Vol. A58 (1980) *ACTA PHYSICA POLONICA* No 6

THE INFLUENCE OF THE MAGNETIC FIELD ON THE THERMODYNAMIC PROPERTIES OF A TWO SUBLATTICE ANTIFERROMAGNET WITH SINGLE-ION ORTHORHOMBIC ANISOTROPY AND TRANSVERSALLY ANISOTROPIC EXCHANGE INTERACTION

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(Received September 28, 1979; revised version received March 13, 1980; final version received July 9, 1980)

By applying the linear theory of spin waves to the two sublattice antiferromagnet with single-ion orthorhombic and exchange anisotropy the formulas are obtained governing the dependences of thermodynamic quantities on the temperature. It is shown that the magnetization components may depend on the temperature in the SF ↔ P phase transition vicinity as T^2 or $T^{3/2}$ depending on whether the exchange anisotropy and/or single-ion anisotropy constants perpendicular to the easy direction are equal to zero or not. These results confirm the conclusions suggested by other authors.

PACS numbers: 75.50.Ee, 75.30.Et, 75.30.Ds, 75.10.Dg

1. *Introduction*

It is known that the behavior of an antiferromagnet in a homogeneous external magnetic field depends strongly upon the value of the exchange and anisotropy constants as well as on the direction of the external field. In a uniaxial antiferromagnet in the presence of the field parallel to the easy direction we will deal with the two phase transitions previously presented [1–6] that is, the first-order transition between the antiferromagnetic phase, AF, and the spin-flop, SF, and the second-order phase transition between the spin-flop phase and paramagnetic phase, P. On the other hand, there is only one second-order phase transition between the SF and P phase when the field is perpendicular to the easy axis. Another case occurs when the exchange anisotropy appears with the single-ion orthorhombic anisotropy. As has been shown [6–8], there may occur three second-order phase tran-

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sitions in the presence of the field parallel to the easy axis: AF ↔ CS, CS ↔ SF, SF ↔ P. This depends only upon the mutual quantities of the single-ion anisotropy in the directions parallel or perpendicular to the easy direction. On the other hand if the field is perpendicular to the easy axis, there occurs only second-order phase transition. The AF ↔ SF and SF ↔ P phase transitions in the magnetic field parallel and perpendicular to the easy axis have been investigated [9, 10]. For $MnCl_2 \cdot 4H_2O$ and $CoCl_2 \cdot 6H_2O$ the deviation of the critical field dependence on the temperature from the $T^{3/2}$-law has been shown. According to the authors of the above paper one should take into account the next term of the expansion proportional to $T^{5/2}$ [1–3] in order to obtain a better fit to the experimental data in a reasonable range. Nevertheless, even this improved law, $\alpha T^{3/2}+\beta T^{5/2}$, does not fit well to some experimental data [9, 10]. The disagreement between the $T^{5/2}$-law and the curves obtained in [9, 10] is explained by the existence of the perpendicular exchange anisotropy in $CoCl_2 \cdot 6H_2O$ and the single-ion orthorhombic anisotropy in $MnCl_2 \cdot 4H_2O$. That is, the reason why in [11] it has been suggested that the T^2-law for the critical field depends on temperature.

In this paper we will show that the presence of the perpendicular exchange anisotropy or single-ion orthorhombic anisotropy causes the thermodynamic quantities (m—magnetization, χ — susceptibility, C_S — spin specific heat and h_{cr} — critical field), the dependence on temperature in the vicinity of the critical field may differ from those obtained previously [1–3].

2. *Hamiltonian*

In our investigations we employ the spin Hamiltonian which can be represented as

$$H = J\sum_{\langle kj\rangle}[XS_k^xS_j^x+S_k^yS_j^y+ZS_k^zS_j^z]-L_x[\sum_k(S_k^x)^2+\sum_j(S_j^x)^2]$$
$$-L_z[\sum_k(S_k^z)^2+\sum_j(S_j^z)^2]-\mu H_x[\sum_k S_k^x+\sum_j S_j^x]-\mu H_z[\sum_k S_k^z+\sum_j S_j^z], \tag{1}$$

where $\langle k,j\rangle$ indicates the summation restricted to the nearest neighbours, S_k^α, S_j^α $(\alpha = x, y, z)$ stand for the spin operator components in $\{k\}$ and $\{j\}$ sublattices each of which includes $N/2$ lattice sites. $X = 1+K_x/J$, $Z = 1+K_z/J$, $K_x \geqslant 0$, $K_z \geqslant 0$ are the exchange anisotropy constants in the x and z-direction, $J > 0$ denotes the exchange constants, $L_x \geqslant 0$ and $L_z \geqslant 0$ are the single-ion anisotropy constants in x and z-directions, $H_x = H\sin\gamma$ and $H_z = H\cos\gamma$ are the components of a homogeneous magnetic field. The μ stands for the effective magnetic moment. After rotating the spin in the $\{j\}$ sublattice by the angle, φ, and the spin $\{k\}$ sublattice by the angle, θ, around the Oy axis (θ is measured from the positive z-direction, whereas φ is measured from the negative z-direction). Applying the Holstein–Primakoff transformation to the spin operators in the sublattices we find that:

$$(S_k^-)^+ = S_k^+ = (2S)^{1/2}f(n_k)b_k, \quad (S_j^-)^+ = S_j^+ = (2S)^{1/2}a_j^+f(n_j),$$
$$S_k^z = S-b_k^+b_k = S-n_k, \quad S_j^z = -S+a_j^+a_j = S-n_j, \tag{2}$$

where $f(n_k)\,(1-n_k/2S^{1/2}$, $f(n_j)=(1-n_j/2S)^{1/2}$. Then having applied the Fourier transformation

$$b_k=(2/N)^{1/2}\sum_{\lambda}\exp(-ik\lambda)b_{\lambda},$$

$$a_j=(2/N)^{1/2}\sum_{\lambda}\exp(ij\lambda)a_{\lambda}, \tag{3}$$

where λ stands for the reciprocal lattice vector. The Hamiltonian (1) is expressed as

$$H=E_0+H_1+H_2. \tag{4}$$

E_0 denotes the energy of the approximate ground state and may be represented as

$$E_0=\tfrac{1}{2}NS^2\gamma_0 J[X\sin\theta\sin\varphi-Z\cos\theta\cos\varphi-h_x(\sin\theta+\sin\varphi)$$
$$-h_z(\cos\theta-\cos\varphi)-a(\cos^2\theta+\cos^2\varphi)-4(2S-1)^{-1/2}(l_z+l_x), \tag{5}$$

where $h_{x,z}=\mu H_{x,z}$, $l_z-l_x=a=(L_z-L_x)\,(1-\tfrac{1}{2}S)/\gamma_0 JS$. The linear formula in the boson operators in (4) is the following:

$$H_1=\sum_{\lambda}[\alpha_{\lambda}(a_{\lambda}^{+}+a_{\lambda})+\beta_{\lambda}(b_{\lambda}^{+}+b_{\lambda})], \tag{6}$$

where

$$\alpha_{\lambda}=\tfrac{1}{2}(NS)^{1/2}\gamma_0 JS[X\sin\varphi\cos\theta+Z\sin\theta\cos\varphi+h_z\sin\theta$$
$$-h_x\cos\theta+2a\sin\theta\cos\theta]\delta_{\lambda,0}, \tag{7}$$

$$\beta_{\lambda}=\tfrac{1}{2}(NS)^{1/2}\gamma_0 JS[X\sin\theta\cos\varphi+Z\cos\theta\sin\varphi-h_z\sin\varphi$$
$$-h_x\cos\varphi+2a\sin\varphi\cos\varphi]\delta_{\lambda,0}, \tag{8}$$

and the bilinear form may be expressed as

$$H_2=\sum_{\lambda}[\alpha_1 b_{\lambda}^{+}b_{\lambda}+\alpha_2 a_{\lambda}^{+}a_{\lambda}+\alpha_{3\lambda}(a_{\lambda}^{+}b_{\lambda}+a_{\lambda}b_{\lambda}^{+})+\tfrac{1}{2}\beta_1(b_{\lambda}^{+}b_{-\lambda}^{+}+b_{\lambda}b_{-\lambda})$$
$$+\tfrac{1}{2}\beta_2(a_{\lambda}^{+}a_{-\lambda}^{+}+a_{\lambda}a_{-\lambda})+\beta_{3\lambda}(a_{\lambda}^{+}b_{\lambda}^{+}+a_{\lambda}b_{\lambda}), \tag{9}$$

where

$$\alpha_1=\gamma_0 JS[Z\cos\varphi\cos\theta-X\sin\varphi\sin\theta+h_x\sin\varphi$$
$$-h_z\cos\varphi-l_z(1-3\cos^2\varphi)-l_x(1-3\sin^2\varphi), \tag{10}$$

$$\alpha_2=\gamma_0 JS[Z\cos\varphi\cos\theta-X\sin\varphi\sin\theta-h_x\sin\theta-h_z\cos\theta$$
$$-l_z(1-3\cos^2\theta)-l_x(1-3\sin^2\theta), \tag{11}$$

$$\beta_1=-\gamma_0 JS[l_z\sin^2\varphi+l_x\cos^2\varphi], \tag{12}$$

$$\beta_2=-\gamma_0 JS[l_z\sin^2\theta+l_x\cos^2\theta], \tag{13}$$

$$\alpha_{\lambda 3}=\tfrac{1}{2}\gamma_0 JS\delta_{\lambda}[X\cos\varphi\cos\theta-Z\sin\varphi\sin\theta-1], \tag{14}$$

$$\beta_{\lambda 3}=\tfrac{1}{2}\gamma_0 JS\delta_{\lambda}[X\cos\varphi\cos\theta-Z\sin\varphi\sin\theta+1], \tag{15}$$

and $\gamma_{\lambda}=\sum_{\Delta}\exp(i\Delta\lambda)$, $\gamma_{\lambda}/\gamma_0=\delta_{\lambda}$ (γ_0 the number of the nearest neigbours).

On the grounds for the minimum condition for the approximate ground state energy, E_0 (5), one can easily notice the disappearance of the linear terms (6) of the Hamiltonian (4) [12, 13]. Therefore, the Hamiltonian will be presently expressed as:

$$H = \mathscr{E}_0 + H_2. \tag{16}$$

By applying the Bogolubov–Tiablikov transformation we obtain in the usual way the spin waves energy spectra in the approximation of non-interacting waves.

$$E_{\lambda r} = [P - (-1)^r R^{1/2}]^{1/2}, \tag{17}$$

where

$$P = \tfrac{1}{2}(\alpha_1^2 + \alpha_2^2) + \alpha_{\lambda 3}^2 - \tfrac{1}{2}(\beta_1^2 + \beta_2^2) - \beta_{\lambda 3}, \tag{18}$$

$$\begin{aligned} R = \tfrac{1}{4}[(\alpha_1^2 - \alpha_2^2) - (\beta_1^2 - \beta_2^2)]^2 - \beta_{\lambda 3}^2(\alpha_1 - \alpha_2)^2 - \alpha_{\lambda 3}^2(\beta_1 - \beta_2)^2 + (\alpha_2\alpha_{\lambda 3} - \beta_1\beta_{\lambda 3})^2 \\ + (\alpha_1\alpha_{\lambda 3} - \beta_2\beta_{\lambda 3})^2 + 4(\alpha_1\alpha_{\lambda 3} - \beta_1\beta_{\lambda 3})(\alpha_2\alpha_{\lambda 3} - \beta_2\beta_{\lambda 3}). \end{aligned} \tag{19}$$

Thus, the Hamiltonian reads

$$H = E_0 + E_D + \sum_{\lambda} (E_{\lambda 1} c_{\lambda 1}^{+} c_{\lambda 1} + E_{\lambda 2} c_{\lambda 2}^{+} c_{\lambda 2}), \tag{20}$$

where E_D [15] is given by

$$E_D = -1/2 \sum_{\lambda} \sum_{r=1}^{2} (\alpha_r - E_{\lambda r}), \tag{21}$$

and α_r is expressed by (10) and (11). The operators, $c_{\lambda 1}$ and $c_{\lambda 2}$, fulfil boson commutation rules similarly as the a_λ and b_λ operators.

One can find that the condition for the spectra (17) to real for all wave vectors, λ, is in agreement with the sufficient condition for the minimum of the approximate ground state [6, 16]. Thus, the disappearance of the linear terms of H_1 (7) (8) and conditions for the spectra to be real (17) to the same solutions and critical curves are the necessary and sufficient conditions for the minimum of the approximate ground state [6, 16].

By applying standard methods [14] to our Hamiltonian and also using the results [6, 16] one can obtain the two branches of the spin waves energy spectra for the field parallel and perpendicular to the easy axis and in particular the magnetic phases (AF, CS, SF, P) discussed in [6, 16]. In the present paper we discuss the dependence of the spin waves spectra on δ_λ. Table I presents the reduced coefficient, e.g. $\tilde{\alpha}_S = \alpha_S/\gamma_0 JS$, in the spectrum. Using this table we can express the spin waves energy spectrum in the AF phase as:

$$\begin{aligned} (\mathscr{E}_{\lambda r}^{\mathrm{AF}})^2 = (E_{\lambda r}/\gamma_0 JS)^2 = (X - a + l_x)^2 + h_x^2 - l_z^2 - Z\delta_\lambda^2 \\ -(-1)^r\{4(X - a + l_x)^2 h_x^2 + \delta_\lambda[[Z(X + 2l_x) - (X - 2a)]^2 - (Z+1)h_x^2]\}^{1/2}. \end{aligned} \tag{22}$$

The schematic dependence of $\mathscr{E}_{\lambda r}$ on δ_λ for the AF phase is shown in Fig. 1. It can be seen that when the value of h_x increases, the branches recede from each other and at the critical value, h_1^x, the distance between them is the longest. The branches, $\mathscr{E}_{\lambda 2} = 0$, for $\delta_\lambda = 1$

TABLE I

	Phase SF $h_2^x \leqslant h_x \leqslant h_3^x,\ Q_x = \dfrac{h_x}{h_3^x}$	Phase SF $0 \leqslant h_z \leqslant h_1^z,\ Q_z = \dfrac{h_z}{h_1^z}$	Phase SF $0 \leqslant h_x \leqslant h_1^x$	Phase P $h_3^x \leqslant h_x < \infty$	Phase P $h_1^z \leqslant h_z < \infty$
$\tilde{\alpha}_r$	$Z+a+l_z-aQ_x^2$	$X-a+l_x+aQ_z^2$	$X-a+l_x-(-1)^r h_x$	$-X-a+l_x+h_x$	$-Z+a+l_z+h_z$
$\tilde{\beta}_r$	$-l_x-aQ_x^2$	$-l_z+aQ_z^2$	$-l_z$	$-l_z$	$-l_x$
$\tilde{\alpha}_{3\lambda}$	$\tfrac{1}{2}\delta_\lambda[X-1-(X+Z)Q_x^2]$	$\tfrac{1}{2}\delta_\lambda[Z-1-(X+Z)Q_z^2]$	$\tfrac{1}{2}\delta_\lambda(Z-1)$	$-\tfrac{1}{2}\delta_\lambda(Z+1)$	$-\tfrac{1}{2}\delta_\lambda(X-1)$
$\tilde{\beta}_{3\lambda}$	$\tfrac{1}{2}\delta_\lambda[X+1-(X+Z)Q_x^2]$	$\tfrac{1}{2}\delta_\lambda[Z+1-(X+Z)Q_z^2]$	$\tfrac{1}{2}\delta_\lambda(Z+1)$	$-\tfrac{1}{2}\delta_\lambda(Z-1)$	$-\tfrac{1}{2}\delta_\lambda(X-1)$

	Phase CS $\quad h_1^x \leqslant h_x \leqslant h_2^x \quad b = (h_x-h_1^x)/4a(X-Z-2a)$
$\tilde{\alpha}_r$	$X-al_x+(Z-X+3a)b-\dfrac{ab(2b-1)h_x^2}{[X+Z+2a(2b-1)]^2}-(-1)^r\dfrac{h_x[X+Z-2a(1+b)]\sqrt{1-b}}{X+Z+2a(2b-1)]}\sqrt{1-\dfrac{h_x^2 b}{[X+Z+2a(2b-1)]^2}}$
$\tilde{\beta}_r$	$-l_z+ab-\dfrac{abh_x^2(2b-1)}{[X+Z+2a(2b-1)]^2}-(-1)^r\dfrac{2abh_x\sqrt{1-b}}{[X+Z+2a(2b-1)]}\sqrt{1-\dfrac{h_x^2 b}{[X+Z+2a(2b-1)]^2}}$
$\tilde{\alpha}_{3\lambda}$	$\tfrac{1}{2}\delta_\lambda\left[Z-1+(X-Z)b-\dfrac{(X+Z)h_x^2 b}{[X+Z+2a(2b-1)]^2}\right]$
$\tilde{\beta}_{3\lambda}$	$\tfrac{1}{2}\delta_\lambda\left[Z+1+(X-Z)b-\dfrac{(X+Z)h_x^2 b}{[X+Z+2a(2b-1)]^2}\right]$

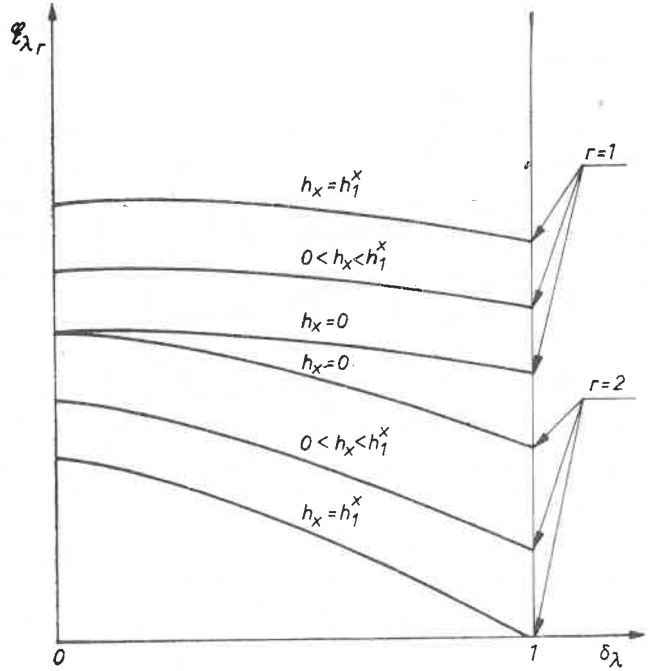

Fig. 1. The schematic dependence of $\mathscr{E}_{\lambda r}$ on δ_λ for the AF phase

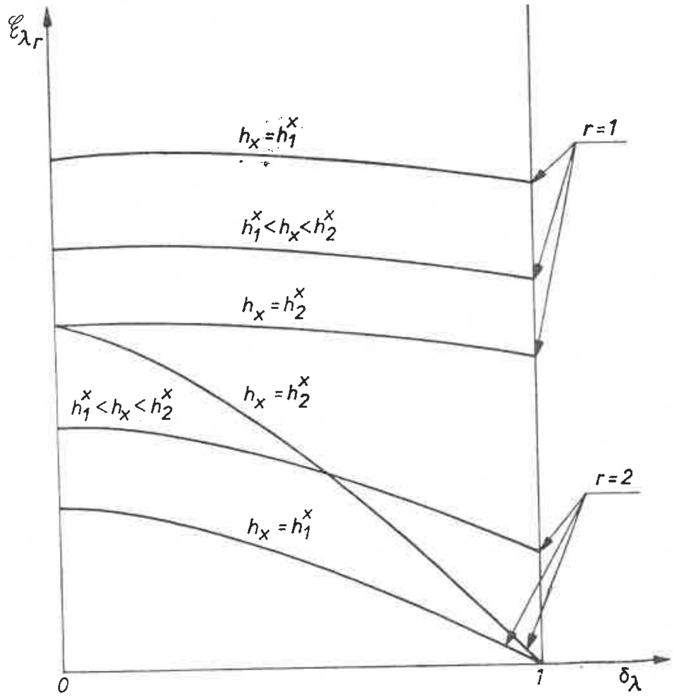

Fig. 2. The schematic dependence of $\mathscr{E}_{\lambda r}$ on δ_λ for the CS phase

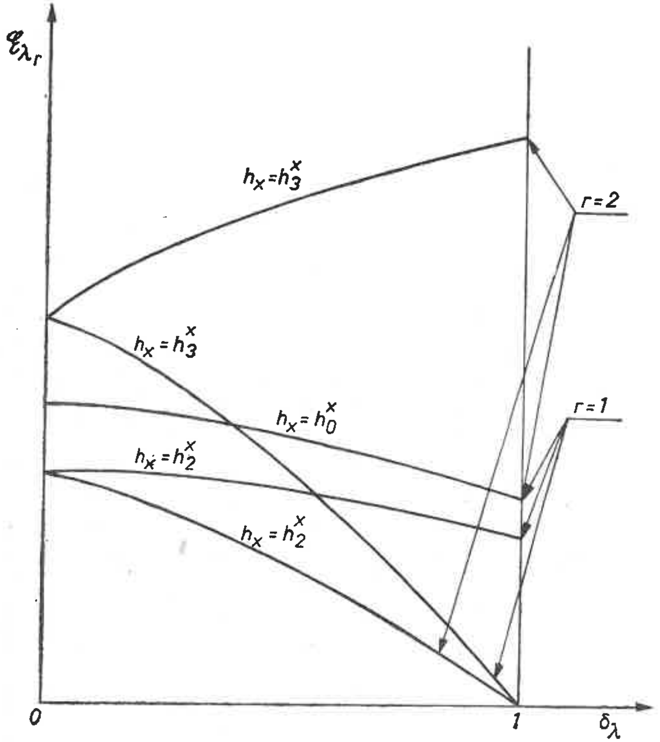

Fig. 3. The schematic dependence of $\mathscr{E}_{\lambda r}$ on δ_λ for the SF phase

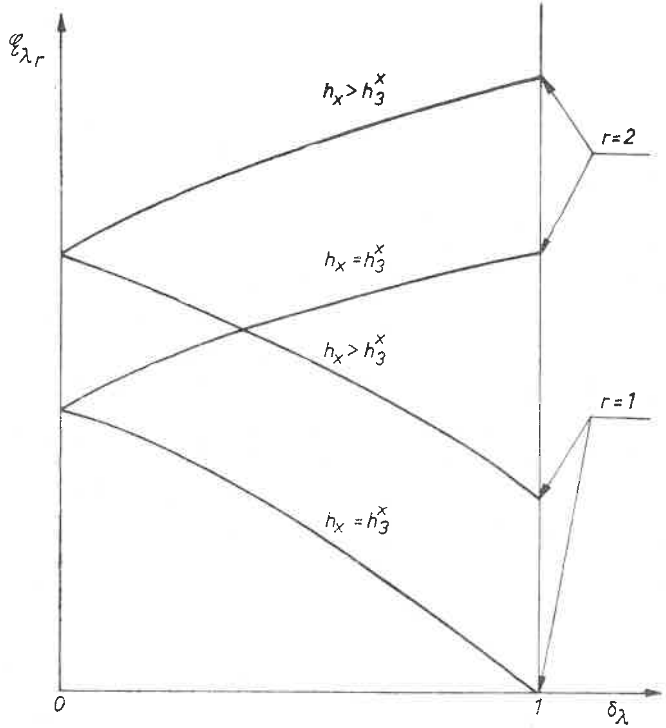

Fig. 4. The schematic dependence of $\mathscr{E}_{\lambda r}$ on δ_λ for the P phase

correspond to $\lambda = 0$. For $h_x > h_1^x$ $\mathscr{E}_{\lambda 2}$ becomes imaginary at $\delta_\lambda = 1$ (i.e. the AF phase is unstable).

The field dependence of the spectra for the CS phase in the field parallel to the easy axis is shown in Fig. 2. For $h_x = h_1^x$ the spectra in the CS phase are the same as in the AF phase. When the field is increasing, they approach each other and for $h_x = h_2^x$ (upper critical field corresponding to the CS ↔ SF transition) and at $\delta_\lambda = 0$ they are equal. At $\delta_\lambda = 1$ and for $h_x = h_2^x$, one of the branches, $\mathscr{E}_{\lambda 2}$ equals 0. The CS phase becomes unstable when the value of the field exceeds h_2^x the $\mathscr{E}_{\lambda 2}$ becomes imaginary. In the interval $h_2^x < h_x < h_3^x$ the SF phase is stable and the spectrum may be presented in the following way:

$$\mathscr{E}_{\lambda r}^{\mathrm{SF}} = [Z+2l_z+(-1)^r\delta_\lambda]^{1/2}\,\{Z+2a-2aQ_x^2-(-1)^r\delta_\lambda[X-(X+Z)Q_x^2]\}^{1/2}, \tag{23}$$

where $Q_x = h_x/h_3^x$, h_3^x is a critical field at the SF ↔ P transition. Fig. 3 presents the behavior of the spectrum for the SF phase. One sees that with the field increase the branches approach each other and when the field value is equal to

$$h_0^x = h_3^x\{[X(Z+2l_z)-(Z+2a)]/[(X+Z)\,(Z+2l_z)-2a]\}^{1/2} \tag{24}$$

they join. This effect is called a degeneration. With a further increase in the field both branches recede. When the field reaches a value of $h_x = h_3^x$, $\mathscr{E}_{\lambda 1}$ equals 0 and (at $\delta_\lambda = 1$) for the field $h_x > h_3^x$, it is unreal (for the fields $h_x < h_2^x$, $\mathscr{E}_{\lambda 2}$ was unreal). For the fields, $h_x > h_3^x$, the P phase is stable and its spectrum may be expressed as:

$$\mathscr{E}_{\lambda r}^{\mathrm{D}} = [h_x-X+2l_x+(-1)^r\delta_\lambda]^{1/2}\times[h_x-X-2a+(-1)^r Z\delta_\lambda]^{1/2}. \tag{25}$$

This spectrum is illustrated in Fig. 4. If the field is perpendicular to the easy axis, the spectra may be obtained by using formulas (23–25) through the application of the following changes: $X \leftrightarrow Z$, $h_x \leftrightarrow h_z$, $l_x \leftrightarrow l_z$.

The schematic plots given in Figs. 1–4 show that the minimum value of the spectra occur at $\delta_\lambda = 1$, which corresponds to $\lambda = 0$. Therefore, it seems reasonable to develop δ_λ with respect to λ in formulas (17) which enables the application of low temperature thermodynamics.

3. *Determination of the temperature dependence on thermodynamic quantities*

By using the diagonal form of the Hamiltonian (20) we determine the spin specific heat, C_S, the reduced magnetization components and the magnetic susceptibility tensor.

The thermodynamic potential for the Hamiltonian (20) is expressed as follows:

$$F = -kT\ln\operatorname{Tr}\exp(-H/kT) = E_0+E_D$$

$$+kT\sum_{\lambda}\sum_{r=1}^{2}\ln[1-\exp(-\mathscr{E}_{\lambda r}/\tau), \tag{26}$$

where k stands for the Boltzmann constant, $\tau = kT/\gamma_0 JS$. In a standard way we derive the formulas for the magnetization components, the parallel $m^{\parallel}$ and the perpendicular

$m^{\perp}$ to the direction of the external magnetic field:

$$m^{\parallel}(\tau) = m_0^{\parallel}(1+\tfrac{1}{2}S)-1/NS\sum_{\lambda}\sum_{r=1}^{2}[\langle n_r\rangle+\tfrac{1}{2}]\left(\frac{\partial\mathscr{E}_{\lambda r}}{\partial h}\right)_{\text{expl}}, \tag{27}$$

$$m^{\perp}(\tau) = m_0^{\perp}(1+\tfrac{1}{2}S)-1/NS\sum_{\lambda}\sum_{r=1}^{2}[\langle n_r\rangle+\tfrac{1}{2}]\frac{1}{h}\left(\frac{\partial\mathscr{E}_{\lambda r}}{\partial \gamma}\right)_{\text{expl}}, \tag{28}$$

where

$$\langle n_r\rangle = \sum_{\lambda}\sum_{r=1}^{2}\exp(-n\mathscr{E}_{\lambda r}/\tau) \tag{29}$$

and we introduced the polar coordinate system $h_x = h\sin\gamma$, $h_z = h\cos\gamma$. It is worth noting that the derivatives in (27) and (28) are calculated explicitly with respect to h and γ occurring in $\mathscr{E}_{\lambda r}$. One the other hand, the specific heat may be expressed as:

$$C_S = kd[\sum_{r=1}^{2}\mathscr{E}_{\lambda r}\langle n_{\lambda r}\rangle]/d\tau. \tag{30}$$

In order to make other calculations we pass from the sums to integrals by the following correspondence

$$\sum_{\lambda}\ldots \to (2\pi)^{-3}V\int\ldots d\lambda, \tag{31}$$

where V stands for the crystal volume. Then, we use the long wave approximation which has been already mentioned in the discussion of the spin wave energy spectrum. This discussion pointed out that at least one of the spectrum branches reaches the minimum for $\lambda = 0$. Therefore, we develop δ_λ into a series in the powers of λ. When assuming for simplicity the cubic lattice, we have

$$\delta_\lambda \approx 1-\tfrac{1}{6}(a_1\lambda)^2+\ldots, \tag{32}$$

where a_1 is (magnetic) lattice constant. By employing (32) to (17) we get

$$\mathscr{E}_{\lambda r} \approx [\mathscr{E}_{0r}^2+\Delta_r(a_1\lambda)^2]^{1/2}, \tag{33}$$

where

$$\mathscr{E}_{\lambda r} = \mathscr{E}_{\lambda r}(\delta_\lambda = 0).$$

Then by using the approximations (31)–(33), one obtains the dependence of the magnetization $m^t(t = \|;\perp)$, susceptibility, χ_t^t, on the temperature

$$\Delta m^{\parallel}(\tau) = \frac{1}{2\pi^2 S}\tau\sum_{r}^{2}\mathscr{E}_{0r}^2\Delta_r\left[\left(\frac{\partial\mathscr{E}_{0r}}{\partial h}\right)_{\text{expl}}\left\{\sum_{n=1}^{\infty}n^{-1}K_1(n\mathscr{E}_{0r}/\tau)\right\}\right.$$
$$\left.+\left(\frac{3\tau}{2\Delta_r}\right)\left(\frac{\partial\Delta_r}{\partial h}\right)_{\text{expl}}\left\{\sum_{n=1}^{\infty}n^{-2}K_2(n\mathscr{E}_{0r}/\tau)\right\}\right], \tag{34}$$

$$\Delta m^{\perp}(\tau) = \frac{1}{2\pi^2 S}\tau \sum_{r=1}^{2} \mathscr{E}^2_{0r}\Delta_r \left[\frac{1}{h}\left(\frac{\partial \mathscr{E}_{0r}}{\partial \gamma}\right)_{\text{expl}} \left\{\sum_{n=1}^{\infty} n^{-1}K_1(n\mathscr{E}_{0r}/\tau)\Big)\right\}\right.$$

$$\left. + \left(\frac{3\tau}{2\Delta_r}\right)\frac{1}{h}\left(\frac{\partial \Delta_r}{\partial \gamma}\right)_{\text{expl}} \left\{\sum_{n=1}^{\infty} n^{-2}K_2(n\mathscr{E}_{0r}/\tau)\right\}\right]. \tag{35}$$

The spin specific heat is expressed as

$$C_S = (1/2\pi^2\tau)\sum_{r=1}^{2} \mathscr{E}^2_{0r}\Delta_r^{-3/2}\left[\sum_{n=1}^{\infty} n^{-1}K_4(n\mathscr{E}_{0r}/\tau)\right], \tag{36}$$

where K_1, K_2, K_4 are the Bessel functions of the Mc Donald type.

We shall not specify here the explicit form of the coefficients in formulae (34–36) for all the magnetic phases, but confine ourselves to examining the behaviour of the thermodynamical quantities described by (34–36) in both cases: in the vicinity of the critical curves and far from them. In the first case, which corresponds to $n\mathscr{E}_{0r}/\tau \ll 1$, we have the following formulae for the magnetization components

$$\Delta m^{\parallel}(\tau) = (\tau^2/24S)\sum_{r=1}^{2} \Delta_r^{-3/2}\left(\frac{\partial \mathscr{E}^2_{0r}}{\partial h}\right)_{\text{expl}}, \tag{37}$$

$$\Delta m^{\perp}(\tau) = (\tau^2/24S)\sum_{r=1}^{2} \Delta_r^{-3/2}\,\frac{1}{h}\left(\frac{\partial \mathscr{E}^2_{0r}}{\partial \gamma}\right)_{\text{expl}}. \tag{38}$$

On the other hand, far from the critical curves ($n\mathscr{E}_{0r}/\tau \gg 1$), one obtains from (34–36) that

$$\Delta m^{\parallel}(\tau) = \frac{\tau^{3/2}}{S}\sum_{r=1}^{2}\left(\frac{\mathscr{E}_{0r}}{2\pi\Delta_r}\right)^{3/2}\cdot\left(\frac{\partial \mathscr{E}^2_{0r}}{\partial h}\right)_{\text{expl}}\cdot Z_{3/2}\left(\frac{\mathscr{E}_{0r}}{\tau}\right), \tag{39}$$

$$\Delta m^{\perp}(\tau) = \frac{\tau^{3/2}}{S}\sum_{r=1}^{2}\left(\frac{\mathscr{E}_{0r}}{2\pi\Delta_r}\right)^{3/2}\frac{1}{h}\left(\frac{\partial \mathscr{E}^2_{0r}}{\partial \gamma}\right)_{\text{expl}}\cdot Z_{3/2}\left(\frac{\mathscr{E}_{0r}}{\tau}\right), \tag{40}$$

where $Z_y(x) = \sum_{n=1}^{\infty} n^{-y}\cdot\exp(-nx)$. The specific heat for the case $n\mathscr{E}_{0r}/\tau \ll 1$ is

$$C_S = (12\pi^2/4S)\cdot\tau^3\cdot\sum_{r=1}^{2}\Delta_r^{-3/2}, \tag{41}$$

whereas, when $n\mathscr{E}_{0r}/\tau \gg 1$ one has

$$C_S = k\tau^{-1/2} \cdot \sum_{r=1}^{2} \left(\frac{\mathscr{E}_{0r}}{2\pi\Delta_r}\right)^{3/2} \cdot Z_{3/2}\left(\frac{\mathscr{E}_{0r}}{\tau}\right). \tag{42}$$

As concerns the magnetic susceptibility, near the critical curves its components read

$$\Delta\chi_{\parallel}^{\parallel}(\tau) = 3\pi^{-2}\zeta(2)\tau^2 \sum_{r=1}^{2} \mathscr{E}_{0r}^{-2}\Delta_r^{-3/2} \left(\frac{1}{24S}\,\frac{\partial\mathscr{E}_{0r}^2}{\partial h}\right)\left(\frac{1}{24S}\,\frac{\partial\mathscr{E}_{0r}^2}{\partial h}\right)_{\text{expl}}, \tag{43}$$

$$\Delta\chi_{\perp}^{\parallel}(\tau) = 3\pi^{-2}\zeta(2)\tau^2 \sum_{r=1}^{2} \mathscr{E}_{0r}^{-2}\Delta_r^{-3/2} \left(\frac{1}{24S}\,\frac{\partial\mathscr{E}_{0r}^2}{\partial h}\right)_{\text{expl}} \cdot \frac{1}{h}\left(\frac{\partial\mathscr{E}_{0r}^2}{\partial\gamma}\right), \tag{44}$$

$$\Delta\chi_{\perp}^{\perp}(\tau) = 3\pi^{-2}\zeta(2)\tau^2 \sum_{r=1}^{2} \mathscr{E}_{0r}^{-2}\Delta_r^{-3/2} \left(\frac{1}{24Sh}\,\frac{\partial\mathscr{E}_{0r}^2}{\partial\gamma}\right)_{\text{expl}} \cdot \frac{1}{h}\left(\frac{\partial\mathscr{E}_{0r}^2}{\partial\gamma}\right), \tag{45}$$

where $\zeta(y) = Z_y(0)$. On can easily prove that $\Delta\chi_{\parallel}^{\parallel}(r)$, $\Delta\chi_{\parallel}^{\perp}(\tau)$ and $\Delta\chi_{\perp}^{\perp}(\tau)$ tend to $+\infty$ because $\mathscr{E}_{0r}^{-2} \to \infty$ for $h(\gamma) \to h_{\text{cr}}(\gamma)$. This is in agreement with the experimental data [17].

4. *Final remarks*

It is worth while to emphasize that the formulae for thermodynamical quantities such as the magnetization, $m = (m^{\perp}(\tau)m^{\parallel}(\tau))$, susceptibility tensor, $\chi = (\chi_{\parallel}^{\parallel}(\tau)\chi_{\perp}^{\perp}(\tau)\chi_{\parallel}^{\perp}(\tau))$, and spin specific heat, $C_S(\tau)$, calculated in this paper are valid for the whole field range (Eqs. (34)–(36)) in contrast to the results obtained in [1–3], restricted either to the fields from the vicinity of the critical ones or to the fields far from them. Moreover, the Hamiltonian we use includes the transversal exchange and single-ion anisotropy (K_z and L_z in our notation) which is absent in the considerations of [1, 2]. Therefore, our temperature dependence of C_S, m and χ in the neigbourhood of the critical fields differs from that obtained in [1, 2]. In particular, this is clearly seen near the critical field h_3^x, separating the phase, P, from SF. According to our estimations the temperature dependence for the magnetization in this region is governed by the T^2-law, whereas according to [1, 2] by the $T^{3/2}$ one. The correspondence ($T^{3/2}$-law) can be obtained by putting $h_x = h_x^3$ and $K_z = L_z = 0$ into Eqs. (33). In this case the coefficient, Δ_r, is equal to zero, and therefore, a further term in the expansion of δ_λ (Eqs. (32)) in the power of λ is required. This leads to $\mathscr{E}_\lambda \sim \lambda^2$ and, consequently, to the $T^{3/2}$-law as in [1, 2]. From the physical point of view it is only important that the presence of a transversal anisotropy (exchange and or single-ion) yields the T^2-like dependence of the magnetization and susceptibility on the temperature instead of the $T^{3/2}$-law.

Though we do not present any dependence of the critical field h_3^x on the temperature, such a dependence can be obtained easily from formulae (34)–(35) which are similar to the formula for the critical curve in [18]. By putting $h_z = 0$ in that formula, one can conclude that h_3^x is proportional to T^2, which corresponds to the result of [11], while according to [1, 2] this dependence reads $\alpha T^{3/2}+\beta T^{5/2}$. In [9, 10] compounds with transversal anisotropy (exchange and single-ion) were investigated; it seems that the dependence of the critical field on the temperature such as T^2 is not excluded.

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