FLUCTUATIONS OF MAGNETIC MOMENT AND THE SPECIFIC HEAT OF AN ANTIFERROMAGNET NEAR THE CRITICAL TEMPERATURE

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The magnetic specific heat near the critical temperature was calculated for an isotropic cubic antiferromagnet with 1/2 spins, for $T \geqslant T_N$. The free energy of the system was calculated in the constant coupling approximation taking into account the influence of fluctuations of magnetic moment. A nonlinear differential equation for the distribution in space of magnetic moment was used. The model consists in the division of the system into N_f equal cells each of which has a volume equal to the mean volume of a fluctuation. As a result the effect of rounding of the specific heat peak at its maximum was obtained. A comparison with the experimental data is presented.

1. *Introduction*

It is a well known fact that the method of critical indices, which is strongly supported by scaling hypothesis, describes very well the behaviour of magnetic specific heat only with a certain interval of reduced temperature $\varepsilon = |T-T_c|/T_c$. It means that a satisfactory fit of experimental data to the asymptotic form

$$C_M = (A^{\pm}/\alpha^{\pm})\varepsilon^{-\alpha^{\pm}} + B^{\pm}, \tag{1}$$

where A^+, α^+, B^+ and A^-, α^-, B^- are constants for $T \geqslant T_c$ and $T \leqslant T_c$ correspondingly, can be achieved only in the range $10^{-3} \leqslant \varepsilon \leqslant 10^{-1}$. So, as the experimental technique allows us to measure the reduced temperature up to the values $\varepsilon = 10^{-5} \div 10^{-6}$, two or three decades in ε are rejected as "not critical" in the typical analysis of the experimental data.

This situation is common for all types of magnetic substances. The experimental results in the cases of ferromagnets and antiferromagnets are presented, for instance, in [1–5]. As there is no doubt that the region for $\varepsilon \geqslant 10^{-1}$ can really be "not critical", the situation for $\varepsilon \leqslant 10^{-3}$ is not quite clear. McCoy and Wu [6] have pointed out that

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very near to critical temperature the deviation of experimental data from the power-law (1), so called rouding, can be caused by random physical impurities like vacancies, dislocations and chemical impurities as well.

The measurements perfoımed on nickel [1] show that in fact, in this case, the crystalline imperfection has much greater effect on the rounding than the chemical impurities. On the other hand, the specific heat measurements [7–9] peformed on DAG, $RbMnF_3$ and $MnBr_2 \cdot 4H_2O$ do not indicate any significant effect of impurities or defects on the rounding. The influence of microscopic imperfections in a single crystal is some times connected with a presumable subdivision of the sample into an array of microscopic regions [1]. These micro-regions could have slightly different critical temperatures continuously distributed around the average value T_c. Assuming this distribution to be Gaussian one can, in fact, reproduce the experimental data of the specific heat exactly throughout the transition region.

However, Sukiennicki, Wojtczak and Mrygoń [10, 11] showed that in the case of an inhomogeneous system the critical temperature has a global character. It means that T_c is the same for the whole system. All these facts suggest that rounding of the specific heat peak close to the critical temperature may be, in the case of magnetics, an intrinsic property of the phase transition. Mrygoń and Wentowska [13] showed that taking into account fluctuations of magnetic moment in the calculation of magnetic specific heat of a cubic ferromagnet leads to the appearance of that rounding. In the present work we show that one obtains the similar result in the case of antiferromagnet. The calculations have been carried out only for temperatures $T \geqslant T_N$.

2. *Fluctuations and specific heat*

We used the same model of the system near the phase transition as in [12–16]. In this model an antiferromagnetic system of N spins, with the fluctuations of magnetic moment, is replaced by the sum of N_f equal cells each of which has a volume equal to the mean volume of a fluctuation.

The each cell, very small from the macroscopical point of view, is sufficiently large to apply statistical mechanics to describe the properties of it. The cell remains in a non-equilibrium state with respect to the whole system but we assume a local equilibrium. To describe a fluctuation the constant coupling approximation [17] of cubic antiferromagnet with spins $s = 1/2$ was used. The spatial distribution of magnetic moment in a fluctuation was taken into account by introducing the fluctuation $c(r)$ of the effective field acting on a pair of spins. The change of free energy of a single fluctuation $\langle \Delta F_1 \rangle$ can be expressed as follows

$$\langle \Delta F_1 \rangle = \frac{1}{2v} \int\limits_{V_1} d^3 r \Delta F[c(r)],$$

$$\Delta F[c(r)] = \sum_{\alpha=1}^{z} \Delta \mathscr{F}_{r,r+\delta}, \tag{2}$$

where $\Delta F[c(\boldsymbol{r})]$ is the free energy of a cluster of $z+1$ spins with central spin at $\boldsymbol{r}$, $\Delta \mathscr{F}_{r,r+\delta}$ is free energy of a pair of spins, v is the volume per one atom in a crystal lattice and V_1 is the volume of a fluctuation. The quantity $\Delta F[c(\boldsymbol{r})]$ was in our case calculated using the form of $\Delta \mathscr{F}_{r,r+\delta}$ given in [15] and then according to procedure described in [13, 18]. The application of the variational method [16] to the function $\Delta F[c(\boldsymbol{r})]$ leads to the differential equation for the distribution of the magnetic moment [15, 18]. An approximate solution of this equation, in the case when it was deiived with the accuracy to the third order terms, has the form [13]

$$M = \mu A_0 \exp(-Kr), \tag{3}$$

where A_0 is the mean value of the magnetic moment at the origin and can take the values from 0 to 1, μ is the Bohr magneton,

$$K^2 = \frac{K_1^2 + \frac{1}{2} K_3 (\mu A_0)^2}{3[1 - \frac{1}{2} K_3 \nu (\mu A_0)^2]}, \tag{4}$$

where a is the lattice constant and K_1, K_3, $K_3\nu$ are given in [15]. The parameter K^{-1} has the meaning of the correlation range which, as it follows from (4), remains finite in the critical point. In writing (3) the following relation [15] between magnetic moment and the fluctuation of effective field was used

$$M(\boldsymbol{r}) = 2\mu^2 \beta \frac{x^2 - 1}{\beta J (x^2 + 3)} c(\boldsymbol{r}), \tag{5}$$

where $x = \exp(\beta J)$, $\beta = 1/kT$, J is exchange constant. The mean number of fluctuations N_f can be expressed by the mean number of spins N_1 in a fluctuation. The number N_1 can be determined from the condition for the maximum of the probability of occurrence of a fluctuation [19].

Thus the total change of free energy ΔF of the system due to fluctuations is equal

$$\Delta F = N_f \langle \Delta F_1 \rangle. \tag{6}$$

The quantity $\langle \Delta F_1 \rangle$ was calculated according to (2) and has the form

$$\langle \Delta F_1 \rangle = -\frac{\pi}{2v} (\mu A_0)^2 P K^{-1} \{P(\mu A_0)^2 [a^2 (C - 6D) + 2zDK^{-2}] + 2a^2 (B - A) + 4zAK^{-2}\}, \tag{7}$$

where A, B, C, D and P are the functions of temperature and are given in [15].

By analogy with the case of ferromagnet [13]

$$N_f \simeq N(kT)^{-1} (\mu A_0)^2 [W_2(T) + W_4(T) (\mu A_0)^2], \tag{8}$$

where

$$W_2(T) = 4zAP, \quad W_4(T) = 16zDP^2. \tag{9}$$

Applying the general relation

$$\Delta C_{\mathrm{M}} = -T \frac{\partial^2}{\partial T^2} (\langle \Delta F_1 \rangle N_{\mathrm{f}}) \tag{10}$$

we obtain the magnetic specific heat related to the fluctuations of magnetic moment in an antiferromagnet. The results of numerical calculations are presented in Fig. 1 for two values of the amplitude A_0.

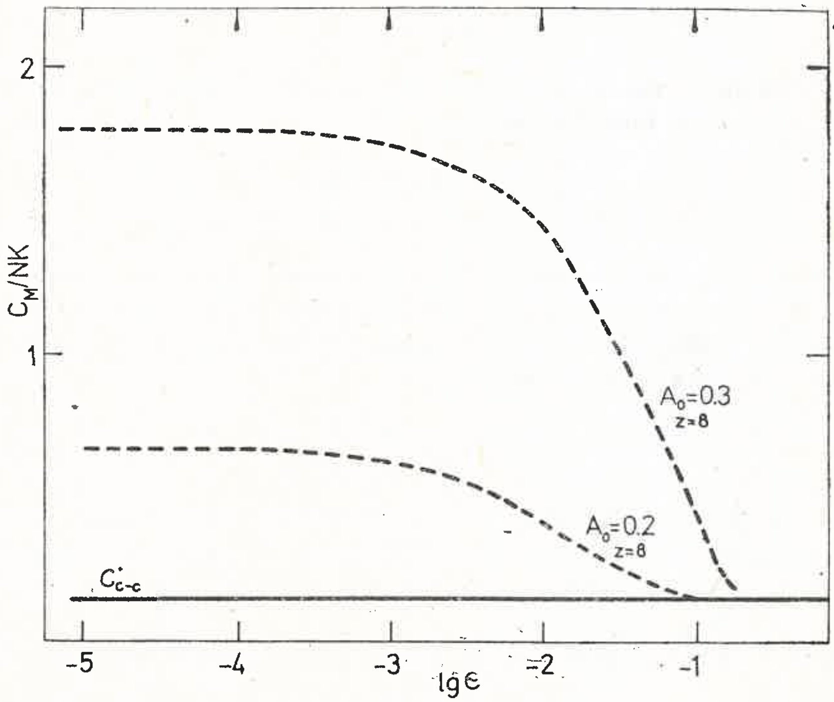

Fig. 1. Magnetic specific heat C_M/Nk as a function of lg ε, for two values of the amplitude A_0. The dashed curves represent the increment of the specific heat ΔC_M due to fluctuations. The curve C_{c-c} represents the approximate specific heat for homogeneous system obtained by means of constant coupling approximation

In our calculations several simplifying mathematical and physical assumptions were made which are as follows:

1. The division of a system into N_f noninteracting cells equal to the most probable volume of a fluctuation.

2. The amplitude A_0 was treated as a parameter of the theory independent on temperature.

3. In calculating the free energy of a fluctuation the summation over the spins was replaced by the integral over the volume of the fluctuation (Eq. (2)). This integral has been calculated in the limits (0, ∞) what is justified by the fast convergence of the function (3).

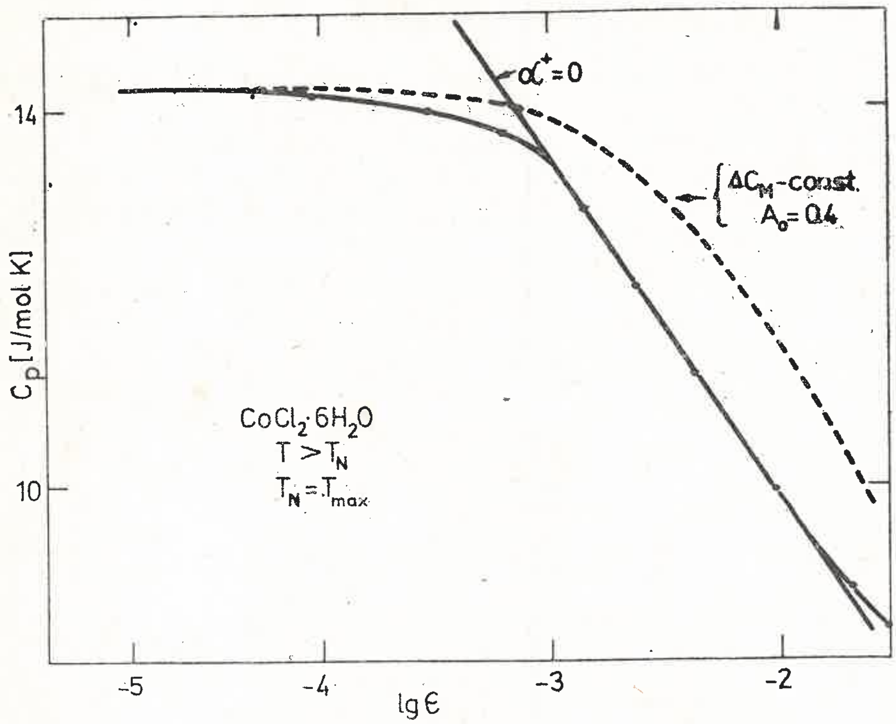

Fig. 2. The specific heat of $CoCl_2 \cdot 6H_2O$ [5] (dotted line) together with a power-law fit (1) for $\alpha^+ = 0$ (straight line) and with present work results (10) (dashed line). Description ΔC_M—const denotes that some constant was subtracted from the absolute values of ΔC_M in order to obtain the best qualitative agreement with the exprimental data. The theoretical and experimental values are equated at the point $\lg \varepsilon = -5$. T_{max} is the temperature of the maximum of the specific heat peak

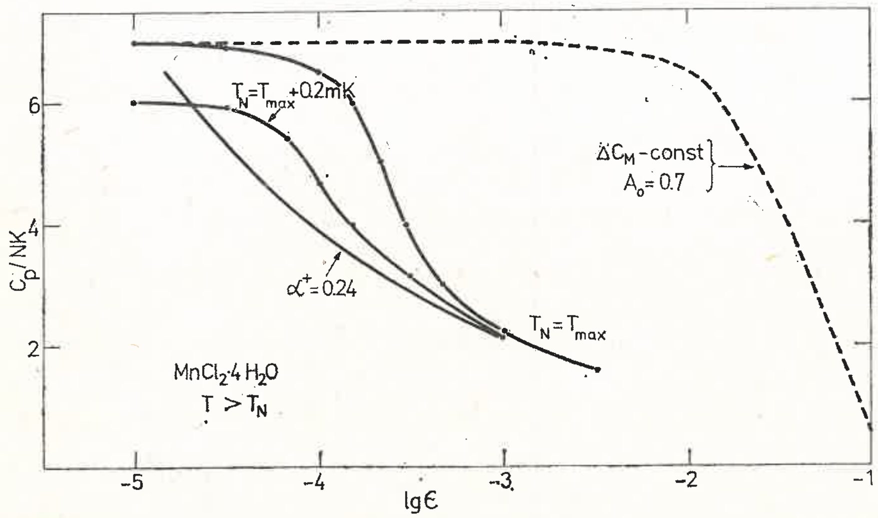

Fig. 3. The specific heat of $MnCl_2 \cdot 4H_2O$ [4] (dotted lines), for two values of T_N, together with a power-law fit (1) for $\alpha^+ = 0.24$ and with present work results (10). The meaning of ΔC_M—const and T_{max} is the same as in Fig. 1

3. *Conclusions*

Results obtained for an antiferromagnet show the same kind of the influence of fluctuations on the specific heat as in the case of a ferromagnet [13].

In both cases the specific heat becomes practically constant (rounded) in the immediate vicinity of critical temperature. A comparison of the experimental data for $CoCl_2 \cdot 6H_2O$ [5] and $MnCl_2 \cdot 4H_2O$ [4] with our theoretical results (10) and with a power-law fit (1) is given in Fig. 2 and 3.

It appears that the results obtained for the fluctuation model of an antiferromagnet are in much better qualitative agreement with experimental data in the whole range of ε than those based on critical indices method.

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