FLUCTUATIONS IN MAGNETIC MOMENT IN ANTIFERROMAGNETS

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The fluctuations in magnetic moment and the static spin pair correlation function in antiferromagnets has been obtained in the regions of temperatures closest to the critical point both below and above it. An antiferromagnet with spins $s = 1/2$ localized in the sites of the cubic lattices has been described by means of the constant coupling approximation of Kasteleijn and Kranendonk into which a uniaxial anisotropy caused by an exchange interaction had been introduced. For generalization this model to cover the nonequilibrium situation the mean effective field acting on a pair of spins has been replaced by local field dependent on the position in the lattice. The spatial distribution of the magnetic moment in a fluctuation, which is treated as a subsystem in a reservoir following from the ideas of Smoluchowski has been calculated by using the variational procedure for the work needed for the fluctuation to appear. This work has been obtained on the basis of the constant coupling approximation for two types of thermodynamic variables: magnetic moment and external field or magnetic moment and local field, which lead to the two correlation functions, namely $\exp(-K_1 r)/r$ form and $|\sin K r|/r$ form. The dependence on temperature and anisotropy of the correlation ranges K_{1x}^{-1} and K_{1z}^{-1} for the transverse and longitudinal components respectively (with respect to the direction distinguished by anisotropy) and of the parameters K_{2z}, K_{2z} has been discussed. The dependence on external magnetic field has been obtained for the longitudinal correlations only. The used method of calculations represents a generalization of paper by J. Kociński and L. Wojtczak (*J. Appl. Phys.*, **39**, 618 (1968)) to the temperatures below the Néel point and to the case of the anisotropic exchange interaction.

1. The constant coupling approximation of Kasteleijn and van Kranendonk with an anisotropic exchange interaction

The method of Kasteleijn and van Kranendonk has been originally formulated for the cubic two sublattices antiferromagnet [3]. The Hamiltonian of the system of N spins $s = \frac{1}{2}$ with the antiparallel exchange coupling between the nearest neighbour spins is given by

$$\hat{H} = 2J \sum_{\langle ij \rangle} \hat{S}_i \cdot \hat{S}_j - 2\mu B^z \sum_i \hat{S}_i^z. \tag{1}$$

The equilibrium properties of the system can be described in the terms of the representative pair of the nearest neighbour spins in the same way as in the case of a ferromagnet by intro-

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ducing the pair density matrix

$$\hat{\varrho}^{(2)} = \exp(-\beta\hat{H}_e)/\mathrm{Tr}\exp(-\beta\hat{H}_e), \tag{2}$$

where the effective Hamiltonian

$$\hat{H}_e = -2A_1\hat{S}_1\cdot\hat{S}_2 - 2A_2\hat{S}_1^z\hat{S}_2^z - \\ -2\mu A_3(\hat{S}_1^z+\hat{S}_2^z) - 2\mu A_4(\hat{S}_1^z-\hat{S}_2^z) \tag{3}$$

contains now the additional term connected with the effective field A_4 ,which alignes the spins from the different sublattices antiparallel to each other. In the so called constant coupling approximation the isotropic coupling constant A_1 is assumed to be equal to $(-J)$, $J>0$, the anisotropic one $A_2 = 0$ and A_3, A_4 are determined from the equilibrium conditions of the system.

We shall alter the Hamiltonian (3) so as to account for an anisotropic exchange interaction. The Hamiltonian of the system will be written in the form

$$\hat{H} = \sum_{\langle ij\rangle}[2J\hat{S}_i\cdot\hat{S}_j + 2J'\hat{S}_i^z\hat{S}_j^z] - 2\mu B^z\sum_i\hat{S}_i^z. \tag{4}$$

Consequently, the anisotropic coupling constant in the effective Hamiltonian will be put equal to $(-J')$

$$\hat{H}_e = 2J\hat{S}_1\cdot\hat{S}_2 + 2J'\hat{S}_1^z\hat{S}_2^z - 2\mu A_3(\hat{S}_1^z+\hat{S}_2^z) - \\ -2\mu A_4(\hat{S}_1^z-\hat{S}_2^z). \tag{5}$$

The presence of the anisotropic term does not change the eigenfunctions of $\hat{H}_e$

$$\Psi_0 = \cos\frac{\omega}{2}\varphi_0 + \sin\frac{\omega}{2}\varphi_2 \qquad \Psi_1 = \varphi_1$$

$$\Psi_2 = -\sin\frac{\omega}{2}\varphi_0 + \cos\frac{\omega}{2}\varphi_2 \qquad \Psi_3 = \varphi_3$$

where

$$\sin\omega = 2\mu A_4[A_1^2+4\mu^2A_4^2]^{\frac{1}{2}}, \qquad |\omega| \leqslant \frac{\pi}{2}$$

and the corresponding eigenvalues are

$$\begin{aligned}
\varepsilon_0 &= \tfrac{1}{2}(A_1+A_2) - [A_1^2+4\mu^2A_4^2]^{1/2}\\
\varepsilon_2 &= \tfrac{1}{2}(A_1+A_2) + [A_1^2+4\mu^2A_4^2]^{1/2}\\
\varepsilon_1 &= -\tfrac{1}{2}(A_1+A_2) - 2\mu A_3\\
\varepsilon_3 &= -\tfrac{1}{2}(A_1+A_2) + 2\mu A_3.
\end{aligned}$$

The internal energy and entropy of a pair are given by

$$E_{12} = \mathrm{Tr}[\hat{\varrho}^{(2)}\hat{H}^{(2)}] \tag{6}$$

$$S_{12} = -k_B\left\{\sum_{\nu=0}^{3} f_\nu \ln f_\nu + \frac{z-1}{z}\ln\left[4(1-S-s)^{\frac{1}{2}(S+s-1)}\times \right.\right. \\ \left.\left.\times(1-S+s)^{\frac{1}{2}(S-s-1)}(1+S+s)^{-\frac{1}{2}(S+s+1)}(1+S-s)^{-\frac{1}{2}(S-s+1)}\right]\right\}, \tag{7}$$

where

$$\hat{H}^{(2)} = 2J\hat{S}_1 \cdot \hat{S}_2 + 2J'\hat{S}_1^z\hat{S}_2^z - 2\mu B^z z^{-1}(\hat{S}_1^z + \hat{S}_2^z) \tag{8}$$

and

$$S = \mathrm{Tr}\,[\hat{\varrho}^{(2)}(\hat{S}_1^z + \hat{S}_2^z)] = f_1 - f_3 \tag{9}$$

$$s = \mathrm{Tr}\,[\hat{\varrho}^{(2)}(\hat{S}_1^z - \hat{S}_2^z)] = (f_0 - f_2)\sin\omega \tag{10}$$

represent the long range order parameters, while f_ν denote the eigenvalues of $\hat{\varrho}^{(2)}$. The form of the equilibrium equations in this case is similar to that given in [3] (formulae (47)–(50)); The coupling constant J must be replaced by $(J'+J)$, S and s attain their values obtained with $A_2 \neq 0$.

$$S+s = \tanh\beta[\mu B^z - \tfrac{1}{2}z(J+J')\{\Phi(S,s,\beta) - \varphi(S,s,\beta)\}]$$

$$S-s = \tanh\beta[\mu B^z - \tfrac{1}{2}z(J+J')\{\Phi(S,s,\beta) + \varphi(S,s,\beta)\}],$$

where the functions Φ and φ are given by

$$\Phi = \beta^{-1}(J+J')^{-1}\left[2\beta\mu A_3 + \frac{1}{2}\ln\frac{(1-S-s)(1-S+s)}{(1+S+s)(1+S-s)}\right]$$

$$\varphi = \beta^{-1}(J+J')^{-1}\left[-2\beta\mu A_4 + \frac{1}{2}\ln\frac{(1+S+s)(1-S+s)}{(1-S-s)(1+S-s)}\right].$$

By introducing the new variables

$$x = \exp(\beta J) \qquad u = \exp(2\beta\mu A_3)$$

$$m = \exp(\beta J') \qquad y = \exp(2\beta\mu B^z)$$

$$\sin\omega = 2\mu A_4[J^2 + 4\mu^2 A_4^2]^{-1/2}$$

the equilibrium conditions take the form

$$y = u\left[\frac{W(1+uX_-)}{u+X_+}\right]^{z-1}$$

$$y = u\left[\frac{1+uX_+}{W(u+X_-)}\right]^{z-1},$$

where

$$X_\pm = \frac{1}{2}\left[(1\pm\sin\omega)vxm + \frac{xm}{v}(1\mp\sin\omega)\right]$$

$$W = x^{\frac{z}{z-1}}\tan\omega, \qquad v = x^{1/\cos\omega}.$$

The calculations based on the method of Kasteleijn and van Kranendonk lead to the equation determining the Néel point

$$2m_N(x_N^2-1)-\frac{z}{z-1}[m_N(x_N^2+1)+2]\ln x_N = 0, \tag{11}$$

which in the limit $A_2 \to 0$ is identical with the result obtained for an antiferromagnet with the isotropic exchange interaction [3],

$$2(x_N^2-1)-\frac{z}{z-1}(x_N^2+3)\ln x_N = 0$$

and in the limit $A_1 \to 0$ gives the well known result for the Ising antiferromagnet

$$J' = k_B T_N \ln\frac{z}{z-2}.$$

Equation (11) has two solutions (except for the case $A_1 = 0$). This means that there appears also the anti-Néel point [3]. We shall represent the exchange Hamiltonian for a pair in the form

$$\hat{H}_{ex} = 2jw(S_1^x S_2^x + S_1^y S_2^y) + 2jS_1^z S_2^z, \tag{12}$$

where the isotropic and anisotropic coupling constants have been replaced respectively by: $J \to jw$, $J' \to (1-w)j$; $w(\leqslant 1)$ is the parameter characterizing the anisotropy of the exchange interaction. The dependence on w of $k_B T_N/j$ is shown in Fig. 1 for s.c. and b.c.c. lattices.

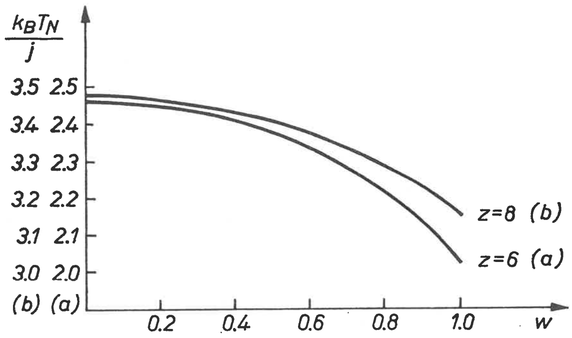

Fig. 1. Dependence of $k_B T_N/j$ on the anisotropy of the exchange interaction for s. c. and b. c. c. lattices

We shall consider the case of a vanishingly small external magnetic field perpendicular to the direction of easy magnetization. In this case

$$\hat{H} = \sum_{\langle ij\rangle}[2J\hat{S}_i\cdot\hat{S}_j + 2J'S_i^z S_j^z] - 2\mu B^x \sum_i \hat{S}_i^x \tag{13}$$

and we can write the effective Hamiltonian in the form

$$\hat{H}_e = -2A_1\hat{S}_1\cdot\hat{S}_2 - 2A_2\hat{S}_1^z\hat{S}_2^z - 2\mu A_3(\hat{S}_1^x+\hat{S}_2^x) - -2\mu A_4(\hat{S}_1^z-\hat{S}_2^z), \tag{14}$$

where $A_1 = -J$, $A_2 = -J'$. For a weak external magnetic field the parameter A_3 can be treated as small as compared with A_4 and J/μ. In the limit $B^x \rightarrow 0$, A_3 goes to zero. The parameter A_4 is connected with the spontaneous antiferromagnetic magnetization along the z direction. In the molecular field approximation the equilibrium equations are given by

$$\frac{S_x}{S} \ln \frac{1+S}{1-S} = 2\beta \left[\mu B^x - \frac{1}{2} zJS_x \right]$$

$$\frac{s_z}{S} \ln \frac{1+S}{1-S} = 2\beta \left[\frac{1}{2} z(J+J')s_z \right], \tag{15}$$

where

$$S = \sqrt{s_z^2 + S_x^2}$$

and in the constant coupling approximation with the anisotropic exchange interaction we have

$$\frac{S_x}{S} \ln \frac{1+S}{1-S} = 2\beta \left[\mu B^x + \frac{1}{2} zJ\Phi_x(\beta, S_x, S) \right]$$

$$\frac{s_z}{S} \ln \frac{1+S}{1-S} = 2\beta \left[\frac{1}{2} z(J+J')\Phi_z(\beta, s_z, S) \right], \tag{16}$$

where Φ_z and Φ_x are defined as

$$\Phi_z = \beta^{-1}(J+J')^{-1} \left[\frac{s_z}{S} \ln \frac{1+S}{1-S} - 2\beta\mu A_4 \right]$$

$$\Phi_x = (\beta J)^{-1} \left[\frac{S_x}{S} \ln \frac{1+S}{1-S} - 2\beta\mu A_3 \right]. \tag{17}$$

The quantities A_3 and A_4 are functions of the temperature and the long range order parameters S_x and s_z

$$S_x = \mathrm{Tr}\, [\hat{\varrho}^{(2)}(\hat{S}_1^x + \hat{S}_2^x)]$$

$$s_z = \mathrm{Tr}\, [\hat{\varrho}^{(2)}(\hat{S}_1^z - \hat{S}_2^z)]. \tag{18}$$

In order to determine the effective fields A_3 and A_4 we diagonalize the Hamiltonian H_e. We obtain the eigenvalue equation in the form

$$\Delta^3 - 2(A_1+A_2)\Delta^2 + [A_2(2A_1+A_2) - 4\mu^2(A_3^2 +$$

$$+ A_4^2)]\Delta + 4\mu^2 A_3^2(2A_1+A_2) = 0 \tag{19}$$

$$\Delta = \tfrac{1}{2}(A_1+A_2) + \varepsilon.$$

In the approximation $A_3 \ll A_4$ the eigenvalues are given by

$$\varepsilon_1 = \tfrac{1}{2}(J+J')$$

$$\varepsilon_3 = \tfrac{1}{2}(J+J')+4\mu^2 A_3^2(2J+J')\cos^2\omega[(J+J')^2\cos^2\omega-J^2]^{-1}$$

$$\varepsilon_0 = -\frac{1}{2}(J+J')-\frac{J}{\cos\omega}-2\mu^2 A_3^2(1-\cos\omega)\cos\omega[J+(J+J')\cos\omega]^{-1}$$

$$\varepsilon_2 = -\frac{1}{2}(J+J')+\frac{J}{\cos\omega}+2\mu^2 A_3^2(1+\cos\omega)\cos\omega[J-(J+J')\cos\omega]^{-1}.$$

For S_x and s_z we find

$$S_x = -(2\mu)^{-1}\sum_\nu f_\nu \frac{\partial\varepsilon_\nu}{\partial A_3} = 2\mu A_3\cos\omega\left\{\frac{1-\cos\omega}{J+(J+J')\cos\omega}f_0-\right.$$

$$\left.-\frac{1+\cos\omega}{J-(J+J')\cos\omega}f_2+\frac{2\cos\omega(2J+J')}{J^2-(J+J')^2\cos^2\omega}f_3\right\} \tag{20}$$

$$s_z = -(2\mu)^{-1}\sum_\nu f_\nu \frac{\partial\varepsilon_\nu}{\partial A_4} = \sin\omega\left\{f_0\left(1+\right.\right.$$

$$\left.+2\mu^2 A_3^2\cos^2\omega\frac{[(J+J')\cos^2\omega+2J\cos\omega]-J}{J[(J+J')\cos\omega+J]^2}\right)$$

$$-f_2\left(1-2\mu^2 A_3^2\cos^2\omega\frac{[-(J+J')\cos^2\omega+2J\cos\omega]+J}{J[-(J+J')\cos\omega+J]^2}\right)-$$

$$\left.-f_3 8\mu^2 A_3^2(2J+J')J\cos^3\omega[(J+J')^2\cos^2\omega-J^2]^{-2}\right\} \tag{21}$$

and the effective fields A_3 and A_4 satisfy the relations:

$$A_3 = B^x+\tfrac{1}{2}(z-1)\mu^{-1}J\Phi_x$$

$$A_4 = \tfrac{1}{2}(z-1)\mu^{-1}(J+J')\Phi_z. \tag{22}$$

2. Fluctuations in molecular field in the constant coupling approximation with the anisotropic exchange interaction

In the case of the longitudinal fluctuations we can write the effective Hamiltonian in the form

$$\hat{H}_{e||} = 2J\hat{S}_r\cdot\hat{S}_{r+\delta}+2J'\hat{S}_r^z\hat{S}_{r+\delta}^z-2\mu A_3(\hat{S}_r^z+\hat{S}_{r+\delta}^z)-$$

$$-2\mu A_4(\hat{S}_r^z-\hat{S}_{r+\delta}^z)-2\mu(c_r^z\hat{S}_r^z-c_{r+\delta}^z\hat{S}_{r+\delta}^z) \tag{23}$$

and for the transverse ones

$$\hat{H}_{e\perp} = 2J\hat{S}_r\cdot\hat{S}_{r+\delta}+2J'\hat{S}^z_r\hat{S}^z_{r+\delta}-2\mu A^1_3(\hat{S}^x_r+\hat{S}^x_{r+\delta})-$$

$$-2\mu A_4(\hat{S}^z_r-\hat{S}^z_{r+\delta})-2\mu(c^x_r\hat{S}^x_r-c^x_{r+\delta}\hat{S}^x_{r+\delta}) \tag{24}$$

where r and $r+\delta$ denote the two lattice sites occupied by the nearest neighbour spins, $c^z_r = c^z(r, 0)$ and $c^x_r = c^x(r, 0)$ are the fluctuations in the molecular field. In the calculations of the transverse fluctuations we shall limit ourselves to the case of the external magnetic field equal to zero and accordingly we put

$$\hat{H}_{e\perp} = 2J\hat{S}_r\cdot\hat{S}_{r+\delta}+2J'\hat{S}^z_r\hat{S}^z_{r+\delta}-2\mu A_4(\hat{S}^z_r-\hat{S}^z_{r+\delta})-$$

$$-2\mu(c^x_r\hat{S}^x_r-c^x_{r+\delta}\hat{S}^x_{r+\delta}). \tag{25}$$

For the longitudinal fluctuations the eigenfunctions of $\hat{H}_{e||}$ have the same form as in the equilibrium conditions but ω is now dependent on the fluctuation in the molecular field

$$\sin\omega = \frac{2\mu A_4+\mu\sum c^z}{[A_1^2+(2\mu A_4+\mu\sum c^z)^2]^{\frac{1}{2}}}$$

and the eigenvalues of $H_{e||}$ are given by

$$\varepsilon_1 = -\tfrac{1}{2}(A_1+A_2)-2\mu A_3+\mu\delta c^z$$

$$\varepsilon_3 = -\tfrac{1}{2}(A_1+A_2)+2\mu A_3-\mu\delta c^z$$

$$\varepsilon_0 = \tfrac{1}{2}(A_1+A_2)-[A_1^2+(2\mu A_4+\mu\Sigma c^z)^2]^{1/2}$$

$$\varepsilon_2 = \tfrac{1}{2}(A_1+A_2)+[A_1^2+(2\mu A_4+\mu\Sigma c^z)^2]^{1/2}$$

where

$$\sum c^z = c^z_{r+\delta}+c^z_r, \qquad \delta c^z = c^z_{r+\delta}-c^z_r.$$

For the transverse fluctuations the effective Hamiltonian $H_{e\perp}$ in the representation of the singlet and triplet functions takes the form

$$\hat{H}_{e\perp} = \begin{bmatrix} -\frac{1}{2}(3J+J') & \mu\Sigma c^x/\sqrt{2} & -2\mu A_4 & -\mu\Sigma c^x/\sqrt{2} \\ \mu\Sigma c^x/\sqrt{2} & \frac{1}{2}(J+J') & \mu\delta c^x/\sqrt{2} & 0 \\ -2\mu A_4 & \mu\delta c^x/\sqrt{2} & \frac{1}{2}(J-J') & \mu\delta c^x/\sqrt{2} \\ -\mu\Sigma c^x/\sqrt{2} & 0 & \mu\delta c^x/\sqrt{2} & \frac{1}{2}(J+J') \end{bmatrix}$$

and the eigenvalues satisfy the following equation

$$\Delta^4+2(J+J')\Delta^3+[J'(2J+J')-4\mu^2A_4^2-$$

$$-\mu^2((\delta c^x)^2+(\Sigma c^x)^2)]\Delta^2-\mu^2[J'((\delta c^x)^2+$$

$$+(\Sigma c^x)^2)+2J(\delta c^x)^2]\Delta+\mu^4(\delta c^x)^2(\Sigma c^x)^2 = 0, \tag{26}$$

where

$$\Delta = -\tfrac{1}{2}(J+J')+\varepsilon$$

$$\sum c^x = c^x_{r+\delta}+c^x_r, \quad \delta c^x = c^x_{r+\delta}-c^x_r.$$

The approximate solutions with accuracy up to second order with respect to the powers of Σc^x or δc^x, are given by

$$\varepsilon_0 = -\frac{1}{2}(J+J')-\frac{J}{\cos\omega_0}-\frac{\mu^2}{2}\frac{[(\delta c^x)^2(1-\cos\omega_0)+(\sum c^x)^2(1+\cos\omega_0)]\cos\omega_0}{(1+\cos\omega_0)J+\cos\omega_0 J'}$$

$$\varepsilon_2 = -\frac{1}{2}(J+J')+\frac{J}{\cos\omega_0}+\frac{\mu^2}{2}\frac{[(\delta c^x)^2(1+\cos\omega_0)+(\sum c^x)^2(1-\cos\omega_0)]\cos\omega_0}{(1-\cos\omega_0)J-J'\cos\omega_0}$$

$$\varepsilon_{1,3} = \tfrac{1}{2}(J+J')+\tfrac{1}{2}\mu^2\{(J'+2J)(\delta c^x)^2+J'(\Sigma c^x)^2\pm(16\mu^2A_4^2+$$
$$+[J'(\Sigma c^x)^2-(J'+2J)(\delta c^x)^2)^{1/2}[J'(2J+J')-4\mu^2A_4^2]^{-1}$$

$$\varepsilon_1 \cong \tfrac{1}{2}(J+J')+\mu^2J'(\Sigma c^x)^2[J'(2J+J')-4\mu^2A_4^2]^{-1}$$

$$\varepsilon_3 \cong \tfrac{1}{2}(J+J')+\mu^2(2J+J')(\delta c^x)^2[J'(2J+J')-4\mu^2A_4^2]^{-1}$$

$$\sin\omega_0 = 2\mu A_4[J^2+4\mu^2A_4^2]^{-1/2}.$$

In ε_1 and ε_3 we have neglected under the square root the terms which contain the parameter A_4. This approximation is valid in the vicinity of the critical point and is the better the smaller the anisotropy.

We shall calculate the work necessary to create a fluctuation in magnetic moment for two pairs of thermodynamic variables: (A) magnetic moment and external magnetic field, (B) magnetic moment and local field. In the case (A) this work is given by the change in the Gibbs free energy ΔG[1]. In the case (B) this work is equal to[2]

$$\Delta\Phi = \Delta G'+I_0\Delta\mathscr{B}, \tag{27}$$

where $\Delta\mathscr{B}$ is the deviation of the effective magnetic field acting on a pair from its equilibrium value, I_0 is the equilibrium value of the magnetic moment of a pair and for the calculation of G' the internal energy has the form [1]

$$\mathscr{E}_{r,r+\delta} = \mathrm{Tr}\,[\hat{\varrho}^{(2)}_{||,\perp}\hat{H}^{(2)}_{||,\perp}]$$

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[1] According to the denotation used in paper [1] $G \equiv F'$ is a thermodynamic potential obtained as the Legendre transform of the internal energy $F' = U'[T] = F'(T, V, B)$.

[2] This work may be derived in another way; in the form $\Delta\Phi = \Delta G - \Delta I\Delta\mathscr{B}$, where ΔI is the deviation of the magnetic moment of a pair from its equilibrium value and the internal energy in G is given by the same expression as in the case (A): $E_{r,r+\delta} = \mathrm{Tr}\,[\hat{\varrho}^{(2)}_{||,\perp}\hat{H}^{(2)}]$. The second term in $\Delta\Phi$ represents the energy of magnetic field connected with the appearance of a fluctuation. This term must be subtracted from the increase in the thermodynamic potential due to the fact that the fluctuation is now treated as produced at the cost of the decrease of heat in the system and not by the action of a fictitious magnetic field. The thermodynamic potential G is taken at the temperature T of the fluctuation in temperature [12].

with

$$\hat{H}^{(2)}_{||,\perp} = \hat{H}^{(2)} - 2\mu(c_r^{z,x}\hat{S}_r^{z,x} - c_{r+\delta}^{z,x}\hat{S}_{r+\delta}^{z,x}) \tag{28}$$

and

$$\hat{H}^{(2)} = 2J\hat{S}_r \cdot \hat{S}_{r+\delta} + 2J'\hat{S}_r^z\hat{S}_{r+\delta}^z - 2\mu B^z z^{-1}(\hat{S}_r^z + \hat{S}_{r+\delta}^z). \tag{29}$$

In both cases (A) and (B) the entropy for a pair of spins is given by (7), where for the f_ν, S, s the nonequilibrium values must be inserted. In the case of the longitudinal fluctuations

$$S = S_z = \mathrm{Tr}\,[\hat{\varrho}^{(2)}_{||}(\hat{S}_r^z + \hat{S}_{r+\delta}^z)]$$
$$s = s_z = \mathrm{Tr}\,[\hat{\varrho}^{(2)}_{||}(\hat{S}_r^z - \hat{S}_{r+\delta}^z)] \tag{30a}$$

and for the transverse ones we have put

$$s = \sqrt{s_x^2 + s_z^2}, \quad S = \sqrt{S_x^2 + S_z^2},$$

where

$$s_\alpha = \mathrm{Tr}\,[\hat{\varrho}^{(2)}_{\perp}(\hat{S}_r^\alpha - \hat{S}_{r+\delta}^\alpha)]$$
$$S_\alpha = \mathrm{Tr}\,[\hat{\varrho}^{(2)}_{\perp}(\hat{S}_r^\alpha + \hat{S}_{r+\delta}^\alpha)] \tag{30b}$$

with $\alpha = x, z$ and

$$\hat{\varrho}^{(2)}_{||,\perp} = \exp(-\beta\hat{H}_{e||,\perp})/\mathrm{Tr}\exp(-\beta\hat{H}_{e||,\perp}). \tag{31}$$

Now we can calculate the spatial behaviour of a fluctuation in the molecular field.

Longitudinal fluctuations

A. The Gibbs free energy for a pair may be written in the form

$$G_{r,r+\delta} = G^{(0)}_{r,r+\delta} + [w_1(\sum c^z)(\delta c^z) + w_2(\sum c^z)^2 + w_3(\delta c^z)^2], \tag{32}$$

where $G^{(0)}_{r,r+\delta}$ is the part of the free energy which does not depend on the fluctuation in the molecular field, $w_i (i = 1, 2, 3)$ are functions of the temperature, external magnetic field and anisotropy. The work necessary for creating a fluctuation including z pairs of spins is given by

$$\mathscr{L}_r = \sum_\delta^z (G_{r,r+\delta} - G^{(0)}_{r,r+\delta}) = A(c_r^z)^2 + E(\nabla^2 c_r^z)c_r^z + D(\nabla c_r^z)^2,$$

$$A = 4zw_2, \qquad E = 2a^2(2w_2 + w_1), \qquad D = 2a^2(w_1 + w_2 + w_3),$$

where a is the lattice constant. We have passed to a continuous variable $\boldsymbol{r}$ and expanded $(\Sigma c)^2$ and $(\delta c)^2$ in Taylor series. The Euler-Lagrange equation for $\mathscr{L}_r$ leads to the following equation for the fluctuation

$$(\nabla^2 - k_{1z}^2)\, c^z(\boldsymbol{r}, 0) = 0$$
$$k_{1z}^{-2} = \frac{a^2}{2z}\left[\frac{w_3}{w_2} - 1\right] \tag{33}$$

and with the assumption of spherical symmetry we have

$$c^z(r,0) = c_0^z \frac{\exp(-k_{1z}r)}{r}.$$

$$w_2 = \beta\mu^2\alpha^{-2}xm\left\{(2\beta J)^{-1}(v-v^{-1})\alpha\cos^3\omega_0+\right.$$

$$+\frac{1}{2}[4xm+(u+u^{-1})(v+v^{-1})]\sin^2\omega_0-$$

$$\left.-\frac{1}{2}xm\frac{z-1}{z}\left[\frac{(p-q)^2}{1-(S_z^{(0)}+s_z^{(0)})^2}+\frac{(p+q)^2}{1-(S_z^{(0)}-s_z^{(0)})^2}\right]\right\}$$

$$w_3 = \frac{1}{2}\beta\mu^2\alpha^{-2}\left\{r-\frac{z-1}{z}\alpha^{-2}\left[\frac{(r-t)^2}{1-(S_z^{(0)}+s_z^{(0)})^2}+\frac{(r+t)^2}{1-(S_z^{(0)}-s_z^{(0)})^2}\right]\right\}$$

where

$$p = \alpha^{-1}(v-v^{-1})(u-u^{-1})\sin\omega_0$$

$$q = (\beta J)^{-1}\cos^3\omega_0(v-v^{-1})+\alpha^{-1}[4xm+(u+u^{-1})(v+v^{-1})]\sin\omega_0$$

$$r = 4+xm(v+v^{-1})(u+u^{-1})$$

$$t = xm(v-v^{-1})(u-u^{-1})\sin\omega_0$$

$$\alpha = xm(v+v^{-1})+u+u^{-1}$$

$$x = \exp(\beta J)\ \sin\omega_0 = 2\mu A_4[J^2+4\mu^2A_4^2]^{-1/2}$$

$$m = \exp(\beta J')\ \cos\omega_0 = J[J^2+4\mu^2A_4^2]^{-1/2}$$

$$u = \exp(2\beta\mu A_3)\ \ v = x^{1/\cos\omega_0}$$

$S_z^{(0)}$ and $s_z^{(0)}$ are the equilibrium values of the long range order parameters

$$S_z^{(0)} = \alpha^{-1}(u-u^{-1})$$

$$s_z^{(0)} = xm\alpha^{-1}(v-v^{-1})\sin\omega_0. \tag{34}$$

B. For the work connected with the appearance of a fluctuation (referred to one pair of spins) we obtain according to (27)

$$\Delta\Phi_{r,r+\delta} = \Delta G'_{r,r+\delta}+I_0\Delta\mathscr{B},$$

$$I\Delta\mathscr{B} = 2\mu\,\mathrm{Tr}\,[\hat{\varrho}_{||}^{(2)}(c_r^z\hat{S}_r^z-c_{r+\delta}^z\hat{S}_{r+\delta}^z)] =$$

$$= \mu(\textstyle\sum c^z s_z-\delta c^z S_z)$$

and

$$I_0\Delta\mathscr{B} = \mu[\Sigma c^z s_z^{(0)}-\delta c^z S_z^{(0)}].$$

Application of the variational procedure to $\mathscr{L}_r = \sum_{\delta}^{z} \Delta\Phi_{r,r+\delta}$ yields the following equation

$$(\nabla^2 + k_{2z}^2)\, c^z(r, 0) = 0$$

$$k_{2z}^{-2} = \frac{a^2}{2z}\left[1 - \frac{u_3}{u_2}\right] \tag{35}$$

with a $|\sin k_{2z}|/r$ type solution.

$$u_2 = \beta\mu^2\alpha^{-2}xm\left\{(2\beta J)^{-1}(v - v^{-1})\alpha\cos^3\omega_0 + \right.$$

$$+\frac{1}{2}[4xm + (u+u^{-1})(v+v^{-1})]\sin^2\omega_0 +$$

$$\left. +\frac{1}{2}xm\frac{z-1}{z}\left[\frac{(p-q)^2}{1-(S_z^{(0)}+s_z^{(0)})^2} + \frac{(p+q)^2}{1-(S_z^{(0)}-s_z^{(0)})^2}\right]\right\}$$

$$u_3 = \frac{1}{2}\beta\mu^2\alpha^{-2}\left\{r + \frac{z-1}{z}\alpha^{-2}\left[\frac{(r-t)^2}{1-(S_z^{(0)}+s_z^{(0)})^2} + \frac{(r+t)^2}{1-(S_z^{(0)}-s_z^{(0)})^2}\right]\right\}.$$

We shall discuss the dependence of k_{1z} and k_{2z} on temperature and anisotropy of the exchange interaction in the limit of $B \to 0$ for $KMnF_3$ which is characterized by $z = 6$

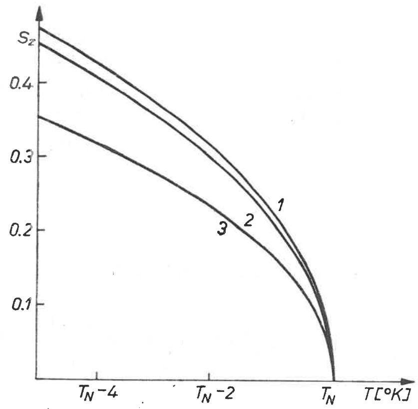

Fig. 2. Temperature dependence of the long range order parameter s_z (without external magnetic field). *1* — for $w = 0$, *2* — for $w = 0.5$, *3* — for $w = 1$

and $T_N = 88°K$ [7–9]. Without the external magnetic field $S_z^{(0)} = 0$ at all the temperatures and $s_z^{(0)}$ varies with temperature in the way illustrated in Fig. 2. The influence of the anisotropy upon the temperature dependence of k_{1z}^{-1} is shown in Fig. 3. k_{2z}^{-1} depends very little on temperature and anisotropy. This is presented in Table I.

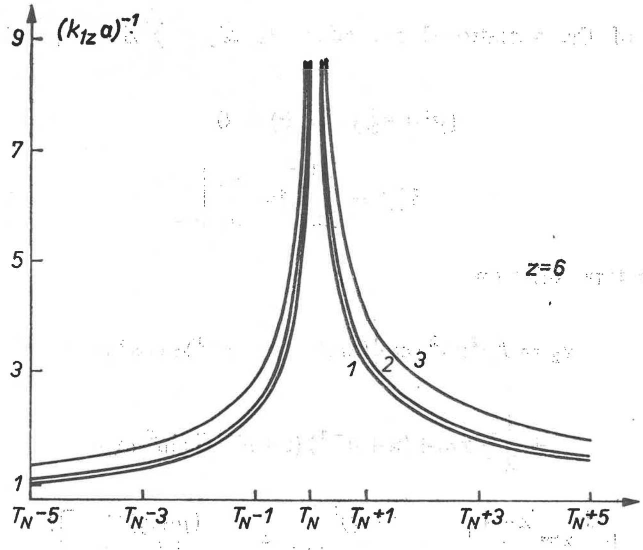

Fig. 3. Temperature dependence of the k_{1z}^{-1} for various values of the anisotropy parameter (in the limit $B=0$), *1* — for $w=0$, *2* — for $w=0.5$, *3* — for $w=1$

TABLE I

w	K_{2z}^{-1} for $z=6$		
	$T_N-5°$	$T_N\pm0.05°$	$T_N+5°$
0	0.184	0.192	0.189
0.1	0.185	0.192	0.189
0.5	0.189	0.196	0.192
0.9	0.204	0.206	0.202
1	0.210	0.211	0.207

Transverse fluctuations

In the case of the transverse fluctuations we get

$$\Delta G_{r,r+\delta} = \mu^2 \left\{ (\sum c^x)^2 h \left(1-4\frac{z-1}{z}\beta^{-1}h\right) + \right.$$
$$\left. +(\delta c^x)^2 f\left(1-4\frac{z-1}{z}\beta^{-1}f\right)\right\} \tag{36}$$

$$\Delta \Phi_{r,r+\delta} = \mu^2 \left\{ (\sum c^x)^2 h \left(1+4\frac{z-1}{z}\beta^{-1}h\right) + \right.$$
$$\left. +(\delta c^x)^2 f\left(1+4\frac{z-1}{z}\beta^{-1}f\right)\right\} \tag{37}$$

where

$$f = \alpha^{-1} \cos\omega_0 [J^2 - (J+J')^2 \cos^2\omega_0]^{-1} [(2J+J')\cos\omega_0 - \tfrac{1}{2} xm \times$$

$$\times \{(J'+2J)\cos\omega_0 (v+v^{-1}) - (v-v^{-1}) [J' \cos^2\omega_0 + J(1+\cos^2\omega_0)]\}]$$

$$h = \alpha^{-1} \cos\omega_0 [J^2 - (J+J')^2 \cos^2\omega_0]^{-1} [J' \cos\omega_0 - \tfrac{1}{2} xm \times$$

$$\times \{J' \cos\omega_0 (v+v^{-1}) + (v-v^{-1}) [J' \cos^2\omega_0 - J(1-\cos^2\omega_0)]\}]$$

$$\mathscr{L}_r = A'(c_r^x)^2 + E' c_r^x (\nabla^2 c_r^x) + D'(\nabla c_r^x)^2 \tag{38}$$

and we obtain the following equations for the transverse fluctuations in the molecular field

$$(\nabla^2 - k_{1x}^2)\, c^x(r, 0) = 0$$

and

$$(\nabla^2 + k_{2x}^2)\, c^x(r, 0) = 0$$

where k_{1x} and k_{2x} are given by

$$k_{1x}^{-2} = \frac{a^2}{2z} \left[\frac{f\left(1 - 4\dfrac{z-1}{z}\beta^{-1} f\right)}{h\left(1 - 4\dfrac{z-1}{z}\beta^{-1} h\right)} - 1 \right], \tag{39}$$

$$k_{2x}^{-2} = \frac{a^2}{2z} \left[1 - \frac{f\left(1 + 4\dfrac{z-1}{z}\beta^{-1} f\right)}{h\left(1 + 4\dfrac{z-1}{z}\beta^{-1} h\right)} \right]. \tag{40}$$

The change of k_{1x} and k_{2x} with temperature for various values of the anisotropy parameter w is shown in Fig. 4 and in Table II. For an isotropic exchange interaction, $w = 1$, and in the region of temperatures above the Néel point the values of k_{1x} and k_{2x} are identical with those for the longitudinal fluctuations. In the limit $T \to T_N$ $k_{2x}^{-1} \approx 0.2a$ and $k_{1x}^{-1} \to \infty$ and below T_N k_{1x}^{-1} becomes infinite. In the case of an Ising type interaction, $w = 0$, k_{1x}^{-1} and k_{2x}^{-1} vanish for every temperature both below and above the critical point. In general k_{1x}^{-1} depends strongly on anisotropy and for all w attains its maximal value at the Néel point, which is finite except for $w = 1$. These results are similar to those obtained by Oguchi and Ono [5]. But the interpretation of the dependence of k_{1x} on temperature by means of transverse transition temperature $T_\perp$ introduced by Moriya [10] is not possible since in our case $T_\parallel = T_\perp = T_N$. Such an interpretation has been accepted in [11] to explain the experimental data. The curve $k_{1x} = k_{1x}(T)$ measured above T_N has been extrapolated to the region of temperatures below T_N and k_{1x} has attained zero at $T_\perp$.

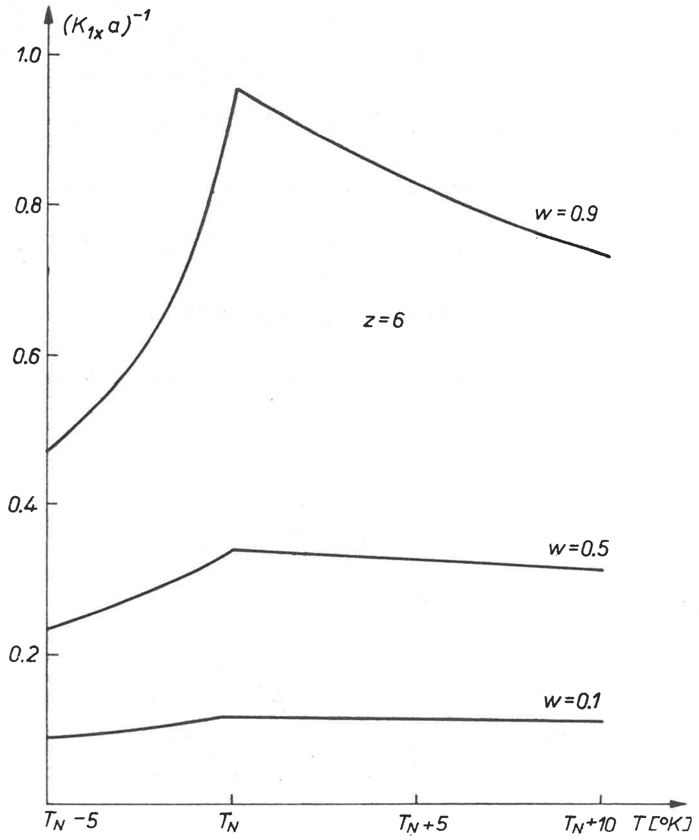

Fig. 4. Temperature dependence of the k_{1x}^{-1} for various values of the anisotropy parameter (in the case $B = 0$)

TABLE II

w	k_{2x}^{-1} for $z = 6$		
	$T_N - 5°$	$T_N \pm 0.05°$	$T_N + 5°$
0	0	0	0
0.1	0.070	0.071	0.069
0.5	0.151	0.152	0.148
0.9	0.200	0.200	—
1	0.212	0.211	0.207

This interpretation seems not to be adequate because it is the so-called staggered-susceptibility (which does not diverge), which is proportional to the cross-section and responsible for the behaviour of the correlation range k_{1x}^{-1} in the Ornstein-Zernike sense and not the susceptibility connected with the magnetization caused by a uniform magnetic field which has been discussed by Moriya.

3. Static spin pair correlation functions

In order to calculate the spin pair correlation functions we shall use the approximate relation [6]

$$\langle \hat{S}_0^\alpha(0)\hat{S}_r^\alpha(0)\rangle = \tfrac{1}{4}[n_0^+ - n_0^-][n_r^+ - n_r^-], \tag{41}$$

where $n^{+,-}$ denote the probabilities of finding the spins at 0, r aligned parallel or anti-parallel to the $\alpha = x, y, z$ direction.

Longitudinal components

The probabilities n_r for the two sublattices have been obtained in [4] (formulae (9), (11)–(13)) and they have been expressed in the terms of the fluctuations in magnetic moment of the sublattices

1)
$$n_r^+ - n_r^- = f_1 - f_3 + (f_0 - f_2)\sin\omega =$$
$$= S_z + s_z = 2\overline{S_r^z} = \mu^{-1} M_r^z$$

2)
$$n_{r+\delta}^+ - n_{r+\delta}^- = f_1 - f_3 - (f_0 - f_2)\sin\omega =$$
$$= S_z - s_z = 2\overline{S_{r+\delta}^z} = \mu^{-1} M_{r+\delta}^z, \tag{42}$$

where M_r^z and $M_{r+\delta}^z$ are the fluctuations in magnetic moment at the lattice sites $\boldsymbol{r}$ and $\boldsymbol{r}+\boldsymbol{\delta}$.

The form of the relations (42) does not change in our case. They differ only in the values of the S_z and s_z parameters, which now are given by

$$S_z = -2\alpha^{-2}\beta\mu x m\,(v - v^{-1})\,(u - u^{-1})\sin\omega_0\, c_r^z$$

$$s_z = 2xm\beta\mu\alpha^{-2}\{(\beta J)^{-1}\alpha\,(v - v^{-1})\cos^3\omega_0 +$$
$$+\sin^2\omega_0\,[4xm + (u + u^{-1})\,(v + v^{-1})]\}\, c_r^z \tag{43}$$

in the approximation of the homogeneous fluctuations that means $\delta c_r^z \to 0$. Because of the nonvanishing external magnetic field B^z the sublattices are not equivalent but in the limit $B^z \to 0$ ($u \to 1$ and $S_z \to 0$) we have $M_r^z = -M_{r+\delta}^z$. Taking into account the distribution of the magnetic moment in the fluctuation [2] we obtain for a single sublattice

$$\langle \hat{S}_0^z(0)\hat{S}_r^z(0)\rangle = (A_1^z)^2\,\frac{1}{4}\,\delta\,\frac{\exp[-k_{1z}(r-\delta)]}{r}$$

$$\langle \hat{S}_0^z(0)\hat{S}_r^z(0)\rangle = (A_2^z)^2\,\frac{1}{4}\,\frac{|\sin k_{2z}r|}{k_{2z}r}. \tag{44}$$

Transverse components

The matrix elements of $\hat{\varrho}_{\perp}^{(2)}$ in the spin product representation referred to the x axis needed for calculating the probabilities n_r are given by

$$\varrho_{+-,+-} = \frac{1}{2}\left(f_0 \cos^2 \frac{\omega_0}{2} + f_2 \sin^2 \frac{\omega_0}{2} + f_1\right) - \frac{1}{2} s_x$$

$$\varrho_{++,++} = \frac{1}{2}\left(f_0 \sin^2 \frac{\omega_0}{2} + f_2 \cos^2 \frac{\omega_0}{2} + f_3\right) - \frac{1}{2} S_x$$

$$\varrho_{-+,-+} = \frac{1}{2}\left(f_0 \cos^2 \frac{\omega_0}{2} + f_2 \sin^2 \frac{\omega_0}{2} + f_1\right) + \frac{1}{2} s_x$$

$$\varrho_{--,--} = \frac{1}{2}\left(f_0 \sin^2 \frac{\omega_0}{2} + f_2 \cos^2 \frac{\omega_0}{2} + f_3\right) + \frac{1}{2} S_x,$$

where s_x and S_x are equal to

$$s_x = 2\mu(\Sigma c^x)\,\alpha^{-1} \cos\omega_0\,[J^2-(J+J')^2 \cos^2\omega_0]^{-1}\{J'\cos\omega_0 -$$

$$-\tfrac{1}{2}xm\,[J'\cos\omega_0\,(v+v^{-1})+(v-v^{-1})\{J'\cos^2\omega_0-J(1-\cos^2\omega_0)\}]\}$$

$$S_x = -2\mu(\delta c^x)\,\alpha^{-1}\cos\omega_0[J^2-(J+J')^2\cos^2\omega_0]^{-1}\{(2J+J')\cos\omega_0 -$$

$$-\tfrac{1}{2}xm\,[(J'+2J)\cos\omega_0\,(v+v^{-1})-(v-v^{-1})\{J'\cos^2\omega_0+J(1+\cos^2\omega_0)\}]\}$$

and we obtain for each of the sublattices, respectively, the following expressions in the limit $\delta c^x \to 0$

1) $n_r^+ - n_r^- = s_x = \mu^{-1}M_r^x$

2) $n_{r+\delta}^+ - n_{r+\delta}^- = -s_x = -\mu^{-1}M_{r+\delta}^x$

and for the spin pair correlation function

$$\langle \hat{S}_0^x(0)\hat{S}_r^x(0)\rangle = (A_1^x)^2\,\frac{1}{4}\,\delta\,\frac{\exp[-k_{1x}(r-\delta)]}{r}$$

$$\langle \hat{S}_0^x(0)\hat{S}_r^x(0)\rangle = (A_2^x)^2\,\frac{1}{4}\,\frac{|\sin k_{2x}r|}{k_{2x}r}. \tag{45}$$

The equations (44), (45) for the spin pair correlation function are valid for all r including the limiting value $r \to 0$. Consequently, the solutions without spherical symmetry like those discussed in [13] also exist. The relevant discussion may be performed for temperatures below the critical one and also in the case of an antiferromagnet. This will be done in a separate paper.

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