INFLUENCE OF SAMPLE INHOMOGENEITY ON THE BEHAVIOR OF THE SUSCEPTIBILITY OF A SYSTEM NEAR THE CRITICAL POINT

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It is shown by the self-consistent field theory method that inhomogeneity of the sample appreciably affects the behavior of the magnetic susceptibility near the critical point. At the point T_{c1} at which unbounded regions with a stable ordering parameter η arise, the susceptibility possesses a weak "unobservable" nonanalyticity. At this point the value of η averaged over the sample is zero. At the point T_{c2}, in which a nonvanishing order parameter throughout the sample arises, the susceptibility is infinite but the peak width is very small.

1. EXPERIMENTAL investigations[1-4] have shown that thermodynamic quantities near the critical point are strongly influenced by impurities and inhomogeneities. The specific-heat curve broadens, becomes finite, and its maximum shifts towards lower temperatures. The susceptibility also becomes finite,[3,4] but its maximum shifts less than the maximum of the specific heat.

Theoretical investigations of the influence of impurities and inhomogeneities on the behavior of thermodynamic quantities near the critical point are carried out in two limiting cases:

1) The sample temperature changes so rapidly, that the impurities do not have time to become redistributed when the temperature changes (the model of "quenched impurities"—MQI).

2) The temperature of the sample changes infinitely slowly, so that the impurities have time to become redistributed when the temperature changes (model of thermodynamic-equilibrium impurities (MTEI)).

In [6], within the framework of the two dimensional Ising model, there was considered the influence of nonmagnetic impurities on the behavior of the specific heat (in the MQI case). It was shown that at a temperature equal to the critical value for the pure substance there arises an essentially singular point, in which all the derivatives of the free energy with respect to the temperature are finite. McCoy and Wu,[7] considering a two dimensional Ising model in which the exchange energy changes randomly from column to column, also arrived at singularities of the same type as in [6].

It was shown in [8] that there exist two non-analytic points. The higher point is connected with the occurrence of regions of unlimited size with a stable ordering parameter η. At this point, the value of η averaged over the sample is equal to zero. In [6,7], a singularity of the specific heat was obtained near this point, The point of lower temperature is connected with the occurrence of a nonzero ordering parameter over the sample. Griffiths[9] proved a theorem of existence of two singular points: T_{c1}, below which the spontaneous magnetization does not exist and the magnetization is a non-analytic function of H and H = 0, and T_{c2}, below which spontaneous magnetization appears. These results agree with [8].

Unlike the "quenched impurities" model, in the MTEI[10] there is one non-analytic point (a point at which a magnetic moment is produced). At this point, the specific heat is finite. Thus, both the MQI and the MTEI lead to a weakening of the singularities of the specific heat.

At the same time, the singularity of the magnetic susceptibility χ becomes stronger in the MTEI (in the case of the three-dimensional Syozi model) compared with χ_0 of an ideal crystal, $\chi = \tau \exp -\gamma(1-\alpha)$, where τ is the dimensionless temperature, and γ and α are the exponents of the susceptibility and specific heat in an ideal crystal.[11] However, the experimental data on the measurement of the magnetic susceptibility in solids[3,4] indicate a weakening of the singularity of χ at the critical point (χ does not become infinite). The behavior of χ in the MQI is therefore of interest. Unlike the MTEI, in this model the singularity of χ becomes weaker. The magnetic susceptibility is finite everywhere, with the exception of a very narrow region in which it tends to infinity.

2. In the theory of the self-consistent field, the free energy of an inhomogeneous sample near the transition point can be expanded in the ordering parameter η:[12]

$$\Phi(\eta)=\int [\alpha(\mathbf{r},T)(T-T_c(\mathbf{r}))\eta^2+\beta(\mathbf{r},T)\eta^4+\gamma(\mathbf{r},T)(\nabla\eta)^2-H\eta]\,dV. \tag{1}$$

The inhomogeneities are described by the dependence of the expansion coefficients and of the critical temperature on the coordinates. We shall assume that the increments to the coefficients α, β, and γ, which are connected with the inhomogeneity, are small compared with their mean value, and then the influence of the inhomogeneity is taken into account only by the dependence of $T_c(\mathbf{r}) = T_0 + T_1(\mathbf{r})$ on the coordinates, where T_0 is the mean value of the function $T_c(\mathbf{r})$ [$T_1(\mathbf{r})$ is not assumed to be small compared with $T - T_0$]. The region of applicability of this theory is given by the inequality[8]

$$1 \gg T_1/T_0 \gg 1/r_0^6, \quad a \gg r_0.$$

From the condition of the minimum of the functional (1) we obtain the equation for the ordering parameter

$$\gamma\Delta\eta = \alpha(T-T_c(\mathbf{r}))\eta+2\beta\eta^3-H/2, \quad \gamma\geqslant 0. \tag{2}$$

The following theorem was proved in [8]. If $T_c(\mathbf{r})$ is an analytic function, as will henceforth be assumed, and η is a real continuous function with a continuous first derivative, then η either identically vanishes in the entire sample, or does not vanish anywhere.

The proof given in [8] can be readily generalized in the case of the presence of a magnetic field, but in this case, first $\eta \equiv 0$ cannot lead to an absolute minimum of the functional (1) at any temperature, and second, whereas both signs of η are possible at $H = 0$, we have $\eta > 0$ when $H \neq 0$. Indeed, if $H \neq 0$, we can choose $\eta > 0$ in all of space to be so small (but finite) that the last term becomes the principal term in (1). Then $\Phi[\tilde{\eta}] < 0$, whereas $\Phi[\eta \equiv 0] = 0$. We shall now show that $\eta \neq 0$ at the points of minimum x_0. Let us assume the opposite, that $\eta(x_0) = 0$ at the point of minimum x_0 and that $\eta'(x_0) = 0$; it then follows from (2) that $\gamma\eta'' = -H/2$ and $\eta'' < 0$, i.e., x_0 is a maximum point, thus contradicting the initial assumption. Thus, if $H \neq 0$ and the conditions given above are satisfied, then $\eta > 0$ at all points of the sample. This theorem was proved without taking into account the fluctuations of the ordering parameter. Fluctuations lead to violation of the theorem.

3. We seek the solution of (2) assuming a weak magnetic field, when $\alpha\tau\eta \gg H$. In this case the magnetic correlation radius $r_c(H) \sim \gamma^{1/2}/H^{1/3}\beta^{1/6}$ is much larger than the temperature correlation radius $r_c(T) \sim \gamma^{1/2}(\alpha\tau)^{1/2}$. We introduce a parameter characterizing the "weakness" of the magnetic field,

$$\sigma = r_c^3(T)\,/\,r_c^3(H) = H\beta^{1/2}\,/\,(\alpha\tau)^{3/2} \ll 1.$$

We shall first consider the case $a \gg r_c(T)$, where a is the characteristic dimension of the inhomogeneity.

In the region $\tau = T_c(x) - T > 0$ we have the estimate

$$\gamma\eta''/\alpha\tau\eta \sim \gamma/a^2\alpha\tau \sim r_c^2(T)/a^2 = \delta \ll 1$$

and in the zeroth approximation

$$\eta_0 = (\alpha\tau/2\beta)^{1/2}. \tag{3}$$

The addition of η_0 in the first approximation in σ and δ will be equal to

$$\eta_1 = \frac{\gamma}{4\sqrt{2\alpha\beta}}\frac{1}{\tau^{3/2}}\left[\tau\frac{d^2T_1}{dx^2} - \frac{1}{2}\left(\frac{dT_1}{dx}\right)^2\right] + \frac{H}{4\alpha\tau}. \tag{4}$$

Near the point x_0 at which $\tau(x_0) = 0$, formulas (3) and (4) no longer hold. The spatial boundaries of the applicability of formulas (3) and (4) are the same as in [8]: $\eta_1/\eta_0 \sim \delta T_1^3/\tau^3 + \sigma \ll 1$, and using the estimate of τ near the point x_0, $\tau \sim T_1(x - x_0)/a$, we obtain $((x - x_0)/a)^3 \gg \delta$.

In the spatial region with $\tau < 0$, the value of the ordering parameter is small and the term $\beta\eta^3$ in Eq. (2) can be neglected. In this approximation, the solution of Eq. (2) consists of the sum of the solutions of the homogeneous equation obtained in [8], and a particular solution of the inhomogeneous equation. For the latter, with accuracy of the order of $H\delta$, we can take $\eta(x) = H/2\alpha|\tau|$. The complete solution of the equation is

$$\eta(x) = \frac{H}{2\alpha|\tau|} + \frac{D}{|\tau|^{1/4}}\exp\left(\pm\int_{x_0}^{x}\sqrt{\frac{\alpha}{\gamma}|\tau|}\,dx\right). \tag{5}$$

The constant D can be obtained by "joining together" (5) with formulas (3) and (4). Just as in [8], we obtain $D \sim \eta T_1^{1/4}\delta^{1/3}$, where η is the order of magnitude of the ordering parameter in the region $\tau > 0$.

4. Unlike the calculation of the specific heat of an inhomogeneous sample, in the calculation of the susceptibility it is necessary to take into account the contributions of the regions with $\tau > 0$ as well as of the regions with $\tau < 0$:

$$\bar{\eta}(H) = \lim_{L\to\infty}\left[\frac{H}{2\alpha}\int_{\tau<0}\frac{1}{|\tau|}dx + \frac{H}{4\alpha}\int_{\tau>0}\frac{1}{\tau}dx\right],$$
$$\chi = \lim_{H\to 0}\frac{\partial\bar{\eta}(H)}{\partial H}. \tag{6}$$

We introduce the function $\rho(T_1)$, which describes the probability distribution of the temperature T_1 in the sample. For simplicity we neglect the variance of the inhomogeneity parameter (unless specially stipulated). Then

$$\chi = \frac{1}{2\alpha}\int_{-\infty}^{T-T_0}\frac{\rho(T_1)}{T - T_0 - T_1}dT_1 + \frac{1}{4\alpha}\int_{T-T_0}^{\infty}\frac{\rho(T_1)}{T_0 - T + T_1}dT_1. \tag{7}$$

Taking into account the limits of applicability of formulas (3)–(5), we integrate with respect to T_1 from $-\infty$ to $T - T_0 - T_1\delta^{1/3}$ and from $T - T_0 + T_1\delta^{1/3}$ to ∞. The regions $\sim T_1\delta^{1/3}$ near $\tau = 0$ make a small contribution to the total susceptibility of the sample.

We shall carry out the calculations for a Gaussian distribution and for a distribution in which T_1 has an upper limit. As shown in [8], without allowance for the fluctuations, the singular point exists only for the second type of distribution.

a) Asymptotic estimates for a Gaussian distribution

$$\rho(T_1) = \frac{1}{\sqrt{2\pi}t}e^{-T_1^2/2t^2}$$

yield

$$\text{for } T - T_0 \gg t \quad \chi = \frac{1}{2\alpha(T - T_0)}\left[1 + \frac{t^2}{(T - T_0)^2}\right], \tag{8a}$$

$$\text{for } T_0 - T \gg t \quad \chi = \frac{1}{4\alpha(T_0 - T)}\left[1 + \frac{t^2}{(T - T_0)^2}\right]. \tag{8b}$$

The contributions of the ferromagnetic regions to formula (8a) and of the paramagnetic regions to (8b) are exponentially small and can be disregarded. If we choose for the distribution $\rho(T_1)$ a function which is asymmetrical with respect to the reversal of the sign of $(T - T_0)$ (but with $\int_{-\infty}^{0}\rho(T_1)dT_1 = \int_{0}^{\infty}\rho(T_1)dT_1$), having no upper limit, then the corrections to the Curie–Weiss law in (8a) and (8b) will be linear in $t/|T - T_0|$ and will enter in (8a) and (8b) with different signs.

b) We now consider the case when T_1 is bounded from above by the temperature $T_{1\,max}$. An essential difference from a Gaussian distribution will be observed only near $T_{1\,max}$, and the form of the distribution itself has little influence on the character of the behavior of the susceptibility. We consider a very simple distribution of the type

$$\rho(T_1) = \begin{cases} 1/2T_{1max}, & -T_{1max} < T_1 < T_{1max} \\ 0, & \text{elsewhere} \end{cases}$$

The susceptibility in the region $T > T_0 + T_{1\,max}$ is

$$\chi = \frac{1}{4\alpha T_{1max}}\ln\frac{T - T_0 + T_{1max}}{T - T_0 - T_{1max}}. \tag{9}$$

The condition for the applicability of this formula is

$-\delta \ll 1$, i.e., $T - T_0 - T_{1\,max} \gg \gamma/\alpha a^2 \sim T_{1\,max}\delta$. In the region $T - T_0 \gg T_{1\,max}$, formula (9) goes over into the usual Curie–Weiss law $\chi = 1/2\alpha(T - T_0)$. In the region $T_{1\,max}\delta \ll T - T_0 - T_{1\,max} \ll T_{1\,max}$, expression (9) takes the form

$$\chi = \frac{1}{4\alpha T_{1max}} \ln \frac{2T_{1max}}{T - T_0 - T_{1max}}.$$

An essential deviation from the Curie–Weiss law takes place in the temperature region $T - T_0 \lesssim t(T_{1\,max})$ where the contributions of the paramagnetic and ferromagnetic regions are comparable. In the case of a distribution that has an upper temperature limit at $T - T_0 - T_{1\,max} \ll \delta T_{1\,max}$, it is necessary to take into account the contributions of regions of dimension l much smaller than the average dimension a. By virtue of the low probability of such regions at a temperature $T_0 + T_{1\,max}$,[8] the susceptibility has essentially a singular point of the same type as the specific heat. The contribution of the ferromagnetic regions at $|T - T_0| \ll t$ can be estimated from the following formula (in the case of a Gaussian distribution)

$$\chi_{\text{п}} \sim \frac{1}{2\alpha\sqrt{2\pi t}} \ln \frac{b}{\delta^{1/3}}, \quad b \sim 1. \tag{10}$$

Let us estimate the contribution made to the susceptibility by the ferromagnetic regions. At temperatures below $T_{1\,max}$ there appear regions ($\tau > 0$) with nonzero ordering parameters. The probability that in one such spatial region there will appear ordering with negative sign is $\omega \sim \exp(-E_{min}/T)$, where E_{min} is the minimum work necessary to produce such a fluctuation.[12] It is easy to see that the work will be minimal if the change of the sign and of the absolute magnitude of η will occur in a spatial region $T > T_c(x)$ of thickness r_c (where the absolute value of η is minimal).

The main contribution to E_{min} is made by two terms: 1) the term proportional to $(\nabla\eta)^2$, which contributes in regions in which a change of the sign and magnitude of η takes place; 2) the work of "reversal of magnetization" of the region $\tau > 0$ makes a contribution proportional to the volume.

We have

$$E_{min} = R_{min} + E_{min}(H) \sim \gamma \int (\nabla\bar{\eta})^2\, dV + L_1^3 H\bar{\eta} \sim L_1^2 e^{-ga_1/r_c} + L_1^3 H\bar{\eta}, \tag{11}$$

where L_1 is the linear dimension of the region $\tau > 0$, $\bar{\eta}$ is the mean value of the ordering parameter in the ferromagnetic region, and a_1 is the mean distance between regions. All these quantities are functions of the temperature. When the temperature changes from $T_0 + T_{1\,max}$ to $T_0 - T_{1\,max}$ the value of L_1 changes from $L_1 \sim a$ to $L_1 \sim \infty$. The increase of L_1 with decreasing temperature consists of the increase of individual ferromagnetic regions and of the coalescence of several ferromagnetic regions into one. The distance a_1 decreases with decreasing temperature, and the interaction between the regions increases. It is seen from (11) that $E_{min} \ll T_0$ in the temperature region $T_0 + T_{1\,max}$ and $E_{min} \gg T_0$ in the temperature region near $T_0 - T_{1\,max}$. This makes the probability of the fluctuations under consideration of the order of unity near $T_0 + T_{1\,max}$, and the probability of such fluctuations is small in the region near $T_0 - T_{1\,max}$.

Such fluctuations can be qualitatively described as a system of interacting regions with a Hamiltonian in the form

$$\hat{H} = R_{min} \sum \sigma_i\sigma_j + L_1^3 H\bar{\eta} \sum \sigma_i, \tag{12}$$

where $\sigma_i \pm 1$ (depending on the sign of η in the region $\tau > 0$). In such a system there occurs a second-order phase transition at $H = 0$; this transition is connected with the occurrence of a nonzero (when averaged over the sample) ordering parameter $\langle\sigma\rangle$ at the transition point defined by the equation $gR_{min}(\bar{T}_c)/T_0 = 1$.[8] Calculating $\langle\sigma\rangle$ from (12) by the self-consistent field method, we obtain the magnetic moment per region:

$$M(H) = \langle\sigma(H)\rangle\,\bar{\eta} \sim \frac{L_1^3 H\bar{\eta}^2}{gR'_{min}(T - \bar{T}_c)} \sim \frac{L_1^3 H\bar{\eta}^2}{T_0(T - \bar{T}_c)} T_1. \tag{13}$$

In deriving the last formula we used the estimate $R'_{min} = \partial R_{min}/\partial T \sim T_0/T_1$. The susceptibility per particle is

$$\chi_f \sim \frac{T_1^2}{T_0^2} \frac{1}{\alpha(T - \bar{T}_c)}, \quad T - \bar{T}_c \ll T_1. \tag{14}$$

Thus, the contribution of the ferromagnetic regions to the susceptibility is determined by the Curie–Weiss law with a small coefficient $\sim T_1^2/T_0^2$. The temperature region in which the contribution of the ferromagnetic regions plays the principal role can be estimated from $\chi_f \gg \chi_p$ (formulas (14) and (10)):

$$\frac{T_1^2}{T_0^2} \frac{1}{\alpha(T - \bar{T}_c)} \gg \frac{1}{\alpha T_1},$$

hence $|T - T_c| \ll T_1^3/T_0^2$. The width of this peak is much smaller than the width of the "smearing" of the temperature distribution.

In the derivation of (14) we have assumed that the system is inhomogeneous over scales larger than a. Actually the system of ferromagnetic regions, described by the Hamiltonian (12), is also inhomogeneous, leading to an additional weakening of the singularity (14), and the Curie–Weiss law

$$\chi \sim \frac{T_1^2}{T_0^2} \frac{1}{\alpha(T - \bar{T}_c)}$$

is the upper bound of this singularity.

The total susceptibility χ in the temperature region $|T - T_0| \lesssim t$ consists of the paramagnetic and ferromagnetic parts, $\chi = \chi_f + \chi_p$, where χ_p is given by (10) and χ_f by (14).

5. Let us consider another limiting case of the solution of Eq. (2), when $r_c(T) \gg a$ ($\delta \gg 1$), the magnetic field being assumed weak as before ($r_c(H) \gg r_c(T)$). We represent $\eta(x)$ in the form $\eta(x) = \eta_0 + \eta_1(x)$, where

$$\eta_0 = \lim_{L\to\infty} \int_{-L}^{+L} \eta(x)\, dx.$$

Just as in [8], it is natural to assume that $\eta_1 \ll \eta_0$. The mean value of the susceptibility of the sample is in this case

$$\chi = \lim_{H\to 0} \frac{\partial \eta_0}{\partial H}.$$

Averaging Eq. (2) written in the form

$$\begin{aligned}\gamma\eta_1'' = \alpha(T - T_0 - T_1(x))\eta_0 + \alpha(T - T_0)\eta_1(x) - \alpha T_1(x)\eta_1(x) \\ + 2\beta(\eta_0^3 + 3\eta_0^2\eta_1(x) + 3\eta_0\eta_1^2(x) + \eta_1^3(x)) - H/2\end{aligned} \tag{15}$$

over the entire sample, we obtain

$$\alpha(T-T_0)\eta_0-\alpha\overline{T_1\eta_1}+2\beta(3\eta_0\overline{\eta_1^2}+\eta_0^3)-H/2=0. \quad (15')$$

Subtracting (15′) from (15) and again using (15′), we arrive at the system of equations

$$\alpha(T-T_0)\eta_0-\alpha\overline{T_1\eta_1}+2\beta\eta_0^3-H/2=0, \quad (16a)$$

$$\gamma\eta_1''=-\alpha T_1\eta_0+\alpha(T-T_0)\eta_1+6\beta\eta_0^2\eta_1. \quad (16b)$$

From the solution of (16b) we obtain the mean value of $\overline{T_1\eta_1}$:[8]

$$\overline{T_1\eta_1}=\frac{\alpha\eta_0}{\gamma\lambda}S, \quad (17)$$

where $\lambda=([\alpha(T-T_0)+6\beta\eta_0^2]/\gamma)^{1/2}$, and S is a constant characterizing the mean value of the paired correlations of the distribution $T_1(x)$.

Substituting (17) in (16a), we obtain an equation for η_0:

$$[\alpha(T-T_0)+6\beta\eta_0^2]\left[\alpha(T-T_0)\eta_0+2\beta\eta_0^3-\frac{H}{2}\right]^2=\frac{\alpha^4\eta_0^2}{\gamma}S^2. \quad (18)$$

The critical point of such a system is given by [8]

$$\tilde{T}_c=T_0+\tilde{T}_1, \quad \tilde{T}_1=(\alpha S^2/\gamma)^{1/3}\sim T_1\delta^{-1/3}\ll T_1. \quad (19)$$

Let us consider the region $T-T_0\gg\tilde{T}_1$. In view of the smallness of η (in the paramagnetic region), (18) takes the form

$$\alpha(T-T_0)[\alpha(T-T_0)\eta_0-H/2]^2=\alpha^4\eta_0^2S^2/\gamma$$

and the susceptibility is

$$\chi=\frac{1}{2\alpha(T-T_0)}\left[1+\frac{\tilde{T}_1^{3/2}}{(T-T_0)^{3/2}}\right]. \quad (20)$$

At a temperature $T_0-T\gg\tilde{T}_1$, in the zeroth approximation, the solution of Eq. (18) is $\eta_0=(\alpha(T_0-T)/2\beta)^{1/2}$. The first approximation yields

$$\eta_{01}=\frac{H}{4\alpha(T_0-T)}+\frac{\alpha^{1/2}\tilde{T}_1^{3/2}}{4\sqrt{\beta}(T_0-T)},$$

i.e., the susceptibility corresponds to the susceptibility of the pure substance.

Let us consider the temperature region close to the transition temperature $|T-\tilde{T}_c|\ll\tilde{T}_1$. In the paramagnetic region we have

$$\chi_1=1/3\alpha(T-\tilde{T}_c), \quad (21a)$$

and in the ferromagnetic region

$$\chi_2=1/6\alpha(\tilde{T}_c-T). \quad (21b)$$

Thus, in this region there occurs a renormalization of the coefficients compared with the homogeneous case, but the ratio χ_1/χ_2 is the same as in the homogeneous case.

The final results of the inhomogeneity of the sample is that there are temperature regions with different dependences of the susceptibility on the temperature. In the region $T-T_0\gg T_1$ there is the usual Curie–Weiss law. In the region $T_1T_1^2/T_0^2\ll T-T_c\ll T_1$ we have $\chi\sim 1/\alpha T_1$, and finally, in the region $T-\tilde{T}_c\ll T_1T_1^2/T_0^2$ we have $\chi\sim T_1^2/T_0^2\,\alpha(T-\tilde{T}_c)$. In addition, in the temperature region $T_0+T_{1\,max}$ there is an essentially singular point of the same type as in [8]. From experiment[3] it is possible to estimate the value of the "smearing" of the temperature $T_1\sim 1°$, and consequently the width of the susceptibility peak at the point T_c is $\Delta T\sim 10^{-4}$ deg. Experiment has revealed a kink in the susceptibility at this point.

The experimentally observed weakening of the susceptibility of the critical point itself indicates that the model with the secured impurities, considered in the present paper and in [8], describes better the behavior of real samples, then the model with movable impurities,[10, 11] in which an intensification of the singularity of the susceptibility is observed at the critical point.

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