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Orientational phase transitions in the vicinity of the Curie point in terbium-gadolinium alloys

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By measurements of the magnetization and of the magnetocaloric effect in monocrystals of the rare-earth alloys Tb_xGd_{1-x} ($x < 0.94$) along various crystallographic directions, it is shown that in the region of the Curie temperature, in a magnetic field directed along an axis of difficult magnetization, a magnetic phase transition of the spin-reorientation type occurs in an anisotropic ferromagnet. The experimental results are discussed on the basis of Landau's thermodynamic theory of phase transitions. By means of the Ginzburg-Levanyuk criterion, the theory is shown to be applicable over a quite broad temperature interval near the Curie point.

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In the study of magnetic phase transitions at the Curie point (of the order-disorder type), use is often made of Landau's theory of phase transitions of second order.[1] On the basis of it, there exists for an isotropic magnetic material a well developed thermodynamic-coefficient procedure[2] that enables one to determine the spontaneous magnetization $\sigma_s(T)$ and also the Curie point Θ.

But in a strongly anisotropic ferromagnet in the presence of a magnetic field, there is a possibility, in the temperature range $T < \Theta$ below the Curie point, of reorientation phenomena, which may produce changes in the phase-transition picure near Θ and, accordingly, may lead to a change of the physical properties of the magnet. Here the order parameter in the theory of a phase transition is, in contrast to the isotropic case, multicomponent. The effect of the anisotropy of the ferromagnet manifests itself in the fact that the vanishing of the thermodynamic coefficients of the second-order terms in the expansion of the thermodynamic potential will occur at different temperatures for different components. This, in particular, may lead to anisotropy of the paramagnetic Curie point.[3,4]

The present paper is devoted to study of the influence of phenomena of the reorientation type on phase transitions in the Curie-point region of a uniaxial magnetic material. The experimental investigations were made on Tb_xGd_{1-x} alloys with various contents of gadolinium ($x < 0.94$). These compounds are solid solutions and provide a typical example of a strongly anisotropic uniaxial ferromagnetic crystal. They have a hexagonal structure, with the axis of hard magnetization along the hexagonal axis c.

The technology of growing monocrystalline terbium-gadolinium alloys and the monitoring of their quality have been described earlier.[5]

The literature[6] contains information about the investigation of the magnetic properties of gadolinium in a magnetic field directed along the axes of easy and of hard magnetization. Gadolinium, however, has a magnetic anisotropy two orders of magnitude smaller than the anisotropy of heavy rare-earth metals. Therefore Tb_xGd_{1-x} alloys are of considerable interest, since no investigation has hitherto been made of high-anisotropy ferromagnets, near the Curie point, in a magnetic field directed along the axis of hard magnetization.

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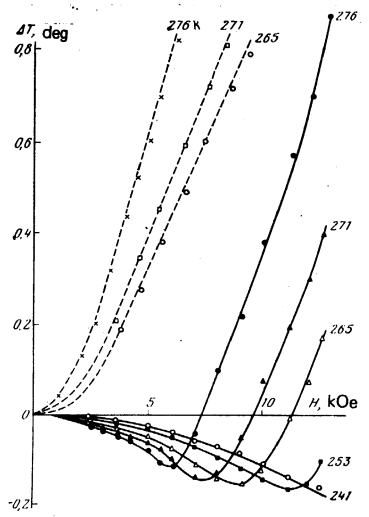

FIG. 1. Variation of the magnetocaloric effect with field in the alloy $Tb_{0.09}Gd_{0.91}$, in a field applied along the hexagonal axis c (solid curve) and along the axis b (dotted curve), in the region of the Curie temperature.

In the present work, measurements were made of the magnetization and of the magnetocaloric effect in fields along the c axis and in the basal plane. It should be noted that the magnetocaloric effect is very informative in the study of magnetic phase transitions, since it is very sensitive to changes of the magnetic structure.[7] This effect occurs during any changes of the entropy of the magnetic subsystem, and in a number of cases a reversal of the sign of the effect is observed when the character of the magnetic ordering changes.[8]

The curves in Fig. 1 show the variation of the magnetocaloric effect (ΔT effect) with magnetic field for the alloy $Tb_{0.09}Gd_{0.91}$ near the Curie point, for the cases in which the field is directed along axes b and c. The $\Delta T(H)$ curves for the b axis have the character that is usual for ferromagnets: $\Delta T > 0$, and the magnetocaloric effect increases monotonically with the field. On the $\Delta T(H)$ curves along the c axis, the absolute value of the ΔT effect increases in weak fields, where $\Delta T < 0$. But beginning with a certain threshold field H_{th} is zero at the Curie point (determined from measurements of the magnetization in a field directed in the basal plane). Below Θ, the field H_{th} increases sharply in magnitude (Fig. 2).

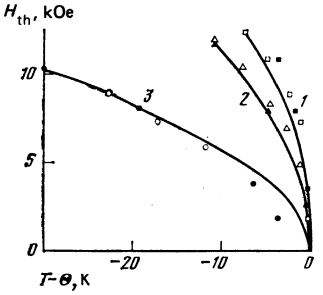

FIG. 2. Theoretical and experimental relations $H_{th}(T)$ for the alloys $Tb_{0.50}Gd_{0.50}$ (Curve 1), $Tb_{0.20}Gd_{0.80}$ (Curve 2), and $Tb_{0.09}Gd_{0.91}$ (Curve 3). The dark points were obtained from measurements of the magnetization, the light points from measurements of the ΔT effect.

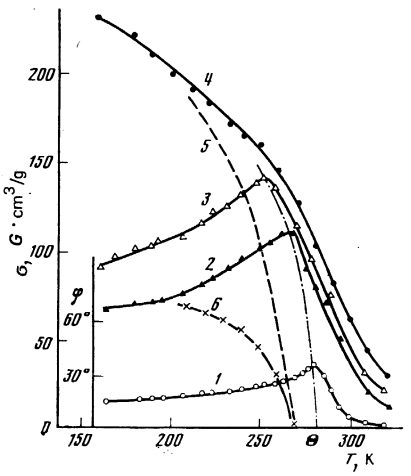

FIG. 3. Temperature variations of the magnetization in a field applied along the c axis, with $H = 1.7$ kOe (Curve 1), 8 kOe (Curve 2), and 12 kOe (Curve 3); and in a field of 8 kOe applied along the axis b (Curve 4). Curve 5 shows $\delta_\perp$ calculated by formula (11) in field 8 kOe. Curve 6 shows the temperature variation of the angle φ that the magnetization vector makes with the c axis, at field 8 kOe. The alloy is $Tb_{0.09}Gd_{0.91}$.

The temperature dependence of the magnetization in a field directed in the basal plane has the same character as in cubic ferromagnets with small magnetic anistropy: a monotonic decrease of the magnetization on increase of temperature, with a sharp drop of σ_s near Θ (Fig. 3, Curve 4).

The curves of temperature variation of the magnetization $\sigma(T)$, measured in a constant magnetic field along the c axis, reveal a maximum in all the Tb_xGd_{1-x} alloys. In weak fields it is close to the Curie point, but on increase of field it shifts toward lower temperatures (Fig. 3, Curves 1, 2, and 3). This maximum occurs at the temperature for which the given constant magnetic field is the threshold field, at which a sharp bend is observed on the $\Delta T(H)$ curves. Figure 2 shows the temperature variations of the threshold field, or transition field, H_{th} for terbium-gadolinium alloys. The values of H_{th} determined by the two methods—from the maximum of the $\sigma(T)$ curve measured in a constant magnetic field along the c axis, and from the sharp bend in the curve $\Delta T(H)$ of the magnetocaloric effect—practically coincide. With increase of the terbium content in the alloys, the $H_{th}(T)$ curve rises more abruptly on cooling below the point Θ This can be explained by the fact that with increase of the constant of uniaxial magnetic anisotropy, larger values of the magnetic field are required for rotation of the magnetization vector into the direction of the c axis.

The experimental results obtained can be explained by the fact that the magnetic phase transition near the Curie point of an anisotropic ferromagnet that retains a large magnetic anisotropy even near Θ has an entirely different character in a magnetic field along the b axis and along the c. In the first case, the magnetic field produces an intense paraprocess near the paramagnetism-ferromagnetism transition temperature. In the second case, the field induces not only a paraprocess but also spin reorientation: rotation of the magnetic moment of

the crystal from the basal plane toward the hexagonal axis along which the magnetic field is directed.

The experimental results obtained on spin reorientation near the Curie point, in a magnetic field applied along the axis of hard magnetization, can be interpreted on the basis of Landau's thermodynamic theory of phase transitions of second order.[1]

We write the thermodynamic potential of a uniaxial ferromagnet near the Curie point for the case of axial symmetry, when the magnetic field is oriented along the hard axis of magnetization (the c axis):

$$\Phi=\frac{\alpha_1}{2}\sigma_\perp^2+\frac{\alpha_2}{2}\sigma_\parallel^2+\frac{\beta_1}{4}\sigma_\perp^4+\frac{\gamma}{2}\sigma_\perp^2\sigma_\parallel^2+\frac{\beta_2}{4}\sigma_\parallel^4-\sigma_\parallel H, \tag{1}$$

where $\sigma_\perp$ and $\sigma_\parallel$ are, respectively, the projections of the magnetization vector on the basal plane and on the c axis; α_1, α_2, β_1, β_2, and γ are thermodynamic coefficients; and H is the external field.

For existence of a phase transition of the second kind, it is necessary that a thermodynamic coefficient of the second-order terms vanish and that the quadratic form of the higher-order terms be positive definite in the vicinity of the phase transition; that is,

$$\beta_1\beta_2-\gamma^2>0. \tag{2}$$

In the case of a uniaxial magnet with axis of hard magnetization along c, $\alpha_2>\alpha_1$ always, so that α_1 vanishes at a higher temperature than α_2; therefore

$$\alpha_1=a_1(T-\Theta^{(1)}),\quad \alpha_2=a_2(T-\Theta^{(2)}),\quad \Theta^{(1)}>\Theta^{(2)}. \tag{3}$$

The necessary conditions for a minimum of the thermodynamic potential (1) lead to the equations

$$\partial\Phi/\partial\sigma_\perp=(\alpha_1+\beta_1\sigma_\perp^2+\gamma\sigma_\parallel^2)\sigma_\perp=0, \tag{4}$$

$$\partial\Phi/\partial\sigma_\parallel=(\alpha_2+\beta_2\sigma_\parallel^2+\gamma\sigma_\perp^2)\sigma_\parallel-H=0. \tag{5}$$

These equations show that there are two phases in the magnet.

A. High-symmetry phase $\sigma_\perp = 0$

The high-symmetry (paramagnetic) phase is characterized by the fact that when $H\neq 0$, we have $\sigma_\perp=0$. Then, as is evident from the condition (5), $\sigma_\parallel$ is described by the equation

$$a_2(T-\Theta^{(2)})+\beta_2\sigma_\parallel^2=H/\sigma_\parallel. \tag{6}$$

The conditions for stability of this phase have the form

$$\partial^2\Phi/\partial\sigma_\parallel^2=\alpha_2+3\beta_2\sigma_\parallel^2>0, \tag{7}$$

$$\partial^2\Phi/\partial\sigma_\perp^2=\alpha_1+\gamma\sigma_\parallel^2>0, \tag{8}$$

since $\partial^2\Phi/\partial\sigma_\perp\partial\sigma_\parallel=0$.

Condition (7) is satisfied if condition (6) is. Therefore the high-temperature phase loses its stability when the condition (8) is violated, i.e., when

$$\sigma_\parallel^2=a_1(T-\Theta^{(1)})/\gamma. \tag{9}$$

On substituting formula (9) in equation (6), we find the limit of stability of this phase:

$$H_{th}=\left[\frac{a_1}{\gamma}(\Theta^{(1)}-T)\right]^{1/2}\left[a_2(T-\Theta^{(2)})+\frac{a_1\beta_1}{\gamma}(\Theta^{(1)}-T)\right]. \tag{10}$$

The curve $H_{th}(T)$ is a curve of phase transition of second order. Decrease of $H_{th}(T)$ as $T\to\Theta^{(1)}$ occurs according to a $(\Theta^{(1)}-T)^{1/2}$ law. The temperature $\Theta^{(1)}$ is essentially an "ordinary" Curie point, below which, when $H=0$, a spontaneous magnetization $\sigma_\perp\neq 0$ appears ($\sigma_\parallel=0$). But the paramagnetic Curie point $\Theta^{(2)}$, as is seen from the magnetization equation (6), does not coincide with the ordinary Curie point $\Theta^{(1)}$.

B. Low-symmetry (ferromagnetic) phase $\sigma_\perp \neq 0$

From Eqs. (4) and (5), we get for $\sigma_\perp\neq 0$

$$\sigma_\perp^2=-\frac{\alpha_1}{\beta_1}-\frac{\gamma}{\beta_1}\sigma_\parallel^2, \tag{11}$$

$$\alpha_2-\alpha_1\frac{\gamma}{\beta_1}+\left(\beta_2-\frac{\gamma}{\beta_1}\right)\sigma_\parallel^2=\frac{H}{\sigma_\parallel}. \tag{12}$$

It is interesting that the magnetization equation (12) is similar in form to equation (6) for the paramagnetic phase.

The conditions for stability of the low-symmetry phase with $\sigma_\perp\neq 0$ have the form

$$\partial^2\Phi/\partial\sigma_\perp^2=2\beta_1\sigma_\perp^2>0, \tag{13}$$

$$\partial^2\Phi/\partial\sigma_\parallel^2=\frac{H}{\sigma_\parallel}+2\beta_1\sigma_\parallel^2>0, \tag{14}$$

$$\frac{\partial^2\Phi}{\partial\sigma_\perp^2}\frac{\partial^2\Phi}{\partial\sigma_\parallel^2}-\left(\frac{\partial^2\Phi}{\partial\sigma_\perp\partial\sigma_\parallel}\right)^2=2\beta_1\sigma_\perp^2\left\{\frac{H}{\sigma_\parallel}+2\left(\beta_1-\frac{\gamma^2}{\beta_2}\right)\sigma_\parallel^2\right\}>0. \tag{15}$$

From the last relations (if we also take Eq. (2) into account) it is seen that destruction of the stability of the low-symmetry phase occurs when $\sigma_\perp=0$, which accordint to (11) is equivalent to the condition

$$\sigma_\parallel^2=-\alpha_1/\gamma. \tag{16}$$

On substituting (16) in (12), we again get a phase-transition curve described by Eq. (10). The phase diagram for the case under consideration is shown in Fig. 4. To the left of the solid curve in the (H, T) plane, the phase with $\sigma_\perp\neq 0$ exists; to the right of this curve, the high-symmetry phase with $\sigma_\perp=0$.

We shall now consider the features of the magnetocaloric effect on the basis of the above-described behavior of the magnetic system of the ferromagnet.

Thermodynamic relations lead to the following expression for the magnetocaloric effect:

$$\frac{dT}{dH}=-\frac{T}{C_H}\frac{\partial\sigma_\parallel}{\partial T}, \tag{17}$$

where C_H is the specific heat at constant magnetic field H.

In the high-symmetry phase ($\sigma_\perp=0$), we have from Eq. (6)

$$\frac{1}{\sigma_\parallel}\frac{\partial\sigma^{(A)}}{\partial T}=-\frac{\partial\alpha_2/\partial T+\sigma_\parallel^2\partial\beta_2/\partial T}{\alpha_2+3\beta_2\sigma_\parallel^2}, \tag{18}$$

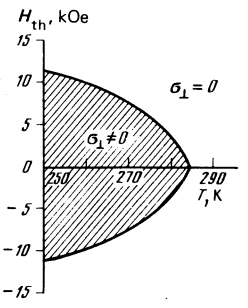

FIG. 4. Phase diagram for the alloy $Tb_{0.09}Gd_{0.91}$.

therefore

$$\frac{dT^{(A)}}{dH}=\frac{T}{C_H^{(A)}}\sigma_\parallel\frac{\partial\alpha_2/\partial T+\sigma_\parallel^2\partial\beta_2/\partial T}{\alpha_2+3\beta_2\sigma_\parallel^2}. \quad (19)$$

Hence it is evident that when the fields are not too large and when the coefficient $\beta_2(T)$ varies only slightly with temperature, $dT/dH>0$ in the paramagnetic phase.

In the low-symmetry phase ($\sigma_\perp\neq 0$), we get from (12)

$$\frac{dT^{(B)}}{dH}=\frac{T}{C_H^{(B)}}\sigma_\parallel\Big[\frac{\partial\alpha_2}{\partial T}+\sigma_\parallel^2\frac{\partial\beta_2}{\partial T}$$
$$-\frac{\partial}{\partial T}\Big(\alpha_1\frac{\gamma}{\beta_1}\Big)-\sigma_\parallel^2\frac{\partial}{\partial T}\Big(\frac{\gamma^2}{\beta_1}\Big)\Big]$$
$$\times\Big[\alpha_2-\alpha_1\frac{\gamma}{\beta_1}+3\Big(\beta_2-\frac{\gamma^2}{\beta_1}\Big)\sigma_\parallel^2\Big]^{-1}. \quad (20)$$

Thus on the curve of phase transition from the paramagnetic to the low-temperature phase, a sharp bend should be observable on the field-dependence curve of the magnetocaloric effect $\Delta T(H)$. In the low-symmetry phase ($\sigma_\perp\neq 0$), even a change of sign, $dT^{(B)}/dH<0$, is possible, as is observed in terbium-gadolinium alloys (see Fig. 1).

It should be noted that the temperature interval within which molecular-field theory is applicable is limited by the growth of fluctuations of the order parameters near the phase-transition point. According to the phenomenological theory of Ginzburg and Levanyuk,[9,10] the magnitude of this temperature interval near $T=\Theta^{(1)}$, for $H=0$, is

$$\frac{T-\Theta^{(1)}}{\Theta^{(1)}}=\frac{\Delta T}{\Theta^{(1)}}=\frac{9\beta_1^2(k\Theta^{(1)})^2I_0^6}{(4\pi)^2A^3a_1\Theta^{(1)}}, \quad (21)$$

where A is an exchange constant.

By using the experimentally determined values $\beta_1\approx 3.9\cdot10^{-5}$ G^{-2}, $a_1=\partial\alpha_1/\partial T|_\Theta\approx 2.1$ deg^{-1}, $A\approx 2.6\cdot10^{-6}$erg/cm, $I_0\approx 2.15\cdot10^3$ G, and $\Theta\approx 3.10^2$ deg, we get, according to formula (21), $\Delta T/\Theta\approx 10^{-3}$.

This estimate shows that the thermodynamic theory developed may be used to describe the experimental results obtained, except within a quite narrow temperature interval (~0.3 degree).

We shall make an analysis of the experimental data on the basis of the thermodynamic theory presented.

Figure 5 shows the relation $H/\sigma_\parallel=f(\sigma_\parallel^2)$ for a field directed along the hexagonal axis c. According to equation (6), for $H>H_{th}$ a linear variation of $H/\sigma_\parallel$ with $\sigma_\parallel^2$

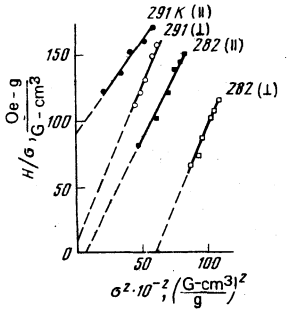

FIG. 5. Variation of H/σ with σ^2 for the alloy $Tb_{0.09}Gd_{0.91}$ in a field applied along the c axis and for a field applied in the basal plane.

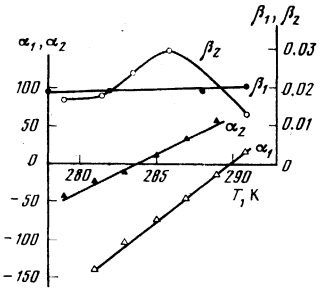

FIG. 6. Temperature variation of thermodynamic coeffecients in the slloy $Tb_{0.09}Gd_{0.91}$.

should be observed; this is confirmed experimentally (Fig. 5). When the magnetic field is directed in the basal plane (along an axis of easy magnetization), as is easily shown by minimization of the thermodynamic potential (1), the equation that holds is

$$\alpha_1+\beta_1\sigma_\perp^2=H/\sigma_\perp, \quad (22)$$

this also describes well the experimental data near the Curie point (Fig. 5).

The thermodynamic coefficients α_1, α_2, β_1, and β_2 for the alloy $Tb_{0.09}Gd_{0.91}$, found from measurements of the magnetization in fields $H\|c$ and $H\perp c$, are shown in Fig. 6. The temperatures $\Theta^{(1)}$ and $\Theta^{(2)}$ at which α_1 and α_2 vanish differ by almost 6 K, and $\Theta^{(2)}<\Theta^{(1)}$.

Another conclusion characteristic of the theory of phase transitions of the second kind is that $H_{th}\sim(T_K^{(1)}-T)^{1/2}$, where $T_K^{(1)}=\Theta$; that is, $T_K^{(1)}$ coincides with the Curie point. The theoretical curves plotted in Fig. 2 show that H_{th} does in fact increase as $(\Theta-T)^{1/2}$ with distance from the Curie point, although some deviations are observed near Θ; these are presumably due to the presence near Θ of fluctuations, which the Landau theory does not take into account.

Equation (11) enables us to calculate $\sigma_\perp$ in the low-symmetry phase for a given value of the magnetic field (the dotted curve 5 in Fig. 3). It is seen that $\sigma_\perp=0$ on the curve $H_{th}(T)$ of the phase diagram and increases monotonically on cooling. The angle φ that the magnetic moment of the crystal makes with the c axis is zero on the curve of the phase diagram that separates the low-symmetry phase from the high-symmetry, and it increases rapidly on cooling (Curve 6, Fig. 3).

Figure 4 shows the magnetic phase diagram in the (H, T) plane. To the left of the solid curve, the phase with $\sigma_\perp\neq 0$ exists; to the right of this curve, the high-symmetry phase with $\sigma_\perp=0$.

Thus we may conclude that in ferromagnets with strong magnetic anisotropy, such as the rare-earth terbium-gadolinium alloys, there is a magnetic phase transition of the spin-reorientation type near the Curie point in a field directed along the axis of hard magnetization. This transition is well described by Landau's thermodynamic theory of phase transitions and is a phase transition of the second kind.

Reorientation phenomena lead to a distinctive temperature variation $\sigma_\parallel(T)$ of the magnetization in a magnetic field near the Curie point, and accordingly to peculiar-

ities of the magnetocaloric effect (see Figs. 1 and 3). It is obvious that the above-described behavior of the magnetic system in the vicinity of the Curie point should manifest itself also in others of its physical properties, such as, for example, the specific heat, the magnetostriction, and the scattering of light.

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Effect of fluctuations on the properties of the phase transition from a nematic liquid crystal to an isotropic liquid

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The critical properties are investigated of a model in which the order parameter is a symmetrical zero-trace $n \times n$ tensor. The particular case $n = 3$ corresponds to the model of the nematic liquid crystal–isotropic liquid phase transition. It is shown that the critical anomalies near this transition can be due to the specific properties of the interaction of the fluctuations of the order-parameter tensor field. An experimental method is proposed with which to establish the cause of the pre-transition anomalies.

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INTRODUCTION

We employ in this paper the method of field renormalization groups (RG) for the investigation of the critical behavior of the Q-model, in which the order parameter is by definition a zero-trace $n \times n$ tensor. Particular interest attaches to the case $n = 3$, which corresponds to the model proposed by de Gennes for the description of phase transitions of the type nematic liquid crystal-isotropic liquid (NLC–IL).[1] Most of them are accompanied by strong pre-critical phenomena, and the Landau expansion for the free energy contains a third-order invariant. Therefore, in the spirit of the predictions of the phenomenological theory, the anomalous behavior of the thermodynamic quantities near the NLC–IL transition was previously attributed to the presence, on the curve of the first-order phase transitions (or near this curve), of an isolated singular point at which the coefficient of this invariant vanishes. The critical properties of the NLC–IL transition, assuming that such a point exists, were investigated by Vigman, Larkin, and Filev, as well as by Lubensky and Priest.[3,4]

Recently Gorodetskiĭ and Zaprudskiĭ,[5] and independently of them one of us[6] have proposed another explanation for the appearance of critical anomalies near this phase transition, without resorting to the assumption that is close to an isolated singular point. It was shown that in contrast to the conclusions of the phenomenological theory the NLC–IL transition can be continous if the system of the RG equations has a scale-invariant solution satisfying definite conditions, and the assumption was advanced that the experimental values of the critical exponents at $\gamma \approx 1.0$ for the susceptibility and $\alpha = 0.3-0.5$ for the heat capacity can pertain to the fluctuation region and not to the region of the Landau theory. In this case the proximity of the exponent γ to unity can be attributed to mutual cancellation of the contribution made to it by the triple or quadruple vertices. For the value $n = 3$ the scale-invariant solution of the system of the RG equations in the single-loop approximation was obtained by Gorodetskiĭ and Zaprudskiĭ[5] without taking into account the renormalization of the Green's function, a procedure which in general is inconsistent for the given problem. Nor did they ascertain whether this solution satisfies the condition that the phase transition be continuous.

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