

Modified Landau Theory of the Second Order Phase Transition^{*)}

Yoshiki KURAMOTO

Department of Physics, Kyoto University, Kyoto

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The difficulty near the critical point encountered by the Landau theory or the classical theory of the second order phase transition is removed with the least modification of the original framework. The essential point of our idea is to note that the temperature region in which the Landau theory valids depends sensitively on the way of defining the local order parameter. It is shown that the relations among the singularities of various thermodynamic quantities predicted by this modified form of the Landau theory are in good agreement with those obtained by nonclassical theories such as the static scaling theory.

§ 1. Introduction

It is often pointed out that the Landau theory of the second order phase transition suffers from an internal inconsistency.¹⁾ In fact under the assumption of small fluctuation this theory predicts the large fluctuation at the critical point. The failure of this theory in the vicinity of the critical point, especially in the system with short range interaction, seems to be the consequence of this inconsistency. When we work with free energy density, there seems to be at least two possible ways to avoid this difficulty. One way is to take into account the higher order terms with respect to the local fluctuations and their spatial derivatives.^{**)} The other way, which is of our main concern in this paper, is to change the definition of the local order parameter with temperature without taking into account the higher order terms. In the next section the definition of the local order parameter is carefully examined, and this is indispensable for our whole discussion. The procedure of removing the inconsistency is then presented. In § 3 the asymptotic behaviors of various thermodynamic quantities are studied and all the independent relations among the critical indices are derived which turn out to be identical with those obtained by the static scaling theory.^{3),4)} Throughout the present paper the language is used appropriate for the Ising ferromagnets though the same kind of arguments may apply to the classical gas or liquid system as well.

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^{**)} This fact was pointed out by several authors but such an approach was found too difficult to obtain satisfactory results (see reference 2)).

§ 2. Elimination of the inconsistency

(a) Local order parameter

Let us start with an unambiguous definition of the local order parameter. In the present case the local order parameter, which we denote by $M(\mathbf{r})$, is the z -component of magnetization density averaged over a semimicroscopical region around the position \mathbf{r} :

$$M(\mathbf{r}) = \frac{1}{v} \int m(\mathbf{r} + \mathbf{r}') d\mathbf{r}'. \tag{2.1}$$

Here

$$m(\mathbf{r}) = \sum_i m_i \delta(\mathbf{r} - \mathbf{r}_i)$$

and m_i is the z -component of the magnetic moment of the spin at \mathbf{r}_i . The integral in (2.1) extends over a spherical region centered at \mathbf{r} , whose volume is denoted by v . The definition (2.1) implies that $M(\mathbf{r}_1)$ and $M(\mathbf{r}_2)$ cannot be regarded as independent variables if $|\mathbf{r}_1 - \mathbf{r}_2| < R$, where R is the radius of the sphere. Therefore it will be more convenient to work with the spatial Fourier transform defined by

$$M_{\mathbf{k}} = \frac{1}{\sqrt{V}} \int M(\mathbf{r}) e^{i\mathbf{k}\mathbf{r}} d\mathbf{r}, \tag{2.2}$$

where V is the total volume of the system. Then the Fourier transform of (2.1) is

$$M_{\mathbf{k}} = \frac{1}{v} \int m_{\mathbf{k}} e^{-i\mathbf{k}\mathbf{r}} d\mathbf{r}, \tag{2.3}$$

where $m_{\mathbf{k}}$ is the Fourier transform of $m(\mathbf{r})$. From (2.3) we have

$$\begin{aligned} M_{\mathbf{k}} &\cong 0 && \text{for } |\mathbf{k}| \gtrsim R^{-1}, \\ &\cong m_{\mathbf{k}} && \text{for } |\mathbf{k}| \lesssim R^{-1}. \end{aligned} \tag{2.4}$$

Therefore if $|\mathbf{k}|$ is considerably smaller than R^{-1} the quantity $M_{\mathbf{k}}$ has the clear physical meaning unaffected by the value of R . Our idea is to change R with temperature as will be seen later.

(b) Landau theory and its inconsistency

Let us briefly recapitulate Landau's idea, which underlies most of the classical theories,⁵⁾ in the form convenient for our purposes. According to Landau, the free energy density $g(\mathbf{r})$ near the critical point is given by

$$g(\mathbf{r}) = g_0 + aM(\mathbf{r})^2 + bM(\mathbf{r})^4 + c[\nabla M(\mathbf{r})]^2. \tag{2.5}$$

Here g_0 is independent of the local order parameter. In the presence of an external field, an additional term $-BM(\mathbf{r})$ appears in the above expression,

where B is the external field in a suitable unit. The effect of the external field is, however, considered separately in later part of the present paper. The total free energy G in an fluctuating state is given by

$$G(\{M(\mathbf{r})\}) = \int g(\mathbf{r}) d\mathbf{r}, \quad (2.6)$$

where $\{M(\mathbf{r})\}$ represents a certain distribution of magnetization density. It is obvious that G is minimized if

$$\begin{aligned} M(\mathbf{r}) &= \bar{M}, \\ \bar{M} &= 0 \quad \text{for } a > 0, \\ &= \sqrt{-\frac{a}{2b}} \quad \text{for } a < 0. \end{aligned} \quad (2.7)$$

In terms of \bar{M} (2.5) can be rewritten as

$$g(\mathbf{r}) = g_0 + a(M(\mathbf{r}) - \bar{M})^2 + b(M(\mathbf{r}) - \bar{M})^4 + c[\mathcal{V}(M(\mathbf{r}) - \bar{M})]^2 \quad \text{for } a > 0, \quad (2.8a)$$

$$\begin{aligned} g(\mathbf{r}) &= g_0 - \frac{|a|\bar{M}^2}{2} - 2a(M(\mathbf{r}) - \bar{M})^2 + 2\sqrt{2|a|b}(M(\mathbf{r}) - \bar{M})^3 \\ &\quad + b(M(\mathbf{r}) - \bar{M})^4 + c[\mathcal{V}(M(\mathbf{r}) - \bar{M})]^2 \quad \text{for } a < 0, \end{aligned} \quad (2.8b)$$

though (2.8a) is a trivial transformation. The expectation value of a certain physical quantity A may be calculated with the aid of a probability function $P(\{M(\mathbf{r})\})$:

$$\langle A \rangle = \int AP(\{M(\mathbf{r})\})d\{M(\mathbf{r})\} / \int P(\{M(\mathbf{r})\})d\{M(\mathbf{r})\}. \quad (2.9)$$

Here

$$P(\{M(\mathbf{r})\}) = \exp\left\{-\frac{G(\{M(\mathbf{r})\})}{k_B T}\right\}. \quad (2.10)$$

In the actual calculation some additional assumptions are necessary. The essential point of the conventional theory is to neglect the higher order fluctuation in (2.8a) and (2.8b), which amounts to approximate those expressions by

$$g(\mathbf{r}) = g_0 + a(M(\mathbf{r}) - \bar{M})^2 + c[\mathcal{V}(M(\mathbf{r}) - \bar{M})]^2 \quad \text{for } a > 0, \quad (2.11a)$$

$$\begin{aligned} g(\mathbf{r}) &= g_0 - \frac{|a|\bar{M}^2}{2} - 2a(M(\mathbf{r}) - \bar{M})^2 + c[\mathcal{V}(M(\mathbf{r}) - \bar{M})]^2 \\ &\quad \text{for } a < 0. \end{aligned} \quad (2.11b)$$

Then the total free energy may be written in terms of the Fourier transform of $M(\mathbf{r})$ in the form

$$G = G_0 + a(M_0 - \sqrt{V} \bar{M})^2 + V \int (a + ck^2) M_{\mathbf{k}} M_{-\mathbf{k}} d\mathbf{k}$$

for $a > 0$, (2.12a)

$$G = G_0 - 2a(M_0 - \sqrt{V} \bar{M})^2 + V \int (2|a| + ck^2) M_{\mathbf{k}} M_{\mathbf{k}} d\mathbf{k}$$

for $a < 0$, (2.12b)

where $G = Vg_0$ and $V(g_0 - |a|\bar{M}^2/2)$ for $a > 0$ and $a < 0$, respectively. We shall see below that such an approximation leads to an apparent contradiction near the critical point. Using (2.12a), (2.12b) and (2.8) various physical quantities may be calculated. For instance, the spontaneous magnetization becomes identical with \bar{M} and the density correlation function assumes Ornstein-Zernike form and so on. Let us now inquire the condition under which the above approximation holds. But since $M(\mathbf{r})$ is contained in $g(\mathbf{r})$ still as a variable and not as an averaged quantity such a condition will be meaningless without presupposing some averaging procedure taken over the possible values of $M(\mathbf{r})$. Speaking precisely, we are in fact inquiring the condition under which the nonlinear fluctuation terms may be neglected in $g(\mathbf{r})$ for such values of $M(\mathbf{r})$ as to allow the probability function take an appreciable value. Consider first the case $a > 0$. We observe that the probability function (2.10) may safely be replaced by a new function $\tilde{P}(\{M(\mathbf{r})\})$ defined by

$$\begin{aligned} \tilde{P}(\{M(\mathbf{r})\}) &= P(\{M(\mathbf{r})\}) & \text{if } |M(\mathbf{r})| < \mu \text{ for all } \mathbf{r}, \\ &= 0 & \text{otherwise.} \end{aligned} \quad (2.13)$$

The value of μ should be considerably larger than the magnitude of the mean fluctuation of $M(\mathbf{r})$, namely

$$\mu \gg \langle M(\mathbf{r})^2 \rangle^{1/2}, \quad (2.14)$$

which we write in the form

$$\mu = \lambda_0 \langle M(\mathbf{r})^2 \rangle^{1/2}, \quad (2.15)$$

where $\lambda_0 (\gg 1)$ is the minimum constant for which the replacement (2.13) remains valid. Therefore it may be concluded that the condition for the validity of the Landau's assumption is equivalent to the one that the term $bM(\mathbf{r})^4$ may be neglected as compared with $aM(\mathbf{r})^2$ when $M(\mathbf{r})$ is of the order of μ . This condition may be expressed as

$$a \gtrsim b\mu^2$$

or using (2.15)

$$a \gtrsim b\lambda_0^2 \langle M(\mathbf{r})^2 \rangle. \quad (2.16)$$

In the case $a < 0$ two kinds of nonlinear fluctuation terms, $2\sqrt{2|a|b}(M(\mathbf{r}) - \bar{M})^3$

and $b(M(\mathbf{r}) - \bar{M})^4$ appear. But the arguments go quite similarly as before, and the relevant condition becomes

$$-a \geq b\lambda_0^2 \langle (M(\mathbf{r}) - \bar{M})^2 \rangle^{1/2}. \quad (2.17)$$

(2.16) and (2.17) are now combined to a single condition:

$$|a| \geq b\lambda_0^2 \langle (M(\mathbf{r}) - \bar{M})^2 \rangle^{1/2}. \quad (2.18)$$

Obviously the inequality (2.18) cannot be satisfied in the Landau theory near T_c , because a vanishes and b as well as $\langle M(\mathbf{r})^2 \rangle$ remains finite at the critical point. Such a breakdown of the Landau's assumption causes serious difficulties. For instance, when $a < 0$, the free energy density $g(\mathbf{r})$ is no more an even function of $M(\mathbf{r}) - \bar{M}$, so that $\bar{M} = \sqrt{-a/2b}$ will not be the spontaneous magnetization. This in turn implies that the critical point should not be determined by setting $a=0$. Moreover it is quite probable that the expression (2.5) itself becomes insufficient.

(c) *Improvement of the Landau theory*

We have seen in the previous subsection that the validity of the Landau theory is not assured unless the condition (2.18) is satisfied. But whether this condition is satisfied or not at a given temperature depends on the way of defining the local order parameter or, more precisely, on the value of R introduced in (a). The reason for this may easily be seen if we note Eq. (2.4) and write the mean square fluctuation of the magnetization density in the form

$$\langle (M(\mathbf{r}) - \langle M \rangle)^2 \rangle \simeq \frac{1}{\pi^d} \int_0^{R^{-1}} \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle d\mathbf{k}, \quad (2.19)$$

where $\int_0^{R^{-1}} d\mathbf{k}$ means the integral over the d -dimensional sphere of radius R^{-1} in the momentum space, d being the dimension of the system under consideration. The above equation shows that when R is kept at a large value the fluctuation remains small and the condition (2.18) is more easily satisfied at a given temperature. On the contrary if R is kept small the region in which the Landau theory is valid will be reduced. It should be noted that the value of R may quite arbitrarily be chosen if we do not mind losing the informations about the local properties of the system. Therefore in what follows we shall change R with temperature in such a way that the condition (2.18) may be satisfied at all temperatures but the ratio $|a|/b\langle (M(\mathbf{r}) - \bar{M})^2 \rangle$ may not depend on the temperature. Thus we have

$$|a| \sim \lambda b \langle (M(\mathbf{r}) - \langle M \rangle)^2 \rangle, \quad (2.20)$$

where λ is a number larger than λ_0^2 but independent of temperature. Under such a procedure R tends of course to infinite as the critical point is approached. It may be asked why R should be changed and why should not be fixed at a large value beforehand. The answer is simple: R cannot be tend to infinite

beforehand, which leads to the trivial result $g(\mathbf{r}) = g_0 = \text{const}$, while if R is kept at large finite value the Landau theory will break down in the immediate vicinity of the critical point. Thus the asymptotic behavior of thermodynamic quantities may be clarified only by changing R with temperature.

As long as (2.18) is satisfied, no positive reason can be found why the Landau's free energy and his treatment should not be applied to our problem. Therefore we shall use hereafter the expressions (2.11a) and (2.11b) and replace \bar{M} by $\langle M \rangle$. Below the critical point the expression (2.5) may also be valid and be used if necessary. Since according to the Landau theory the contributions of the two kinds of the fluctuation terms appearing in the right-hand side of (2.11a) or (2.11b) are comparable in magnitude, we also assume this to be correct in the new description. This means that

$$|a| \langle (M(\mathbf{r}) - \langle M \rangle)^2 \rangle \sim c \langle (\nabla(M(\mathbf{r}) - \langle M \rangle))^2 \rangle \sim cR^{-2} \langle (M(\mathbf{r}) - \langle M \rangle)^2 \rangle. \quad (2.21)$$

The last expression in the above equation is derived as follows. First we have

$$\langle (\nabla(M(\mathbf{r}) - \langle M \rangle))^2 \rangle \simeq \frac{1}{\pi^d} \int_0^{R^{-1}} k^2 \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle d\mathbf{k} \sim \bar{k}^2 \langle (M(\mathbf{r}) - \langle M \rangle)^2 \rangle, \quad (2.22)$$

where

$$\bar{k}^2 = \int_0^{R^{-1}} k^2 \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle d\mathbf{k} \bigg/ \int_0^{R^{-1}} \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle d\mathbf{k}. \quad (2.23)$$

The quantity \bar{k}^2 is interpreted as the square of the typical wavenumber characterizing the magnetization fluctuation with the wavenumber smaller than R^{-1} . Therefore if we put

$$\sqrt{\bar{k}^2} = AR^{-1} \quad (A < 1)$$

A will not vanish at the critical point and may be considered constant.*) Thus Eq. (2.22) leads to the last expression of Eq. (2.21). From (2.21) we obtain

$$|a| \sim cR^{-2}. \quad (2.24)$$

So far the behavior of R near the critical point has not been referred to. It is shown in the following manner that R behaves like the correlation length. Using (2.9), (2.10) and (2.12a) the mean square fluctuation of the k -mode in the paramagnetic region is calculated as

*) The fact that A does not vanish at the critical point is assured except the case that the dominant part of the integral $\int_0^{R^{-1}} \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle d\mathbf{k}$ comes only from an infinitesimally small region in the k -space (much smaller than R^{-d}) near the critical point, which seems to be impossible except in the lower dimensional systems whose transition points are absolute zero.

$$\begin{aligned}\phi_{\mathbf{k}} &\equiv \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle = \frac{k_B T}{2(a + ck^2)} \\ &= \frac{k_B T}{2c} \frac{1}{(a/c) + k^2} \quad (k \ll R^{-1}).\end{aligned}\quad (2.25)$$

From (2.25) and (2.24) it will turn out that $\phi_{\mathbf{k}}$ has the Ornstein-Zernike form and that R plays the role of the correlation length. Quote an analogous argument holds for $a < 0$ also and, following the usual notation, we put

$$R^{-1} \sim \kappa. \quad (2.26)$$

From (2.24) and (2.26) we obtain

$$c \sim |a| \kappa^{-2}. \quad (2.27)$$

At first sight, our theory seems to have made no progress as compared with the classical theory as far as the correlation function is concerned. But this is not the case because of the following two reasons. The first is that in our framework the factor c^{-1} multiplying the Ornstein-Zernike type correlation function is expected to become singular at the critical point. This leads, as will be seen in the next section, to the correct relation among the critical indices different from that obtained by the Landau theory or the molecular field theory. The second reason is that in our treatment the expression (2.25) valid only in the region $k \ll R^{-1} (\sim \kappa)$, which implies that the region in which the Ornstein-Zernike form valid becomes indefinitely small as the critical point is approached. More detailed study of these points will be made in the next section.

Before ending this section let us summarize the asymptotic behavior of the quantities a , b and c appearing in the Landau's free energy. Throughout this paper Fisher's notations⁶⁾ are used for the critical indices which are listed in Table I, where the deviation from the critical temperature is indicated by ε :

$$\varepsilon \equiv (T - T_c) / T_c.$$

Let us first note that $|a|$ is proportional to the inverse susceptibility:

Table I.

Physical quantity	Sign of ε	Behavior of quantity	Critical index
Spontaneous magnetization $\langle M \rangle$	< 0	$\sim \varepsilon ^\beta$	β
	$= 0$	$\sim B^{1/\delta}$	δ
Correlation function $\phi(r)$	$= 0$	$\sim r^{-d+2-\eta}$	η
Inverse correlation length κ	> 0	$\sim \varepsilon^\nu$	ν
	< 0	$\sim \varepsilon ^{\nu'}$	ν'
Specific heat C	> 0	$\sim A\varepsilon^{-\alpha} + B$	α
	< 0	$\sim A' \varepsilon ^{-\alpha'} + B'$	α'
Uniform susceptibility χ_0	> 0	$\sim \varepsilon^{-\gamma}$	γ
	< 0	$\sim \varepsilon ^{-\gamma'}$	γ'

$$|a| \sim \chi_0^{-1}. \quad (2.28)$$

Also note that the right-hand side of (2.20) can be expressed as

$$\begin{aligned} \lambda b \langle (M(\mathbf{r}) - \langle M \rangle)^2 \rangle &\simeq \lambda b \frac{1}{\pi^d} \int_0^{R^{-1}} \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle d\mathbf{k} \\ &\sim \lambda b \frac{1}{\pi^d} \int_0^{R^{-1}} \langle m_0 m_0 \rangle d\mathbf{k} \sim \lambda b \kappa^d \chi_0, \end{aligned} \quad (2.29)$$

where we have used (2.25), (2.24) and the proportionality of $\langle m_0 m_0 \rangle$ to χ_0 . Then (2.20) can be written as

$$b \sim \lambda^{-1} |a| \kappa^{-d} \chi_0^{-1}. \quad (2.30)$$

On the other hand, by definition (see Table I),

$$\begin{aligned} \kappa &\sim \varepsilon^\nu \quad \text{for } T > T_c, \\ &\sim |\varepsilon|^{\nu'} \quad \text{for } T < T_c \end{aligned}$$

and

$$\begin{aligned} \chi_0 &\sim \varepsilon^{-r} \quad \text{for } T > T_c, \\ &\sim |\varepsilon|^{-r'} \quad \text{for } T < T_c. \end{aligned}$$

Thus (2.28), (2.30) and (2.27) reduce to

$$a \sim \varepsilon^r, \quad (2.31a)$$

$$b \sim \lambda^{-1} \varepsilon^{2r-d\nu}, \quad (2.31b)$$

$$c \sim \varepsilon^{r-2\nu} \quad (2.31c)$$

for $T > T_c$, and

$$|a| \sim |\varepsilon|^{r'}, \quad (2.32a)$$

$$b \sim \lambda^{-1} |\varepsilon|^{2r'-d\nu'}, \quad (2.32b)$$

$$c \sim |\varepsilon|^{r'-2\nu'} \quad (2.32c)$$

for $T < T_c$. Equations (2.31) and (2.32) together with (2.11a) and (2.11b) (and (2.5) if necessary) form a set of basic equations to our theory.

§ 3. Asymptotic behaviors of physical quantities

(a) Pair correlation function

According to our treatment, the spatial correlation function $\phi(r)$ of the magnetization density may have different forms when $r \gg \kappa^{-1}$ and when $r \ll \kappa^{-1}$.

First consider the large distance ($r \gg \kappa^{-1}$) correlation function. In the previous section we have seen that the mean square fluctuation of the k -mode for $k \ll \kappa$ has the Ornstein-Zernike form multiplied by c^{-1} (see Eq. (2.25)). Using Eqs. (2.31c) and (2.32c) we obtain

$$\begin{aligned} \phi_{\mathbf{k}} &\sim \frac{\varepsilon^{-\gamma+2\nu}}{\kappa^2 + k^2} \quad \text{for } T > T_c, \\ &\sim \frac{|\varepsilon|^{-\gamma'+2\nu'}}{\kappa^2 + k^2} \quad \text{for } T < T_c. \end{aligned} \quad (3.1)$$

In real space these equations can be expressed as

$$\begin{aligned} \phi(r) &\sim \frac{\varepsilon^{-\gamma+2\nu}}{(\kappa r)^{(d-1)/2}} e^{-\kappa r} \quad \text{for } T > T_c, \\ &\sim \frac{|\varepsilon|^{-\gamma'+2\nu'}}{(\kappa r)^{(d-1)/2}} e^{-\kappa r} \quad \text{for } T < T_c, \end{aligned} \quad (3.2)$$

which are valid only for $r \gg \kappa^{-1}$. For $r < \kappa^{-1}$ let us assume the form of $\phi(r)$ as

$$\phi(r) \sim \frac{e^{-\kappa r}}{r^{d-2+\eta}}, \quad (3.3)$$

where η is the critical index characterizing the deviation from the Ornstein-Zernike form. For $r < \kappa^{-1}$, $e^{-\kappa r}$ may approximately be replaced by unity. Therefore, integrating (3.3) over the d -dimensional region satisfying $r < \kappa^{-1}$ and dividing by its volume v we obtain

$$\begin{aligned} \frac{1}{v} \int_v \phi(r) dr &\sim \kappa^{d-2+\eta} \sim \varepsilon^{\nu(d-2+\eta)} \quad \text{for } T > T_c, \\ &\sim |\varepsilon|^{\nu'(d-2+\eta)} \quad \text{for } T < T_c, \end{aligned} \quad (3.4)$$

where we have assumed that $\eta \neq 2$. On the other hand, noting Eqs. (2.3) and (3.1), we obtain another equation

$$\begin{aligned} \frac{1}{v} \int_v \phi(r) dr &= \frac{1}{v} \int_v \langle m(\mathbf{r}_0 + \mathbf{r}) m(\mathbf{r}_0) \rangle d\mathbf{r} \\ &\simeq \frac{1}{\pi^d} \int_0^\kappa \langle m_{\mathbf{k}} m_{-\mathbf{k}} \rangle d\mathbf{k} \sim \varepsilon^{d\nu-\gamma} \quad \text{for } T > T_c, \\ &\sim |\varepsilon|^{d\nu'-\gamma'} \quad \text{for } T < T_c. \end{aligned} \quad (3.5)$$

Comparing Eqs. (3.4) and (3.5), we obtain the relations⁷⁾

$$\frac{\gamma}{2-\eta} = \nu \quad \text{and} \quad \frac{\gamma'}{2-\eta} = \nu'. \quad (3.6)$$

It will be instructive to test the validity of our theory by applying it to the

two-dimensional Ising spin system. Using $\gamma = \gamma' = 7/4$, $\nu = \nu' = 1$ and $\eta = 1/4$,⁸⁾ which are known to be exact in this case, Eq. (3.2) is reduced to

$$\phi(r) \sim \frac{|\varepsilon|^{1/4}}{(|\varepsilon|r)^{1/2}} e^{-|\varepsilon|r} \quad \text{for } r \gg \kappa^{-1} \quad (3.7)$$

both above and below the critical point. On the other hand it was shown by Kadanoff⁹⁾ that $\phi(r)$ coincides with (3.7) for $r \gg \kappa^{-1}$ and $T > T_c$. But below T_c , $\phi(r)$ was found to behave like

$$\phi(r) \sim \frac{|\varepsilon|^{1/4}}{(|\varepsilon|r)^2} e^{-2|\varepsilon|r} \quad \text{for } r \gg \kappa^{-1}, \quad (3.8)$$

which is different from (3.7). Therefore our theory seems to be insufficient for $T < T_c$ though the reason for it is not clear.

(b) *Specific heat*

The thermodynamic potential of the total system is given by

$$\Psi = -k_B T \ln \int \exp\left(-\frac{G}{k_B T}\right) d\{M(\mathbf{r})\}, \quad (3.9)$$

where G is given by (2.12a) and (2.12b) for $T > T_c$ and $T < T_c$, respectively. In those equations the effective range of k -integration is of the order of κ^a on account of (2.4) and (2.26). First consider the case of $T > T_c$. Inserting (2.11a) into (3.9) Ψ can be expressed as

$$\Psi = \Psi_0 - \frac{V}{\pi^a} \int \left[\ln \int \exp\{- (a + ck^2) M_{\mathbf{k}} M_{-\mathbf{k}}\} dM_{\mathbf{k}} \right] d\mathbf{k}, \quad (3.10)$$

where

$$\Psi_0 = G_0 = Vg_0.$$

Noting that $a + ck^2$ is proportional to the inverse of the k -dependent susceptibility and that the integral in the logarithm is estimated to be $\chi_k^{1/2}$, (3.10) becomes

$$\Psi \sim \Psi_0 - \frac{V}{\pi^a} \int \ln \chi_k^{1/2} d\mathbf{k}. \quad (3.11)$$

Since χ_k may approximately be replaced by χ_0 for $|k| < \kappa$, the above expression becomes

$$\Psi \sim \Psi_0 - \frac{V}{\pi^a} \int \ln \chi_0^{1/2} d\mathbf{k} \sim \Psi_0 - \frac{V\gamma}{2\pi^a} \varepsilon^{d\nu} \ln \varepsilon. \quad (3.12)$$

In (3.12) the first term on the right-hand side represents the contribution from the short wavelength magnetization density and is expected not to exhibit a singular behavior near the critical point; The singularity in the specific heat will come from the second term. Therefore, leaving only the dominant part, the specific heat C will be expressed as

$$C = -\frac{1}{T} \frac{\partial^2 \Psi}{\partial T^2} = -\frac{1}{T} \frac{\partial^2 \Psi}{\partial \varepsilon^2} \sim \varepsilon^{d\nu-2} \ln \varepsilon. \quad (3.13)$$

The critical index associated with the specific heat (see Table I) is then given by^{3),4),10)}

$$\alpha = d\nu - 2. \quad (3.14)$$

In particular if $d\nu = 2$ C diverges logarithmically. For $T < T_c$ quite an analogous argument leads to

$$\alpha' = d\nu' - 2. \quad (3.15)$$

(c) *Continuation of one phase to the other*

So far our study has been based on (2.11a) and (2.11b) together with (2.31) and (2.32), which are valid if the cell radius R is properly changed with temperature. But such a treatment is useful only when a single phase (e.g. the paramagnetic phase) is under consideration. In order to relate the critical indices associated with one phase to those of the other a slightly different procedure seems to be necessary. In the present and the next subsection we introduce an argument which goes as follows: Down to the temperature $\varepsilon = \varepsilon_0$ R is changed as before, but below ε_0 , R is kept at the value $\sim \varepsilon_0^{-\nu}$:

$$R \sim \varepsilon^{-\nu} \quad \text{for } \varepsilon \geq \varepsilon_0$$

and

$$\sim \varepsilon_0^{-\nu} \quad \text{for } \varepsilon < \varepsilon_0. \quad (3.16)$$

For $\varepsilon \geq \varepsilon_0$ the free energy density (2.11a) is expected to give satisfactory results as has already been seen. But in the region $\varepsilon < \varepsilon_0$ the expression (2.11a) becomes insufficient and cannot describe the phase transition even in an approximate way. Therefore we supplement (2.11a) by the term $bM(\mathbf{r})^4$ and follow Landau's procedure for solving the problem. The results thus obtained are admittedly incorrect in some temperature region below ε_0 . But as we shall see below it is sufficient for our purpose only to ask whether these results are valid again at $\varepsilon = -\varepsilon_0$. We observe that as the temperature is lowered passing through the critical point the fluctuation $M(\mathbf{r}) - \langle M \rangle$ becomes smaller and the condition (2.18) may again be satisfied below a certain temperature. In the following it is proved that this temperature is proportional to $-\varepsilon_0$, the proportionality constant being independent of ε_0 .*) For this purpose it should first be noted that a may quite naturally be assumed to be an analytic and monotonously increasing function of ε since a is a local quantity insensitive to the critical anomaly:**)

*) Implicit in this statement, we are presupposing numerous times of the procedure of changing R for different ε_0 's. Therefore ε_0 may be regarded as a variable.

**) The assumption of the analyticity of a of course breaks down when the free energy G is expressed only in terms of the long range order parameter: $G = G_0 + aM^2 + bM^4 + \dots$. The nonanalyticity of a in this case reflects the fact that when R is changed as before a is no more analytic even when it is in the expression of $g(\mathbf{r})$.

$$\begin{aligned}
 a &= -a_0 + a_1\varepsilon + a_2\varepsilon^2 + \dots \\
 &\simeq -a_0 + a_1\varepsilon \quad (\varepsilon \ll 1).
 \end{aligned}
 \tag{3.17}$$

Here the difference of the two transition temperature obtained by the present and earlier procedures of changing R is taken into account, and this gives rise to the term $-a_0$. The true critical point is of course the one obtained by the earlier procedure. Put this temperature difference divided by the true T_c as $\Delta\varepsilon$. Then $\Delta\varepsilon$ is determined by setting

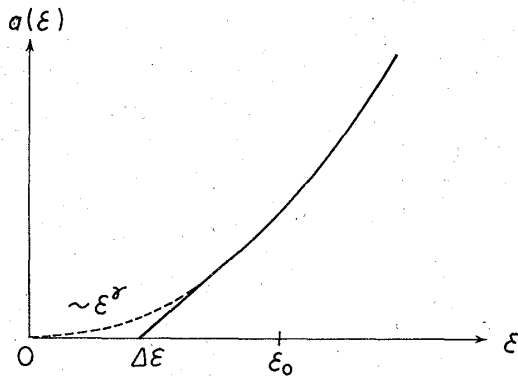
$$a(\Delta\varepsilon) = -a_0 + a_1\Delta\varepsilon = 0$$

or

$$\Delta\varepsilon = a_0/a_1. \tag{3.18}$$

On the other hand, as is easily seen from Fig. 1, $\Delta\varepsilon$ satisfies the inequality

$$0 < \Delta\varepsilon \leq \varepsilon_0 \left(1 - \frac{1}{\gamma}\right). \tag{3.19}$$



Since a_1 should be positive a_0 should also be positive, which is obvious from (3.18) and (3.19). From (3.17), (3.18) and (3.19) we obtain

$$a_1\varepsilon > a(\varepsilon) > a_1 \left\{ \varepsilon - \varepsilon \left(1 - \frac{1}{\gamma}\right) \right\}, \tag{3.20}$$

Fig. 1. Dotted curve behaves like the inverse of the true susceptibility. Solid curve corresponds to the second procedure of changing R (below ε_0 , R is kept constant). The part of the solid curve in the neighborhood of $\varepsilon = \Delta\varepsilon$ is a straight line.

where we have taken into account that $a_0, a_1 > 0$. In effect, it may be put that

$$a(\varepsilon_0) \sim a_1\varepsilon_0, \tag{3.21a}$$

$$a(-\varepsilon_0) \sim -a_1\varepsilon_0. \tag{3.21b}$$

On the other hand b and c will not drastically change near the critical point if R is fixed. In other words, b and c will have the zeroth order term in ε . Therefore we may put (see (2.31))

$$b(-\varepsilon_0) \sim b(\varepsilon_0) \sim \lambda^{-1}\varepsilon_0^{2\gamma-d\nu}, \tag{3.22a}$$

$$c(-\varepsilon_0) \sim c(\varepsilon_0) \sim \varepsilon_0^{\gamma-2\nu}. \tag{3.22b}$$

(3.21b) and (3.22) show that the condition (2.18) is just satisfied at $\varepsilon = -\varepsilon_0$.* This implies that the present procedure gives correct results at $\varepsilon \sim -\varepsilon_0$, though not in the intermediate region between ε_0 and $-\varepsilon_0$.

On taking account that

*) Rigorously speaking, the condition (2.18) is satisfied at $\varepsilon \sim -\varepsilon_0'$, where $\varepsilon_0'/\varepsilon_0$ is independent of ε_0 .

$$a(\varepsilon_0) \sim \chi_0^{-1}(\varepsilon_0) \sim \varepsilon_0^{\gamma}, \quad (3.23)$$

we have from (3.21)

$$-a(-\varepsilon_0) \sim \varepsilon_0^{\gamma'}. \quad (3.24)$$

Indicating the singularity of the susceptibility below T_c by γ' (see Table I), we have from (3.24)

$$\gamma = \gamma'. \quad (3.25)^*)$$

If we put as usual $\kappa \sim |\varepsilon|^{\nu'}$ below T_c , the relation

$$\nu = \nu' \quad (3.26)^*)$$

is directly obtained. This is because the cell radius at $\varepsilon = -\varepsilon_0$ in the present treatment is equal to that at $\varepsilon = \varepsilon_0$ and this cell radius should be proportional to the correlation length at $\varepsilon = -\varepsilon_0$ since whenever the condition (2.18) is satisfied the cell radius is proportional to the correlation length (see (2.26)).

(d) Magnetization

Let us now turn to the spontaneous magnetization. Since the use of the Landau theory at $\varepsilon = -\varepsilon_0$ has been justified, the magnetization at this temperature is given by

$$\langle M \rangle_{\varepsilon = -\varepsilon_0} = \sqrt{-\frac{a(-\varepsilon_0)}{2b(-\varepsilon_0)}}, \quad (3.27)$$

which becomes on using (3.22a) and (3.24) as

$$\langle M \rangle_{\varepsilon = -\varepsilon_0} \sim \varepsilon_0^{-(r-d\nu)/2}. \quad (3.28)$$

From (3.28) and Table I we obtain^{3), 4), 12)}

$$2\beta = d\nu - \gamma. \quad (3.29)$$

(e) Effect of the external field

Returning now to the earlier procedure of changing R , let us investigate the effect of the uniform external field B on the magnetization. Consider below T_c the free energy density of the form

$$g(\mathbf{r}) = g_0 + aM(\mathbf{r})^2 + bM(\mathbf{r})^4 + c[\nabla M(\mathbf{r})]^2 - BM(\mathbf{r}), \quad (3.30)$$

which in the absence of the last term is expected to be valid up to the critical point with a , b and c given by (2.32a) \sim (2.32c). Let us now estimate the value of the additional magnetization near the critical point produced by the external field. The relation between the external field and the field-induced magnetization thus obtained may also hold just at the critical point, because

^{*)} The relations $\gamma = \gamma'$, $\nu = \nu'$ and $\alpha = \alpha'$ are not convincing in the scaling theory (see references 3) and 4)). In the scaling theory one of these is assumed, then the others are derived automatically. Griffiths argued that the unequal values for critical indices above and below the critical point is thermodynamically inconsistent (see reference 11)),

the induced part of magnetization is considered to be approximately constant near the critical point. Therefore for a given B the temperature at which the induced magnetization is calculated may suitably be chosen so that the Landau's treatment may be applied at that temperature. At such a temperature the induced magnetization should be much smaller than the spontaneous one:

$$\lambda \langle M \rangle_{\text{ind}} \sim \langle M \rangle_0 \quad (\lambda \gg 1). \tag{3.31}$$

Here $\langle M \rangle_{\text{ind}}$ and $\langle M \rangle_0$ represents respectively the field-induced and the spontaneous magnetization, and we change ε and B simultaneously so that λ may be kept constant. The total magnetization $\langle M \rangle$ is the sum of these two parts:

$$\langle M \rangle = \langle M \rangle_0 + \langle M \rangle_{\text{ind}}. \tag{3.32}$$

For the condition (3.31) to be satisfied, the induced magnetization for different values of B should be calculated at different temperatures. $\langle M \rangle_{\text{ind}}$ can be obtained by minimizing $g(\mathbf{r})$ with respect to $\langle M \rangle$:

$$\frac{d}{d\langle M \rangle} (a\langle M \rangle^2 + b\langle M \rangle^4 - B\langle M \rangle) = 0. \tag{3.33}$$

On the other hand, $\langle M \rangle_0$ satisfies

$$\frac{d}{d\langle M \rangle_0} (a\langle M \rangle_0^2 + b\langle M \rangle_0^4) = 0. \tag{3.34}$$

From (3.32), (3.33) and (3.34) and noting (3.31) we have

$$B \simeq -4a\langle M \rangle_{\text{ind}}. \tag{3.35}$$

Recalling that

$$\langle M \rangle_0 \sim |\varepsilon|^\beta \sim |a|^{\beta/\tau},$$

(3.35) becomes

$$\begin{aligned} B &\sim \langle M \rangle_0^{\tau/\beta} \langle M \rangle_{\text{ind}}, \\ &\sim \lambda^{\tau/\beta} \langle M \rangle_{\text{ind}}^{1+(\tau/\beta)}. \end{aligned} \tag{3.36}$$

This is the relation between the applied field and the induced magnetization near the critical point, and this relation may hold at the critical point because of the reason mentioned previously. Thus we obtain^{3,4)} (see Table I)

$$\delta = 1 + \frac{\tau}{\beta}. \tag{3.37}$$

§ 4. Discussion

On the basis of the natural assumptions, we have succeeded in modifying the classical treatment of the second order phase transition and in deriving the relations among the critical indices (Eqs. (3.6), (3.14), (3.15), (3.25), (3.26),

(3.29) and (3.37)). All these relations are equivalent to those derived from the static scaling law.^{3),4)} Through the process of the modification, the connection has considerably been clarified between the classical picture and the more realistic mechanism of the second order phase transition. It should be remarked that our most important assumption is (2.21). At some points this assumption seems to be connected with the similarity assumption in the static scaling theory.

One serious discrepancy between our conclusion and the expected feature is the functional form of the pair correlation below the critical point (see § 3 (a)). Although the reason for this is not quite clear, it is probable that the form of $g(\mathbf{r})$ given by (2.5) is no more sufficient below the critical point even if a , b and c behave like (2.32). If this is true, it is remarkable that the critical indices derived from such an insufficient free energy are related to each other in a correct way. Since our purpose in this paper was only to remove the apparent inconsistency involved in the Landau theory, there seems to be no reason why the additional terms should be put in $g(\mathbf{r})$. But a further study seems to be necessary to clarify this point.

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