ON THE DECAY OF THERMODYNAMIC FLUCTUATIONS IN ANTIFERROMAGNETIC ORDER

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The decay of thermodynamic fluctuations in antiferromagnetic order has been investigated on the basis of the master equation. The calculations have been performed in the constant coupling approximation for Heisenberg antiferromagnet with spin $s = 1/2$ at the lattice site, generalized to cover the nonequilibrium conditions, analogously to the case of ferromagnets. The decay appears to be described by the diffusion equation for the magnetic moment fluctuation in each sublattice separately. The diffusion coefficient changes very slowly with temperature in the vicinity and above the Néel point and has a nonzero value at T_N.

I. Introduction

The decay of fluctuations in antiferromagnetic order has been investigated theoretically by De Gennes and Villain (1960) and by Halperin and Hohenberg (1967). However, the results of these papers cannot be treated as conclusive. Our calculation will be based on the master equation as in the ferromagnetic case (Kociński 1963, 1964, 1966) and will lead to the description of the decay exactly analogous to that of the ferromagnetic fluctuations.

II. Method of calculation

We shall consider cubic two sublattice Heisenberg antiferromagnets of N spins $s = 1/2$, at the equilibrium temperature above or at the Néel point T_N. A fluctuation in antiferromagnetic order is treated as a subsystem of N_1 spins in the reservoir of $N-N_1$ spins (Ko-

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ciński and Wojtczak 1968). A fluctuation is described by the set of values of magnetic moment $M_A^z(\vec{r}_i, t)$, $M_B^z(\vec{r}_i + \vec{\varrho}, t)$ $i = 1, 2, \ldots N_1/2$ in the A and B sublattices, with the condition:

$$M_B^z(\vec{r}_i + \vec{\varrho}, t) = -M_A^z(\vec{r}_i, t) \tag{1}$$

where $\vec{\varrho}$ is a constant vector, which connects two nearest neighbour spins. The magnetic moments represent average values over an ensemble of fluctuations, which is realized in the spin system. A fluctuation in antiferromagnetic order is treated as a superposition of two fluctuations in magnetic moment each in one sublattice. The simultaneous decay of magnetic moment fluctuations in each sublattice determines the decay of the fluctuation in antiferromagnetic order.

When the deviation from equilibrium value at the time considered is large, a fluctuation almost surely decays. The degree of deviation from equilibrium is expressed by the so called abnormality of the fluctuation state taken as initial, which is measured by the ratio of this initial state to the root mean square fluctuation (for each Fourier component of the fluctuation). The decay of a fluctuation may be described in the terms of master equation for a pair of spins in the formulation of the constant coupling approximation (Kociński 1963, 1964, 1966). Two limiting cases will be considered, of very weak and very strong exchange interaction of a spin with its z nearest neighbours as compared with that with its ζ next nearest neighbours.

III. Weak coupling between sublattices

In the case of weakly coupled sublattices, the decay in time of magnetic moment in a single one is mainly determined by the exchange interaction between spins within it. This fluctuation in magnetic moment decays according to the diffusion equation of the ferromagnetic case (Kociński 1963, formula (30)) with the diffusion coefficient

$$\Lambda = \frac{4k_B T_0 d^2}{\zeta h} \frac{(1 - \eta^2)(1 + \eta^2 + \lambda^{-1} + \lambda)}{(1 + \eta^2)(\lambda^{-1} + \lambda) + 4} \tag{2}$$

where d is the distance to the nearest neighbour and ζ the number of nearest neighbours in a single sublattice, $\eta = \exp(-J_2/k_B T_0)$, $\lambda = \exp(-2\mu(B_m + B)/k_B T_0)$. The exchange integral J_2 for the nearest neighbour spin in a single sublattice is related with the Néel temperature by means of the ferromagnetic case relation:

$$k_B T_N \ln \frac{\zeta}{\zeta - 4} = 2J_2 \tag{3}$$

(vide formula (55) of Kasteleijn and Van Kranendonk 1956a). The field B_m in λ is the molecular field of a ferromagnetic sublattice to be determined from the constant coupling approximation (Kasteleijn and Van Kranendonk 1956a) and B denotes the external field. At the Néel temperature we get

$$\Lambda = \frac{8k_B^* T_N d^2}{\zeta^2 h} \tag{4}$$

The temperature dependence of Λ is the same as in a ferromagnet.

IV. Strong coupling between sublattices

The calculation will follow the course of the ferromagnetic case (Kociński 1963, 1964, 1966) and will be limited to temperatures above or at the Néel point and zero external magnetic field.

A pair of nearest neighbour spins in the fluctuation[1] is described by the effective Hamiltonian

$$\hat{\mathscr{H}}_e = 2J_1\,\hat{\vec{S}}_{A,r}\,\hat{\vec{S}}_{B,r+\alpha} - 2\mu\left[(|a^z_r|+b^z_r)\,\hat{S}^z_{A,r} - (|a^z_{r+\alpha}|+b^z_{r+\alpha})\,S^z_{B,r+\alpha}\right] \tag{5}$$

where $a^z_r = a^z(\vec{r},\,t)$ and $b^z_r = b^z(\vec{r},\,t)$ denote the fluctuations in the molecular fields of the A and B sublattices in the minus and plus z directions respectively, μ is Bohr's magneton, J_1 the exchange integral for the nearest neighbour spins which belong to different sublattices. The fields represent average values over an ensemble of fluctuations, realized in the spin system. We have to calculate the matrix elements of the pair density operator

$$\hat{\varrho}^{(2)} = \exp\left(-\beta\,\hat{\mathscr{H}}_e\right)/\mathrm{Tr}\,\exp\left(-\beta\,\hat{\mathscr{H}}_e\right),\; \beta = (k_B T_0)^{-1} \tag{6}$$

in the representation of the spin product functions $|++\rangle$, $|+-\rangle$, $|-+\rangle$, $|--\rangle$. To this end we observe that the eigenvalues of the Hamiltonian (5) are given by

$$\varepsilon_0 = -\frac{1}{2}J_1 - (J_1^2 + (\mu\Sigma c)^2)^{1/2}, \quad \varepsilon_2 = -\frac{1}{2}J_1 + (J_1^2 + (\mu\Sigma c)^2)^{1/2}$$

$$\varepsilon_1 = \frac{1}{2}J_1 - \mu\delta c, \quad \varepsilon_3 = \frac{1}{2}J_1 + \mu\delta c \tag{7}$$

with $c = |a| + b$, and Σc, δc denoting the sum and the difference of the fields at the adjacent lattice sites. The corresponding diagonal elements of the operator $\hat{\varrho}^{(2)}$ have the form:

$$f_\nu = e^{-\beta\varepsilon_\nu}(\Sigma_\nu e^{-\beta\varepsilon_\nu})^{-1}, \quad \nu = 0, 1, 2, 3. \tag{8}$$

Now, the matrix elements of $\hat{\varrho}^{(2)}$ in the spin product representation are equal to

$$\varrho_{+-,\,+-} = \frac{1}{2}[f_0 + f_2 + (f_0 - f_2)\sin\omega]$$

$$\varrho_{-+,\,-+} = \frac{1}{2}[f_0 + f_2 - (f_0 - f_2)\sin\omega] \tag{9}$$

$$\varrho_{++,\,++} = f_1, \quad \varrho_{--,\,--} = f_3, \quad \varrho_{-+,\,+-} = \varrho_{+-,\,-+} = \frac{1}{2}(f_2 - f_0)\cos\omega$$

with

$$\sin\omega = \mu\Sigma c(J_1^2 + (\mu\Sigma c)^2)^{-1/2} \tag{10}$$

The probabilities $n^{ik}_{r,r+\alpha}$ $(i, k = +, -)$ of finding the state of "$+-$" or "$-+$" spins at the lattice sites $\vec{r}$, $\vec{r}+\vec{\alpha}$ are determined by the diagonal elements in (9). The probabilities of

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[1] Note that the direction of $\vec{\alpha}$ varies, since $\vec{\alpha}$ runsover the nearest neighbours of a spin, while the direction of $\vec{\varrho}$ in (1) is constant.

finding a "+" or "−" spin at the lattice site $\vec{r}$ are given by the expressions

$$n_r^+ = n_{r,r+\alpha}^{++} + n_{r,r+\alpha}^{+-}$$

$$n_r^- = n_{r,r+\alpha}^{--} + n_{r,r+\alpha}^{-+} \tag{11}$$

These are not microscopic probabilities which refer to a particular pair of spins, but macroscopic probabilities, which refer to macroscopic states for which the spin is meant a classical vector.

The probabilities n will be expressed in terms of the fluctuations in magnetic moment in the sublattices by means of the relations

$$m_+^z(\vec{r},\ \vec{r}+\vec{\alpha},\ t) = 2\mu z^{-1}\ \mathrm{Tr}\ [(\hat{S}_r^z + \hat{S}_{r+\alpha}^z)\hat{\varrho}^{(2)}] = 2\mu z^{-1}S$$

$$m_-^z(\vec{r},\ \vec{r}+\vec{\alpha},\ t) = 2\mu z^{-1}\ \mathrm{Tr}\ [(\hat{S}_r^z - \hat{S}_{r+\alpha}^z)\hat{\varrho}^{(2)}] = 2\mu z^{-1}s \tag{12}$$

$$S = f_1 - f_3,\ s = (f_0 - f_2)\sin\omega \tag{13}$$

for the fluctuation in magnetic moment of the pair, which follow from the relations (33) and (34) of Kasteleijn and Van Kranendonk (1956b). The average value of magnetic moment of the spins at the points $\vec{r}$ and $\vec{r}+\vec{\alpha}$ treated as members of the pair are given by

$$m^z(\vec{r},\ t) = \mu z^{-1}(S+s)$$

$$m^z(\vec{r}+\vec{\alpha},\ t) = \mu z^{-1}(S-s) \tag{14}$$

while the values of the fluctuations in magnetic moment at the lattice sites $\vec{r}$ and $\vec{r}+\vec{\alpha}$ are z times larger (each spin belongs to z pairs) and denoted by

$$M^z(\vec{r},\ t) = zm^z(\vec{r},\ t)$$

$$M^z(\vec{r}+\vec{\alpha},\ t) = zm^z(\vec{r}+\vec{\alpha},\ t). \tag{15}$$

The expressions for the f_ν and $\sin\omega$ depend on the values of the molecular field at the points $\vec{r}$ and $\vec{r}+\vec{\alpha}$. We want to express the magnetic moment at the point $\vec{r}$ in the terms of the molecular fields at this point. In the limit $\delta|a|$, $\delta b \to 0$ we have $f_1 - f_3 = 0$ and we get the expressions

$$M_r^z(t) \simeq \mu(f_0 - f_2)\sin\omega = \frac{1-\eta^2}{1+3\eta^2}\frac{2\mu^2}{J_1}\ (|a_r^z| + b_r^z)$$

$$M_{r+\alpha}^z(t) \simeq \mu(f_2 - f_0)\sin\omega = \frac{\eta^2-1}{1+3\eta^2}\frac{2\mu^2}{J_1}\ (|a_{r+\alpha}^z| + b_{r+\alpha}^z) \tag{16}$$

with $\eta = \exp(-J_1/k_BT_0)$, which show that the magnetic moments in the two sublattices point in opposite directions. These relations enable the expression of the probabilities n in the terms of magnetic moments and the resulting formulas exhibit complete analogy with the ferromagnetic case, as far as the dependence on the sum and difference of the magnetic moments at the lattice sites $\vec{r}$ and $\vec{r}+\vec{\alpha}$ is concerned.

We can now pass over to the determination of the decay of the fluctuation in magnetic moment of a single sublattice. From the master equation for the change in time of the probability of finding a "+" or "−" spin at the point $\vec{r}$ we get the equation for the change

in time of the average value of the z component of the spin at point $\vec{r}$:

$$\frac{1}{2} \frac{\partial}{\partial t} (n_r^+ - n_r^-) = W(n_{r,\,r+\alpha}^{-+} - n_{r,\,r+\alpha}^{+-}) \tag{17}$$

where $W = W_{+-,\,-+} = W_{-+,\,+-}$ denotes the transition probability per unit of time from the $|+-\rangle$ to $|-+\rangle$ state.

It is emphasised that the probabilities n refer to macroscopic states and the transition probabilities to transitions between these macroscopic states (Van Kampen 1962). To the macroscopic spin product state corresponds the microscopic one, connected with the Hamiltonian (5) without the exchange term, in which the fields a, b are now treated as external fields acting on the pair, and not averages over an ensemble of fluactuations. The relevant $|+-\rangle$ and $|-+\rangle$ eigenstates are degenerated with energy $\varepsilon = 0$ in the limit of $\delta|a|, \delta b \to 0$. The exchange term $2J_1 \vec{S}_1 \cdot \vec{S}_2$ is now introduced as a perturbation which acts between antiparallel spins. It causes transitions between the $|+-\rangle$ and $|-+\rangle$ eigenstates. The transition time is determined from the time dependent $|+-\rangle$ (or $|-+\rangle$) state:

$$|+-\rangle_t = \frac{1}{\sqrt{2}} (\varphi_0 \, e^{i3J_1 t/2\hbar} + \varphi_2 \, e^{-iJ_1 t/2\hbar}) = e^{iJ_1 t/2\hbar} \left(\cos \frac{J_1 t}{\hbar} \, |+-\rangle + i \sin \frac{J_1 t}{\hbar} \, |-+\rangle \right) \tag{18}$$

where

$$\varphi_0 = \frac{1}{\sqrt{2}} (|+-\rangle - |-+\rangle), \qquad \varphi_2 = \frac{1}{\sqrt{2}} (|+-\rangle + |-+\rangle)$$

are the perturbed eigenfunctions of the pair for $\delta|a|, \delta b \to 0$. The pair is in the $|+-\rangle$ state at $t = 0$ and passes to the $|-+\rangle$ state after the time $\tau = h/4J_1$. The transition probability W is defined as τ^{-1}.

We now take the formulas (9) and (11) for the probabilities expressed in the terms of the magnetic moment fluctuations, expand them in series with respect to δM and ΣM and insert into the equation (17) together with the calculated value for W. Having taken an average over the z nearest neighbours of the spin at $\vec{r}$ we get the diffusion equation for the decay of the fluctuation in the magnetic moment of a single sublattice with the diffusion coefficient

$$\Lambda = \frac{2k_B T_0 a^2}{zh} (\eta^{-2} - 1), \tag{19}$$

where a denotes the lattice constant, and the exchange integral J_1 is related to the Néel temperature T_N by the condition (69) of Kasteleijn and Van Kranendonk (1956b):

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

For $z = 8$ one gets from (19) at the Néel point and at infinite temperature respectively:

$$\Lambda(T_N) = 0.22 \frac{k_B T_N a^2}{h} \tag{20}$$

$$\Lambda(\infty) = \frac{4J_1 a^2}{zh} = 0.75 \, \Lambda(T_N) \tag{21}$$

Contrary to the case of weak coupling between the sublattices and to the ferromagnetic one, for strong coupling the diffusion coefficient diminishes with growing temperature. The relation

$$\Lambda(2T_N) = 0.86\Lambda(T_N) \qquad z = 8 \tag{22}$$

indicates the degree of bending down of the $\Lambda(T)$ curve.

Comparison of the diffusion coefficients for a model with weak coupling and strong coupling between the two sublattices and the same Néel temperature shows that

$$\Lambda \text{ (weak coupling)} \geqslant \Lambda \text{ (strong coupling)}$$

where the equality sign corresponds to Néel point. It may be expected that a model which takes into account both the interactions between nearest and next nearest neighbours, will lead to intermediate values of the diffussion coefficient. However, the net result will be that above the Néel point the diffusion coefficient changes very little with temperature for ranges of temperature of experimental interest. The situation is completely analogous with that of the ferromagnetic case.

V. Conclusion

The description of the decay is macroscopic in character, because it starts from the master equation and uses the concept of molecular field. The transitions which appear in the master equation refer to macroscopic states and their calculation by means of spin reversal represents a simplification. However, the transition probability W contributes only a constant factor to the diffusion coefficient and does not affect its temperature dependence.

We have not discussed the influence of the fluctuations in temperature in the crystal lattice on the fluctuations in antiferromagnetic order. This effect is essential in ferromagnets for the building up as well as for the decay of the fluctuation. It undoubtedly exists in antiferromagnets and its investigation would be necessary.

Thus far the diffusion coefficient in antiferromagnets has not been determined experimentally. The present calculation enables a confrontation of the theory of critical scattering previously presented (Kociński 1968) with the results of future measurements.

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