of the transition field $h_{tr}(T)$. Thus, for example, for an antiferromagnet with spin $s=\frac{1}{2}$ the inequality $h_t<h_{tr}^0$ is satisfied for $-\delta>2\lambda$ (the minus sign means that $\delta<0$; that is, in this case there is ferromagnetic ordering within the sublattices); for a system with spin $s=\frac{7}{2}$, this inequality is already valid when $-\delta>\lambda$. Allowance for single-ion anisotropy l in this case reduces to introduction in (15) of a factor $[(\beta-\beta_1+l_t)/(\beta-\beta_1+l_0)]^{1/2}$, where l_t and l_0 are the values of the single-ion anisotropy constant at $T=T_t$ and at $T=0$ respectively; $l_t<l_0$. The considerations advanced here can be used to explain the anomalous temperature variation of the transition field $h_{tr}(T)$ in $GdAlO_3$ ($s=\frac{7}{2}$): whereas for most antiferromagnets $h_{tr}(T)$ increases with increase of temperature, in $GdAlO_3$ the opposite behavior is observed[5] (see Fig. 2).

2. We turn to the determination of the instability boundaries of the AF and SF phases.

The instability boundary of the AF phase, $h_{\parallel}(T)$, is determined by simultaneous solution of the system of equations (2) and of the equation $|A_{ik}|_{AF}=0$, where $|A_{ik}|_{AF}$ is the determinant of the matrix A_{ik} for the AF phase. The determinant $|A_{ik}|_{AF}$ can be expressed as the product of two second-order determinants:

$$|A_{ik}|_{AF}=D_1D_2,$$

$$D_1=\begin{vmatrix}\delta-\beta-T\dfrac{d^2S}{d\sigma_1^2} & -(\lambda-\beta_1)\\ -(\lambda-\beta_1) & \delta-\beta-T\dfrac{d^2S}{d\sigma_2^2}\end{vmatrix}, \tag{16}$$

$$D_2=\begin{vmatrix}(\lambda-\beta_1)\sigma_2+\beta\sigma_1+h & \lambda\\ \lambda & (\lambda-\beta_1)\sigma_1+\beta\sigma_2-h\end{vmatrix}\sigma_1^2\sigma_2^2.$$

The determinant D_1 expresses the stability of the AF state with respect to enlargement of the magnetic moments. In Ref. 14, which was devoted to the study of phase transitions in metamagnets, it was shown that D_1 can change sign when the external field approaches the exchange field in order of magnitude ($h\sim\lambda$). In antiferromagnets, the transition to the SF phase occurs in fields considerably smaller than the exchange field ($h\sim[2\lambda(\beta-\beta_1)]^{1/2}$). In this range, $D_1>0$. Thus the instability boundary $h_{\parallel}(T)$ of the AF phase is determined by the equation $D_2=0$ and can be expressed in terms of $\sigma_1(T)$ and $\sigma_2(T)$:

$$h_{\parallel}(T)=\frac{(\lambda-\beta-\beta_1)(\sigma_1-\sigma_2)}{2}+\frac{1}{2}[(\beta-\beta_1)(2\lambda+\beta-\beta_1)(\sigma_1+\sigma_2)^2+\lambda^2(\sigma_1-\sigma_2)^2]^{1/2}. \tag{17}$$

By introducing the quantities $\sigma_+=(\sigma_1+\sigma_2)/2$, $\sigma_-=(\sigma_1-\sigma_2)/2$, and $h_{\parallel}^0=[(\beta-\beta_1)(2\lambda+\beta-\beta_1)]^{1/2}$ (the instability boundary of the AF phase at $T=0$[2]), we can write $h_{\parallel}(T)$ thus:

$$h_{\parallel}(T)=(\lambda-\beta-\beta_1)\sigma_-+[(h_{\parallel}^0)^2\sigma_+^2+\lambda^2\sigma_-^2]^{1/2}. \tag{18}$$

It should also be noted that the determinant D_2 vanishes when $\sigma_2=0$; but this condition does not determine an instability boundary: D_2 does not change sign on passage through this value.

The expression (11) together with the equations (2) forms a system of three equations for the three unknowns $h_{\parallel}(T)$, $\sigma_1(T)$, and $\sigma_2(T)$. This system can be reduced to a single equation for $\sigma_1(T)$. For this purpose we introduce $z(\sigma_1)$:

$$\sigma_2=z\sigma_1. \tag{19}$$

On substituting (19) in the first equation of the system (2), we get the following equation for σ_1:

$$\frac{dS(z\sigma_1)}{d\sigma}=\frac{\sigma_1}{2T}f(z), \tag{20}$$

where

$$f(z)=(\lambda+\beta-\beta_1)+(\lambda+\beta-\beta_1-2\delta)z-[(\lambda+\beta-\beta_1)^2(z+1)^2-4\lambda^2z]^{1/2},$$

$$z=\left[-\frac{T}{\sigma_1}\frac{dS}{d\sigma}-(\lambda+\delta-\beta-\beta_1)\right]\left\{(\lambda+\beta-\beta_1)-\lambda^2\left[-\frac{T}{\sigma_1}\left(\frac{dS}{d\sigma}\right)+\delta\right]^{-1}\right\}^{-1}.$$

For the SF phase, the determinant $|A_{ik}|_{SF}$ can, by identical transformations, be reduced to the product of two second-order determinants:

$$|A_{ik}|_{SF}=D_1D_2,$$
$$D_1=2(\lambda-\delta)(2\lambda-\beta-\beta_1)(1-\cos 2\theta),$$
$$D_2=2(\lambda-\delta+2Td^2S/d\sigma^2)(2\lambda+\beta-\beta_1)\cos 2\theta+(2\lambda+\beta-\beta_1)^2-(2\delta-\beta+\beta_1-4Td^2S/d\sigma^2). \tag{21}$$

The determinant $D_1\geq 0$ and vanishes on the SF-PM line; thus the boundary $h_{\perp}(T)$ of the SF phase is determined by the equation $D_2=0$:

$$h_{\perp}(T)=h_{\perp}^0\left[1+\frac{2\lambda}{-Td^2S/d\sigma^2-(\lambda-\delta)}\right]^{1/2}\sigma(T) \tag{22}$$

where $h_{\perp}^0=[(\beta-\beta_1)(2\lambda+\beta-\beta_1)]^{1/2}(2\lambda-\beta-\beta_1)/(2\lambda+\beta-\beta_1)$ is the instability boundary of the SF phase at $T=0$.[2]

The transition field $h_{tr}(T)$ is determined by equality of the free energies $F=F_{SF}$. Using the systems of equations (2) and (3), we express $h_{tr}(T)$ in terms of the values of the magnetizations $\sigma_+(T)$, $\sigma_-(T)$, and $\sigma(T)$:

$$h_{tr}(T)=(2\lambda-\beta-\beta_1)\sigma_-+\{(2\lambda-\beta-\beta_1)[(\lambda-\delta+\beta-\beta_1)\sigma_+^2-(\lambda-\delta)(\sigma^2-\sigma_-^2)-TS(\sigma_1)-TS(\sigma_2)+2TS(\sigma)]\}^{1/2}, \tag{23}$$

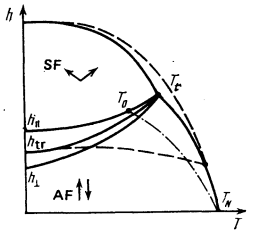

FIG. 2. Phase diagram of a uniaxial antiferromagnet ($\beta>0$). The dotted line shows the phase diagram of $GdAlO_3$;[5] the dashed-dotted line represents the line $h_{\sigma_2=0}(T)$.

$\sigma(T)$ is determined by equation (3), $\sigma_+(T)$ and $\sigma_-(T)$ by the system (2) with $h=h_{tr}(T)$.

3. We turn now to a study of the asymptotic behavior of the fields h_{tr}, $h_\parallel$, and $h_\perp$. In order to simplify the calculations, we restrict ourselves to a study of systems with spin $s=\frac{1}{2}$.

In the low-temperature range ($T<\frac{1}{2}T_N$), the values of the sublattice magnetizations σ_1, σ_2, $\sigma\sim 1$. Then the expressions for h_{tr}, $h_\parallel$, and $h_\perp$ simplify considerably:

$$h_{tr}(T)=h_{tr}^0\left[1+\frac{2T_N}{(\beta-\beta_1)}\exp\left(-\frac{2T_N-h_{tr}^0}{T}\right)\right], \tag{24}$$

$$h_\parallel(T)=h_\parallel^0+(\lambda-\beta-\beta_1)\exp\left(-\frac{2T_N-h_\parallel^0}{T}\right), \tag{25}$$

$$h_\perp(T)=h_\perp^0\left[1+\frac{4\lambda}{T}\exp\left(-\frac{2(\lambda-\delta)}{T}\right)\right], \tag{26}$$

$$\Delta h(T)=h_\parallel-h_\perp=(h_\parallel^0-h_\perp^0)+\left[(\lambda-\beta-\beta_1)\right.$$

$$\left.\times\exp\left(-\frac{2T_N-h_\parallel^0}{T}\right)-\frac{4\lambda}{T}\exp\left(-\frac{2(\lambda-\delta)}{T}\right)\right]. \tag{27}$$

If a transition of the first kind occurs in the system ($h_\parallel > h_\perp$), then $\Delta h(T)$ determines the width of the region of existence of metastable states; for transitions of the second kind ($h_\parallel < h_\perp$), $|\Delta h(T)|$ determines the width of the region of existence of the IP.

At temperatures close to the triple-point temperature, it may be supposed that $\sigma=\sigma_t+\Delta\sigma$, $\sigma_1=\sigma_t+\Delta\sigma_1$, $\sigma_2=\sigma_t-\Delta\sigma_2$ ($\Delta\sigma$, $\Delta\sigma_1$, $\Delta\sigma_2\ll 1$). On expanding in the small parameters $\Delta\sigma$, $\Delta\sigma_1$, and $\Delta\sigma_2$ in equations (2) and (3), we arrive at the following relations:

$$h_{tr}(T)=h_t[1-a\Delta T], \tag{28}$$

$$h_\parallel(T)=h_t\left[1-\left(\frac{\lambda+\delta}{\lambda-\delta}-\frac{2\beta}{\lambda}\right)\frac{\Delta T}{2\lambda}\right] \tag{29}$$

$$h_\perp(T)=h_t\left[1-\frac{(5\lambda+2\delta)}{(\lambda-\delta)}\frac{\Delta T}{4\lambda}\right], \tag{30}$$

$$\Delta h(T)=h_t\left[\frac{3\lambda}{\lambda-\delta}+\frac{2\beta}{\lambda}\right]\frac{\Delta T}{2\lambda}, \tag{31}$$

where $\Delta T=T_t-T$ and $a=(\frac{8}{5}\lambda+\delta)/2\lambda(\lambda-\delta)$.

We note that $\Delta\sigma/\sigma_t=\varepsilon^{-1}\Delta T/T_t$; therefore the expressions obtained are valid within a temperature interval $\Delta T\lesssim 10^{-3}T_t$.

It follows from the relations (28)–(31) that in all easy-axis antiferromagnets, in the vicinity of the triple point the transition from the AF state to the SF phase occurs as a phase transition of the first kind. The width $\Delta h(T)$ of the region of metastable states decreases according to a linear law. The transition field $h_{tr}(T)$ and the instability fields $h_\parallel(T)$ and $h_\perp(T)$ of the individual phases also vary, in this range, according to a linear law. The coefficients of ΔT in formulas (28), (29), and (30) may take different signs, depending on the relation between the exchange constants λ and δ. In other words, on approach to the triple point each of the fields h_{tr}, $h_\parallel$, and $h_\perp$ may either decrease or increase; depending on the values of λ and δ, all variants consistent with the inequalities $h_\parallel > h_{tr} > h_\perp$ are possible.

On the phase diagram of the states of an antiferromagnet (the $H-T$ plane), there is in the AF phase a line $h_{\sigma_2=0}(T)$ on which the value of the magnetization of the sublattice oriented opposite to the field, σ_2, vanishes.[5] The temperature T_0 at which the lines $h_\parallel(T)$ and $h_{\sigma_2=0}(T)$ intersect is determined by simultaneous solution of the equations of the system (2) and of equation (17) under the condition $\sigma_2=0$:

$$T_0=\frac{\lambda^2}{\lambda+\beta-\beta_1}-\delta. \tag{32}$$

At this temperature, the instability fields $h_\parallel$ and $h_\perp$ are determined by the following relations:

$$h_\perp=h_t\left[1+\frac{(\beta-\beta_1)}{4\lambda}+\varepsilon f(\varepsilon)\right], \tag{33}$$

$$h_\parallel=h_t\left[1+\frac{(\beta-\beta_1)}{4\lambda}+\varepsilon g(\varepsilon)\right], \tag{34}$$

$$\Delta h=h_t(g-f)\varepsilon \tag{35}$$

$f(\varepsilon)$ and $g(\varepsilon)$ are certain functions of ε, and $f(\varepsilon)<g(\varepsilon)$.

It follows from the relations (33), (34), and (35) that at temperature T_0 the transition from the AF state to the SF phase, independently of the character of the anisotropy, occurs as a phase transition of the first kind.

Thus analysis of the temperature dependence of the transition field h_{tr} and of the instability fields $h_\parallel$ and $h_\perp$ leads to the conclusion that for antiferromagnets with anisotropy $\beta>0$, the transition from the AF state to the SF phase occurs, over the whole temperature interval from zero to T_t, as a phase transition of the first kind (Fig. 2).

For antiferromagnets with anisotropy $\beta<0$ at sufficiently low temperatures ($T<\frac{1}{2}T_N$), the SF transition is of the same character as at $T=0$; that is, there are two phase transitions of the second kind with formation of an IP. With increase of temperatures, the width $|\Delta h(T)|$ of the region of existence of the IP decreases [see (27)]; and at temperatures near the triple-point temperature, the SF transition occurs as a first-order phase transition.

Thus for an antiferromagnet with anisotropy $\beta<0$, the lines $h_\perp(T)$ and $h_\parallel(T)$ intersect at a certain temperature T^*. This situation requires additional discussion. The fact is that lines of second-order transitions from symmetric phases (in this case the AF and the SF phases) to an asymmetric (the IP) in general cannot terminate at a single point. In fact such a point is also the termination of a first-order transition line between the symmetric phases, and therefore at it both symmetric phases lose their stability. Thus if the system has other stable states, then in the vicinity of the intersection point there will occur phase transitions to these states. In order to study these additional states in the vicinity of the point T^*, it is necessary, in the phenomenological expression (1) for the free energy, to introduce additional terms, describing the magnetic anisotropy energy. A possible form of the phase diagram is shown in Fig. 3. In Fig. 3(a) are plotted the lines of instability of the individual phases, and in Fig. 3(b) the corresponding lines of first-order phase transitions. It is quite obvious that some of the points $T_\perp$, $T_\parallel$, $T_\perp'$, and $T_\parallel'$ may coincide with T^*. We remark also that the possibility of existence of the point T^* was first

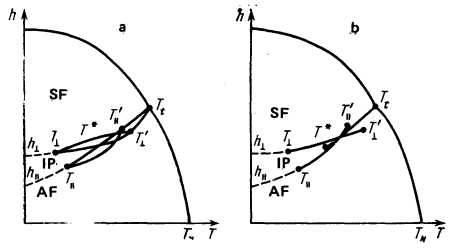

FIG. 3. Phase diagram of a uniaxial antiferromagnet ($\beta < 0$). The dotted lines denote line of transition of the second kind to the IP from the AF and SF phases: $T_{\parallel}$, $T_{\perp}$, $T_{\parallel'}$, and $T_{\perp'}$ are the terminal points of lines of phase transition of the first kind to the IP.

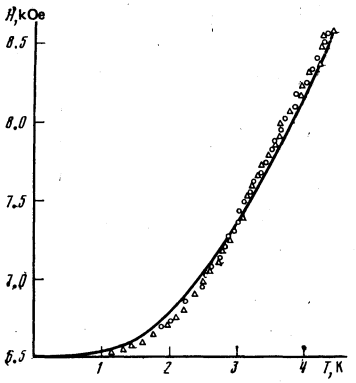

FIG. 4. Temperature dependence of the transition field h_{tr} for $CuCl_2 \cdot H_2O$. Points, results of experimental investigation by the AFMR method (according to the data of Ref. 7); triangles, results of experimental investigation by the NMR method. The solid line represents the results of a numerical calculation.

indicated in Ref. 11.

4. As is well known, in fields close to the flop field, an antiferromagnet is in the intermediate state.[12] Analysis of the nuclear magnetic resonance signal from protons in this range makes it possible to determine, comparatively easily, the temperature dependence of the SF transition field. In the present research, this method was used to determine the transition field in $CuCl_2 \cdot 2H_2O$ ($s=\frac{1}{2}$). The experimental method and the theoretical considerations applied to the interpretation of the experimental data were presented in detail in Refs. 8 and 13. We mention only that the investigations were carried out over the temperature interval 0.96 to 4.3 K, the accuracy of the temperature measurement was ±0.005 K, the accuracy of adjustment and measurement of the field in the region of the specimen was ±1 Oe, and the accuracy of orientation of the crystal was 1′. The results of the experiment are in good agreement with data obtained by the method of antiferromagnetic resonance[1,14] (Fig. 4).

By use of experimntal data for $CuCl_2 \cdot 2H_2O$ ($T_N = 4.36$ K, $T_t = 4.31$ K, $H_{tr}^0 = 6.5$ kOe, $H_t = 8.5$ kOe[1,10]) and of the relations (8), (9), and (15), one can determine the molecular-field constants

$$\lambda = 2.47 \text{ K}, \ \delta = -1.86 \text{ K}, \ \beta - \beta_1 = 0.03 \text{ K}. \qquad (36)$$

The comparative closeness of the values of λ and of $|\delta|$ is in agreement with the model of antiferromagnetic ordering in $CuCl_2 \cdot 2H_2O$ proposed by Poulos and Hardeman.[15] In this model, it is suggested that this antiferromagnet consists of alternating layers with ferromagnetic ordering. Since in this case the difference in the distances between lattice sites with parallel and with antiparallel orientation of the magnetic moments is small, it is reasonable to expect that the intersublattice and intrasublattice exchange interactions will be close in magnitude.

A theoretical $h_{tr}(T)$ relation for $CuCl_2 \cdot 2H_2O$ was obtained by numerical solution of the system of equations (2), (3), and (23); it is in good agreement with the results of the experimental investigations (Fig. 4). Results of a numerical calculation of $h_{\perp}(T)$ and $h_{\parallel}(T)$ for various values of β and β_1 show that at low temperatures, the instability fields remain practically constant; on further increase of the temperature, $h_{\perp}(T)$ and $h_{\parallel}(T)$ increase, while $\Delta h(T)$ decreases monotonically, vanishing at the triple point.

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[1)]More detailed bibliographic information can be found in Ref. 5.
[2)]An expression for T_t ws obtained in Ref. 5.

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