Polarized defects and anomalies in the properties of crystals at phase transitions

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A continuum approach is used to study theoretically the behavior of the thermodynamic quantities, order-parameter correlation functions, and certain kinetic coefficients for crystals containing defects giving the same sign of the order parameter (polarized defects). It is taken into account that real defects consist of both the "random local field" and "random local transition temperature" type. For cases in which the field component of the defect is not particularly small, a theory is constructed which is valid over an entire temperature interval including the phase transition point of the ideal crystal. The temperature dependence of the thermodynamic quantities and of certain kinetic coefficients has the same qualitative character as in the defectless crystal if allowance is made for the shift of the transition temperature and the presence of an effective field conjugate to the order parameter. However, under certain conditions effects due to the randomness of the defect distribution can be appreciable. These effects also determine completely the elastic scattering of light and neutrons and the defect contribution to the anomaly in the diffuse x-ray scattering. It is shown that the scattering intensity can have a rather nontrivial dependence on the temperature and on the defect concentration, owing to the interference of the contributions from the different components of the defect. The possibilities of an experimental check of the theory are discussed.

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The theoretical papers which have been published on the influence of defects on the properties of materials near phase-transition points can be divided into two groups. One (see, e.g., Refs. 1–4 and the literature cited therein) consists of studies of rather narrow neighborhood of the transition point—the so-called scaling region—which has yet ot be reliably detected for the case of structural phase transitions. The other group, smaller in number, consists of studies whose results pertain to temperatures which are not too close to the transition point and to defect concentrations which are not too large (see, e.g., Refs 5–8). In this case the defects can be treated as independent, and their contributions to the physical quantities can be assumed additive. This approach is clearly valid when the order-parameter correlation length r_c, which determines the size of the perturbation region around a defect, is smaller than the average distance between defects.

In the present paper we consider a case in which one can construct a theory which in fact applies to a whole range of temperatures, including the phase transition temperature in the ideal crystal. This is the case of so-called "polarized defects." Let us explain what is meant by this term.

In describing the defect-caused matrix distortions one usually distinguishes between defects of the "random local transition temperature" type (T defects) and "random local field" type in various state (S, P, etc. defects).[7,8,5] These are discussed in more detail in Sec. 1. A real defect can, of course, include both types of contribution and different states simultaneously, and we shall therefore refer to the T, S, and P components of a defect. A system of defects is called polarized when the signs of the S components of the defects are the same. In this paper we shall consider only frozen defects whose orientation and distribution do not change with temperature.

Obviously, in a crystal with polarized defects there is no phase transition in the true sense of the word, i.e., the correlation length r_c of the order parameter remains finite at all temperatures. It is preciesely this circumstance which permits one ultimately to obtain results which pertain to an entire temperature interval including the phase transition point of the defectless crystal.

In constructing the theory we take into account all of the interesting components of the defect. We note that even in the case when the S component of the defect is small or entirely absent, i.e., the concept of polarization of the system of defects loses meaning, the results given below are valid for a substantially wider region than the results obtained in the isolated-defect approxmation,[5] even though one cannot construct a theory which is valid for the entire range of temperatures.

An important feature of the approach developed below is the use of the continuum approximation for describing the distortions caused in the crystal by the defect. This approach is valid if the defect concentration is sufficiently small: $Nd^3 \ll 1$, where N is the defect concentration and d is the dimension of the core of the defect.[5] We note that in considering defects in the framework of the microscopic theory, the authors of Refs. 9 and 10 used the self-consistent-field approximation, for which the applicability condition is that the average distance between defects be small compared to the effective range of the forces. Since this range is of the same order of magnitude as d, the applicability condition for the approach of Refs. 9 and 10 is the direct opposite of our condition $Nd^3 \ll 1$.

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Crystals with polarized defects are of great interest from an experimental standpoint. It is defects of the polarized type that are created through radiation damage of a crystals in an asymmetric phase, during growth of a crystal below the phase transition point, and also in the processing of a sample into a single-domain state. In interpreting the experimental data it is customary to take the presence of polarized defects into account by introducing an effective field conjugate to the order parameter and also a shift of the transition temperature. Although such an approach is qualitatively reasonable it does not take into account effects due to the randomness of the spatial distribution of the defects (fluctuations in the defect concentration). In this paper we employ a continuum-theory approach to evaluate the effective field and the shift of the transition temperature and to calculate the contributions to various quantities from effects due to fluctuations in the defect concentration. Under certain conditions these fluctuations necessitate appreciable corrections to the expressions for the thermodynamic quantities and for certain kinetic coefficients as evaluated with allowance for the effective field and the shift of the transition temperature. They also govern the elastic light-scattering and neutron-scattering intensity and the intensity of diffuse x-ray scattering.

By considering effects due to the randomness of the defect distribution, one can understand on the basis of simple considerations that the relative role of the various defect components changes as the concentration N changes. For an isolated defect the S component gives the most strongly temperature dependent contribution to all the physical quantities,[11] i.e., the contribution containing the highest power of the correlation length r_c. This statement, of course, pertains to the case of low concentrations of defects. As the concentration of polarized defects increases, the effective field produced by these defects grows. This field eliminates the divergence of r_c and leads to a decrease in the contribution of an individual defect to the anomalies. Naturally, this decrease is most important for the S-component contributions, which contain the highest power of r_c. One can therefore expect that at large concentrations N the contribution of the T and P components will become predominant.

The investigation below pertains to the case of structural phase transitions which are not proper ferroelectric or ferroelastic transitions. In these last two cases it is necessary to take into account the long-range forces arising in the presence of spatial inhomogeneities of the order parameter. These cases must be treated separately.

1. SPATIAL DISTRIBUTION OF THE ORDER PARAMETER

Throughout the rest of this paper we consider the simplest case of a one-component order parameter η. In this case the free energy density of an ideal crystal is of the form

$$\frac{A}{2}\eta^2+\frac{B}{4}\eta^4+\ldots-h\eta+\frac{D}{2}(\nabla\eta)^2\equiv\varphi(\eta)+\frac{D}{2}(\nabla\eta)^2, \tag{1}$$

where h is the generalized field conjugate to the order parameter η. In the region where Landau theory applies we have

$$A=A_0(T-T_c)/T_c\equiv A_0\tau, \qquad B=\text{const}.$$

Allowance for the presence of defects is made by including in (1) terms of the type

$$\frac{A_d(\mathbf{r})}{2}\eta^2; \quad -h_d(\mathbf{r})\eta; \quad -g_d(\mathbf{r})\frac{\partial\eta}{\partial x}, \tag{2}$$

where x denotes one of the crystallographic directions. These terms correspond respectively to the T, S, and P components of the defect. All the remaining terms taking into account the defect contribution to the free energy of the crystal are of no interest in the study of anomalies near phase transitions points. In the present paper we consider only polarized point defects, which do not create appreciable elastic stresses in their neighborhoods. For such defects

$$A_d=A_1V_1\sum_i\delta(\mathbf{r}-\mathbf{r}_i), \quad h_d=h_1V_1\sum_i\delta(\mathbf{r}-\mathbf{r}_i),$$
$$g_d=g_1V_1\sum_i\delta(\mathbf{r}-\mathbf{r}_i), \tag{3}$$

where $V_1=(4\pi/3)d^3$ is the volume of the core of the defect and $\mathbf{r}_i$ is the coordinate of the ith defect.

It will be convenient to separate out in all the expressions the volume average $\bar\eta$ of the order parameter. We recall that a feature of the polarized defects under study is that they have a nonzero $\bar\eta$ at all temperatures. Taking into account the contributions (2) of all three defect components, we write the expression for the free energy and the corresponding equation of state as expansions in powers of $(\eta-\bar\eta)$ in the form

$$\Phi=V\left[\varphi(\bar\eta)+\frac{\bar A_d}{2}\bar\eta^2-\bar h_d\bar\eta\right]+\int d\mathbf{r}\left\{\frac{\varphi''(\bar\eta)}{2}(\eta-\bar\eta)^2+\frac{\varphi'''}{3!}(\eta-\bar\eta)^3\right.$$
$$+\frac{\varphi^{\mathrm{IV}}}{4!}(\eta-\bar\eta)^4+\ldots+\frac{D}{2}[\nabla(\eta-\bar\eta)]^2+\frac{A_d}{2}(\eta-\bar\eta)^2+A_d\bar\eta(\eta-\bar\eta)$$
$$\left.-h_d(\eta-\bar\eta)-g_d\frac{\partial(\eta-\bar\eta)}{\partial x}\right\}, \tag{4}$$

$$D\Delta(\eta-\bar\eta)=(\varphi''+A_d)(\eta-\bar\eta)+\varphi'(\bar\eta)-h_d+A_d\bar\eta$$
$$+\frac{\partial g_d}{\partial x}+\frac{\varphi'''}{2}(\eta-\bar\eta)^2+\frac{\varphi^{\mathrm{IV}}}{6}(\eta-\bar\eta)^3+\ldots; \tag{5}$$

here V is the volume of the crystal. We note that whereas in Ref. 5 the defects assigned the boundary conditions for the equation of a continuous medium, here they enter Eq. (5) in the form of inhomogeneous terms and a coefficient of $(\eta-\bar\eta)$. The two approaches are equivalent and lead to identical results.

Let us solve Eq. (5) in the approximation linear in $(\eta-\bar\eta)$, determining the average value of the order parameter from the self-consistency condition

$$\langle\eta-\bar\eta\rangle=0, \tag{6}$$

where the angle brackets denote an average over the random spatial distribution of the defects.

Let us first consider the case of S defects, i.e., let us set $A_d=g_d=0$. Taking (3) into account, we find for this case the solution of (5) in the linear approximation:

$$(\eta-\overline{\eta})_{(\mathrm{I})}=-\varphi'/\varphi''+\eta_0\sum_i \varphi(\mathbf{r}-\mathbf{r}_i), \tag{7}$$

where $\eta_0 = h_1V_1/4\pi Dd$ has the meaning of the order parameter at the core of the defect,

$$\varphi(\mathbf{r})=(d/r)\exp(-r/r_c),$$

while the correlation length r_c is given by the expression

$$r_c^2=D/\varphi''(\overline{\eta}_{(\mathrm{I})}). \tag{8}$$

Using solution (7), we obtain for the self-consistency condition (6)

$$\varphi'(\overline{\eta}_{(\mathrm{I})})=NV_1h_1, \tag{9}$$

i.e., $\overline{\eta}_{(I)}$ has the meaning of the average value of the order parameter in the effective field $h_{\mathrm{eff}} = NV_1h_1$.

The first nonlinear correction to solution (7) is of the form

$$(\eta-\overline{\eta})_{(\mathrm{II})}=\int\frac{\varphi(\mathbf{r}-\mathbf{r}')}{4\pi Dd}d\mathbf{r}'\left\{\frac{\varphi'''}{2}(\eta-\overline{\eta})_{(\mathrm{I})}^2+\frac{\varphi^{\mathrm{IV}}}{6}(\eta-\overline{\eta})_{(\mathrm{I})}^3\right\}. \tag{10}$$

Using (6) and (10) and evaluating the corresponding integrals, we obtain the correction to the average value:

$$\overline{\eta}_{(\mathrm{II})}=-6\pi\left(\frac{\eta_0}{\eta_{\mathrm{at}}}\right)^2Nr_c^3\overline{\eta}_{(\mathrm{I})}-4\pi\frac{\eta_0^3}{\eta_{\mathrm{at}}^2}Ndr_c^2\ln\frac{3r_c}{d}, \tag{11}$$

where $\eta_{\mathrm{at}} = (D/Bd^2)^{1/2}$ is the atomic value of the order parameter (see Ref. 5). Taking into account the temperature dependence of the correlation length (8) and of the average value of the order parameter (9), one is readily convinced that $\overline{\eta}_{(\mathrm{II})}$ is small compared to $\overline{\eta}_{(I)} - \eta_\infty$, where η_∞ is the average value of the order parameter in the defectless crystal, i.e., the solution of (9) with $N = 0$ is valid over the entire temperature range if

$$\eta_0/\eta_{\mathrm{at}}\ll 1. \tag{12}$$

Calculations show that condition (23) also ensures that the next nonlinear correction of the average value $\overline{\eta}$ will be small, as will the nonlinear corrections to the other quantities of interest.

Let us discuss the meaning of condition (12). Far from the phase transition point solution (7) goes over to the additive (in the defects) solution obtained previously in Ref. 5. Condition (12) coincides with the condition for the linear approximation in the case of an isolated defect.[5] Near T_c soluton (7) is not additive in the defects. In this case condition (12) can be interpreted as the requirement that fluctuations of η due to the randomness of the defect distribution be small compared to the characteristic value of the order parameter:

$$\langle(\eta-\overline{\eta})^2\rangle\ll\varphi''/B. \tag{13}$$

Let us turn not to defects of the T type ($h_d = 0, g_d = 0$). In this case the linearized equation (5) is of the form

$$D\Delta(\eta-\overline{\eta})=(\varphi''+A_d(\mathbf{r}))(\eta-\overline{\eta})+\varphi'+\overline{\eta}A_d(\mathbf{r}), \tag{14}$$

for which we are not able to write an exact solution. Let us first replace the coefficient $A_d(\mathbf{r})$ on the right-hand side of equation (14) with its average value $\overline{A}_d$. Then we can easily write a solution of the equation. Taking into account that $A_d(\mathbf{r})$ is of the form (3), we have

$$(\eta-\overline{\eta})_{(\mathrm{I})}=-\varphi'/(\varphi''+NV_1A_1)-\xi\overline{\eta}\sum_i\varphi(\mathbf{r}-\mathbf{r}_i), \tag{15}$$

where $\xi = A_1V_1/4\pi Dd$ characterizes the "stiffness" of the defect, $\varphi(\mathbf{r})$ has the same form as in solution (7), and

$$r_c^2=D/(\varphi''+NV_1A_1).$$

Self-consistency condition (6) gives

$$\varphi'(\overline{\eta}_{(\mathrm{I})})+NV_1A_1\overline{\eta}_{(\mathrm{I})}=0. \tag{16}$$

It is seen from this equation that in the given approximation the effect of T type defects reduces to a renormalization of the phase-transition temperature.

We wish to stress that solution (15) was obtained without allowance for fluctuations of the random temperature $A_d(\mathbf{r})-\overline{A}_d$ in the first term on the right-hand side of equation (14). One is able to take these fluctuations into account since equation (14) is of the same form as the equation describing the propagation of waves in media with random inhomogeneities. A diagrammatic method of solving such equations is described, for example, in the book by Rytov, Kravtsov, and Tatarskiĭ.[12] Using this method, we have carried out a partial summation of the diagram series for the averages of powers of the function $\eta(\mathbf{r})$ and for the correlation functions. We shall not give the corresponding calculations here. It turns out that the averages and correlation functions of interest to us coincide with those obtained with the simple solution (15), but with A_1 replaced by $A_1/(1+\xi)$. An estimate of the diagrams dropped from the summation yields the following applicability condition for this approximation:

$$[\xi/(1+\xi)]^2 2\pi Nd^2r_c\ll 1. \tag{17}$$

It can be shown that condition (17) is simultaneously the condition that the nonlinear terms in equation (5) are small. Therefore, the corrections obtained by allowing for a wider class of diagrams in the summation would be smaller than the limits of accuracy of the present treatment. Using solution (15), one readily sees that condition (17) specifies that the order-parameter fluctuations $\langle(\eta-\overline{\eta})^2\rangle$ are small compared to the characteristic value $(\varphi''+A_d)/B$ of the order parameter, i.e., condition (17) is analogous in meaning to inequality (13).

Solutions (7) and (15) are easily generalized to the case in which the defect has all three components: S, P, and T. If we neglect $A_d(\mathbf{r})-A_d$ in the first term on the right-hand side of (5), as we did in obtaining solution (15), we find that in the linear approximation

$$(\eta-\overline{\eta})_{(\mathrm{I})}=-\frac{\varphi'}{\varphi''+NV_1A_1}+(\eta_0-\xi\overline{\eta})\times\sum_i\varphi(\mathbf{r}-\mathbf{r}_i)+\eta_{0P}d\sum_i\psi(\mathbf{r}-\mathbf{r}_i), \tag{18}$$

where

$$\eta_{0P}=\frac{g_1V_1}{4\pi d^2D},\quad \psi(\mathbf{r})=\frac{\partial\varphi}{\partial r}\cos\theta,$$

and θ is the angle between vector **r** and the x axis. As we have already mentioned, solution (15) gives the right answers for the moments and correlation functions, with correct allowance for the fluctuations $A_d - \bar{A}_d$, if we make the replacement

$$A_1 \to A_1^* = A_1/(1+\xi).$$

Analysis shows that h_1 and g_1 undergo the same renormalization:

$$h_1 \to h_1^* = h_1/(1+\xi), \quad g_1 \to g_1^* = g_1/(1+\xi).$$

We shall assume that this renormalization has been carried out in solution (18) and in all the formulas which follow. The renormalized quantities η_0, η_{0P}, and ξ will also be denoted by an asterisk (*).

The correlation length in (18) is given by the expression

$$r_c^2 = D/(\varphi'' + NV_1A_1^*), \tag{19}$$

and the equation for the average order parameter incorporates both the shift in T_c and the effective field:

$$\varphi'(\bar{\eta}_{(I)}) + NV_1A_1^*\bar{\eta}_{(I)} = NV_1h_1^*. \tag{20}$$

Analysis shows that when the P component is taken into account in the form shown in solution (18), there is an auxiliary condition

$$\frac{4\pi}{3}\left(\frac{\eta_{0P}^*}{\eta_{\text{at}}}\right)^2 Ndr_c^2 \ll 1, \tag{21}$$

which means that the fluctuations of η due to the P component must be small and also requires $\eta_{0P}/\eta_{\text{at}} \ll 1$. We note that for polarized defects even with a comparatively small S component conditions (17) and (21) are satisfied over the entire range of temperatures. This becomes obvious when one estimates the left-hand sides of inequalities (17) and (21) for the temperature at which the correlation length is maximum. In fact, according to (19) and (20)

$$\begin{gathered}
A_{max} = -NV_1A_1^* + 3[\pi B^{1/2}NdD\eta_0^*]^{2/3},\\
r_{c\,max}^2 = {}^1/_6\, D[\pi B^{1/2}NdD\eta_0^*]^{-2/3},\\
\bar{\eta}_{(I)max}^2 = B^{-1}[\pi B^{1/2}NdD\eta_0^*]^{2/3}.
\end{gathered} \tag{22}$$

Thus, for $r_c = r_{c\,\max}$ conditions (17) and (21) reduce to

$$\frac{\eta_0^*}{\eta_{\text{aT}}} \gg \xi^6(Nd^3)^2, \quad \frac{\eta_0^*}{\eta_{\text{at}}} \gg \left(\frac{\eta_{0P}^*}{\eta_{\text{at}}}\right)^3 (Nd^3)^{1/2}.$$

For $N = 10^{18}$–$10^{19}\,\text{cm}^{-3}$, $d = 10^{-7}$–$10^{-8}\,\text{cm}^{-3}$, $\eta_{0P}/\eta_{\text{at}} = 0.1$ and $\xi = 1$, these conditions yield $\eta_0/\eta_{\text{at}} \gg 10^{-4}$–$10^{-12}$ and $\eta_0/\eta_{\text{at}} \gg 10^{-3}$–$10^{-5}$, respectively.

2. THERMODYNAMIC QUANTITIES

Using solution (18) and discarding the nonlinear terms in expression (4), we find the free energy of a crystal contianing defects:

$$\begin{aligned}
\frac{\Phi}{V} = {} & \varphi(\bar{\eta}_{(I)}) + \frac{NV_1A_1^*}{2}\bar{\eta}_{(I)}^2 - NV_1h_1^*\bar{\eta}_{(I)}\\
& + 2\pi NDd(\eta_0^* - \xi^*\bar{\eta}_{(I)})^2\frac{d}{r_c} + \frac{4\pi}{3}NDd\eta_{0P}^{*2}\left(\frac{d^2}{2r_c^2} - \frac{d^3}{3r_c^3}\right).
\end{aligned} \tag{23}$$

We note that since $\bar{\eta}_{(I)}$ and r_c in this expression are determined by equations (20) and (19), respectively, the free energy (23) contains terms of any order in the defect concentration. To go over the expressions of first order in the concentration [see formulas (1.1), (1.11) and (5.3) of Ref. 5] it is sufficient to replace $\bar{\eta}_{(I)}$ by η_∞ [$\varphi'(\eta_\infty) = 0$] and r_c by $[D/\varphi''(\eta_\infty)]^{1/2}$. The second term in the expression for the energy (23) makes allowance for the shift of the phase transition temperature, the third takes into account the presence of the effective field, and the last two terms are due to randomness effects, i.e., to fluctuations of the defect concentration.

By differentiating the free energy (23) with respect to h and τ, one can obtain expressions for the thermodyanamic quantities. In view of the length of these expressions, we shall not write them out completely here. Let us examine the two limiting cases in which the randomness effects are due predominatly to: 1) the S component of the defects; 2) to the T and P components. In the first case we have

$$\bar{\eta} = \bar{\eta}_{(I)} - 6\pi\left(\frac{\eta_0^*}{\eta_{\text{at}}}\right)^2 Nr_c^3\bar{\eta}_{(I)}, \tag{24a}$$

$$\chi = \chi_{(I)} - 6\pi\left(\frac{\eta_0^*}{\eta_{\text{at}}}\right)^2 Nr_c^3\left(1 - \frac{3}{2}t\right)\chi_{(I)}, \tag{24b}$$

$$C = C_{(I)} + \pi\left(\frac{\eta_0^*}{\eta_{\text{at}}}\right)^2 Nr_c^3(1 - 8t + 3t^2)\Delta C. \tag{24c}$$

Here $\Delta C = A_0^2/2BT_c$ is the specific-heat discontinuity given by Landau theory for an ideal crystal,

$$t = \varphi'''(\bar{\eta}_{(I)})\bar{\eta}_{(I)}[\varphi''(\bar{\eta}_{(I)}) + NV_1A_1^*]^{-1},$$

and

$$C_{(I)} = {}^1/_3 t\Delta C, \quad \chi_{(I)} = [\varphi''(\bar{\eta}_{(I)})]^{-1}$$

and $\bar{\eta}_{(I)}$ are respectively the anomalous part of the specific heat and susceptibility and the average value of the order parameter with the shift in the transition temperature and the effective field taken into account [see (20)].

In the second case

$$\bar{\eta} = \bar{\eta}_{(I)} - 4\pi Nd^2r_c\xi^{*2}\left(1 + \frac{1}{4}t\right)\bar{\eta}_{(I)} - 4\pi\left(\frac{\eta_{0P}^*}{\eta_{\text{at}}}\right)^2 Ndr_c^2\bar{\eta}_{(I)}, \tag{25a}$$

$$\begin{aligned}
\chi = {} & \chi_{(I)} - 4\pi Nd^2r_c\xi^{*2}\left(1 + \frac{1}{4}t - \frac{3}{8}t^2\right)\chi_{(I)}\\
& - 4\pi\left(\frac{\eta_{0P}^*}{\eta_{\text{at}}}\right)^2 Ndr_c^2(1 - t)\chi_{(I)},
\end{aligned} \tag{25b}$$

$$\begin{aligned}
C = {} & C_{(I)} - \pi\left(\frac{\bar{\eta}_{(I)}\xi^*}{\eta_{\text{at}}}\right)^2 Nr_c^3(9 + 8t - 3t^2)\Delta C + 2\pi\left(\frac{\eta_{0P}^*}{\eta_{\text{at}}}\right)^2 Nd^2r_c\\
& \times(1 + 4t - t^2)\Delta C.
\end{aligned} \tag{25c}$$

Comparing expression (24) and (25) with inequalities (11), (17), and (21), we see that the contribution of concentration fluctuations to the thermodynamic quantities [i.e., $\bar{\eta} - \bar{\eta}_{(I)}$, $\chi - \chi_{(I)}$, and $C - C_{(I)}$] are proportional to the small parameters of our approximation. The difference $\bar{\eta} - \bar{\eta}_{(I)}$ in (24a) was in fact already taken into account in formula (11). The difference between these formulas is that (11) contains a second term which could also be obtained in (24a) by simply considering the nonlinear terms in the free energy (4). This term is of no interest, however, since it plays an appreciable role only far from the phase transition. On the whole, fluctuations of the concentration of polarized defects give an appreciable contribution either for rather strong defects, when $\eta_0 \sim \eta_{at}$ (in this case fluctuations of the S component are important) or at a high concentration of defects with a small value of η_0 [see inequalities (17) and (21)].

Let us now discuss the conditions which correspond to the two limiting cases under consideration. For this purpose let us compare the fluctuation contributions corresponding to the different defect components at $r_c = r_{c\ \max}$ (22), when these contributions are a maximum. We easily see as a result that the fluctuations corresponding to the S component [formulas (24)] play the predominant role at low defect concentrations. The corresponding conditions are

$$\xi^3 N d^3 < (\eta_0/\eta_{at})^2, \tag{26}$$

$$N d^3 < (\eta_0/\eta_{0P})^6 (\eta_{at}/\eta_0) \tag{27}$$

for the contributions to the susceptibility and to the average value of the order parameter. For the contribution to the specific heat the second inequality is replaced by

$$N d^3 < (\eta_0/\eta_{0P})^3 (\eta_{at}/\eta_0) \tag{28}$$

with increasing defect concentration the fluctuations associated with the T and P components [formulas (25)] begin to play the predominant role.

The "non-field" contribution to $C - C_{(I)}$ in formula (24c) is shown in Fig. 1. We note that the temperature dependence of the first fluctuational correction to the specific heat (the correction due to the thermal fluctuations of η) has a similar form in the presence of a field conjugate to the order parameter. The ratio of these corrections at $r_c = r_{c\ \max}$ is

$$\frac{C - C_{(I)}}{C_f} = 2\pi \frac{T_{at}}{T_c} \left(\frac{d}{r_{c\ max}} \right)^2 = 12\pi^{5/2} \frac{T_{at}}{T_c} (N d^3)^{2/3} \left(\frac{\eta_0^*}{\eta_{at}} \right)^{2/3}, \tag{29}$$

where $T_{at} = D^2/Bdk_B$ is of the order of the atomic temperature for transitions of the displacive type; $T_{at} \sim T_c$ for transitions of the order-disorder type. For $T_c = 100$ K, $N = 10^{18}$ cm^{-3}, $d = 10^{-7}$ cm, and $\eta_0^*/\eta_{at} = 0.1$ we obtain $(C - C_{(I)})/C_f \sim 10^2$ for a displacive phase transition and $(C - C_{(I)})/C_f \sim 1$ for an order-disorder transition. We stress that on the whole the specific heat (24c) depends montonically on the temperature.

Formulas (25) are, of course, also applicable in the absence of an S component of the defects. In this case they reduce, to within a renormalization of T_c, to the formulas which can be obtained in the approximation of isolated defects. We stress that the region around T_c in which formulas (25) do not apply is substantially smaller than in the case of the isolated-defect approximation.

3. CORRELATION FUNCTIONS

Fluctuations of the polarized-defect concentration can cause significant static scattering of neutrons, light, and x rays. We note that in contrast to the thermal-fluctuation correlation functions, the correlation functions governing the scattering by static fluctuations of the defect concentration are not directly related to the corresponding generalized susceptibilities.

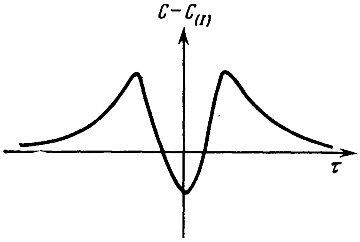

FIG. 1. Temperature dependence of the contribution to the specific heat from fluctuations in the concentration of S defects.

Let us first consider the correlation function

$$\langle (\eta(0) - \bar{\eta}) (\eta(\mathbf{r}) - \bar{\eta}) \rangle,$$

which determines the intensity of the elastic neutron scattering and diffuse x-ray scattering. For its Fourier components with $k \ll 1/d$, i.e., sufficiently close to the Bragg reflection, we obtain with the aid of (18) the equation

$$\langle (\eta(0) - \bar{\eta}) (\eta(\mathbf{r}) - \bar{\eta}) \rangle_k = 16\pi^2 N d^2 r_c^4 (\eta_0^* - \xi^* \bar{\eta}_{(I)})^2 \times \frac{1}{(1 + k^2 r_c^2)^2} + 16\pi^2 N d^4 r_c^2 \eta_{0P}^{*2} \frac{(k_x r_c)^2}{(1 + k^2 r_c^2)^2}. \tag{30}$$

The temperature dependence$^{1)}$ of this correlation function in the case $kr_c \ll 1$ is given for different defect concentrations in Fig. 2. At low values of N [in the same of inequality (26)] it has a peak due to fluctuations of the S component. The size of this peak decreases with increasing N, and, simultaneously, a second peak, due to fluctuations of the T component, appears. We note that if the S component of the defects is nonzero, then the peak in the temperature dependence coincides with the temperature of the maximum of r_c only in the limiting case of small N.

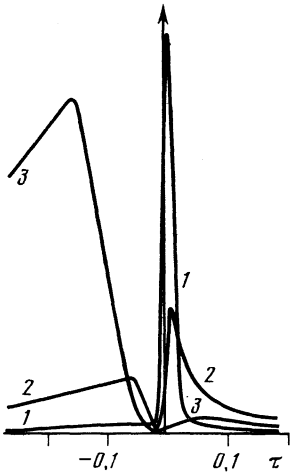

FIG. 2. Temperature dependence of the elastic neutron-scattering (diffuse x-ray-scattering) intensity for various concentrations of polarized defects: 1) $N = 10^{17}$ cm^{-3}, 2) 10^{18}, 3) 10^{19}. The values of the remaining parameters are: $d = 10^{-7}$ cm, $D/A_0 = 10^{-14}$ cm^2, $\eta_0/\eta_{at} = 0.1$, $\xi = 1$.

Let us turn now to the correlation function of the square of the order parameter, which determines the elastic-scattering intensity for light. Here, as usual, one can set $k \approx 0$. This correlation is of the form

$$\langle(\eta^2(0)-\bar{\eta}^2)(\eta^2(\mathbf{r})-\bar{\eta}^2)\rangle=\langle(\eta(0)-\bar{\eta})^2(\eta(\mathbf{r})-\bar{\eta})^2\rangle$$
$$+4\bar{\eta}\langle(\eta(0)-\bar{\eta})(\eta(\mathbf{r})-\bar{\eta})^2\rangle+4\bar{\eta}^2\langle(\eta(0)-\bar{\eta})(\eta(\mathbf{r})-\bar{\eta})\rangle; \quad (31)$$

The last correlator in this expression is given by formula (30), and for the other two correlators we find, using solution (18), at $k=0$

$$\langle(\eta(0)-\bar{\eta})(\eta(\mathbf{r})-\bar{\eta})^2\rangle=8\pi^2Nd^3r_c^3(\eta_0^*-\xi^*\bar{\eta}_{(\mathrm{I})})^3+\frac{64\pi^2}{3}$$
$$\times Nd^4r_c^5(\eta_0^*-\xi^*\bar{\eta}_{(\mathrm{I})})\eta_{0P}^{*2}. \quad (32)$$

$$\langle(\eta(0)-\bar{\eta})^2(\eta(\mathbf{r})-\bar{\eta})^2\rangle=4\pi^2Nd^4r_c^2(\eta_0^*-\xi^*\bar{\eta}_{\mathrm{I}})^4$$
$$+\frac{128\pi^2}{3}Nd^5r_c(\eta_0^*/\xi^*\bar{\eta}_{(\mathrm{I})})^2\eta_{0P}^{*2}+8\pi^3N^2d^4r_c^5(\eta_0^*-\xi^*\bar{\eta}_{(\mathrm{I})})^4$$
$$+\frac{2}{3}\pi^3N^2d^6r_c^3(\eta_0^*-\xi^*\bar{\eta}_{(\mathrm{I})})^2\eta_{0P}^{*2}+\frac{3}{10}\pi^3N^2d^8r_c\eta_{0P}^{*4}. \quad (33)$$

In the last expression we have not written out the terms which do not depend on the temperature.

At a low defect concentration [conditions (26) and (27)] the main contribution to the correlation function is that of the S component of the defects. In this case we find for $k=0$, using (31), (32), and (33),

$$\langle(\eta^2(0)-\bar{\eta}^2)(\eta^2(\mathbf{r})-\bar{\eta}^2)\rangle=N\eta_0^{*2}\{2\pi d^2r_c\eta_0^*+8\pi dr_c^2\bar{\eta}_{(\mathrm{I})}\}^2$$
$$+N^2\eta_0^{*4}(2\pi)^3d^4r_c^5. \quad (34)$$

The contribution of the S component to the correlation function, as given by formula (34), has a temperature peak whose size increases with increasing N.

For sufficiently large N [in the sence of inequality (26)] the interference of the contribution of the S and T components causes the growth of the peak with increasing N to give way to a fall-off. This stage of the evolution of the temperature dependence is illustrated in Fig. 3a. Upon further growth in N all of the defect components can contribute to the scattering, and the temperature dependence can have a rather complex form. An example of a possible temperature dependence is given in Fig. 3b.

At a high defect concentration [i.e., when the inequalities opposite to (26) and (28) hold], the main contribution to (31) is from the T and P components of the defects. In this case we have for $k=0$

$$\langle(\bar{\eta}^2(0)-\bar{\eta}^2)(\eta^2(\mathbf{r})-\bar{\eta}^2)\rangle=64\pi^2Nd^2r_c^4\xi^{*2}\bar{\eta}_{(\mathrm{I})}^4$$
$$-\frac{256\pi^2}{3}Nd^4r_c^2\xi^*\bar{\eta}_{(\mathrm{I})}^2\eta_{0P}^{*2}+(2\pi)^3N^2d^4r_c^5\xi^{*4}\bar{\eta}_{(\mathrm{I})}^4$$
$$+\frac{2\pi^3}{3}N^2d^6r_c^3\xi^{*2}\bar{\eta}_{(\mathrm{I})}^2\eta_{0P}^{*2}+\frac{3\pi^3}{10}N^2d^8r_c\eta_{0P}^{*4}. \quad (35)$$

Let us discuss this last expression first for the case when the defect has no component. The first two terms nonvanishing to first order in N evidently give a jump of the scattering intensity at the phase transition; when the P component is taken into account, the sign of this jump can be positive or negative. As the temperature approaches T_c and the quadratic terms in (35) become importnat, the light-scattering intensity grows as $\tau^{-1/2}$. In the asymmetric phase this growth is due to both the T and P components of the defects, while in the symmetric phase it is due only to the P component. The presence of the S component causes a smearing of these curves. Figure 3c gives the temperatue dependence of the scattering intensity at high defect concentrations [see (26)] for the case in which the defects have only T and S components.

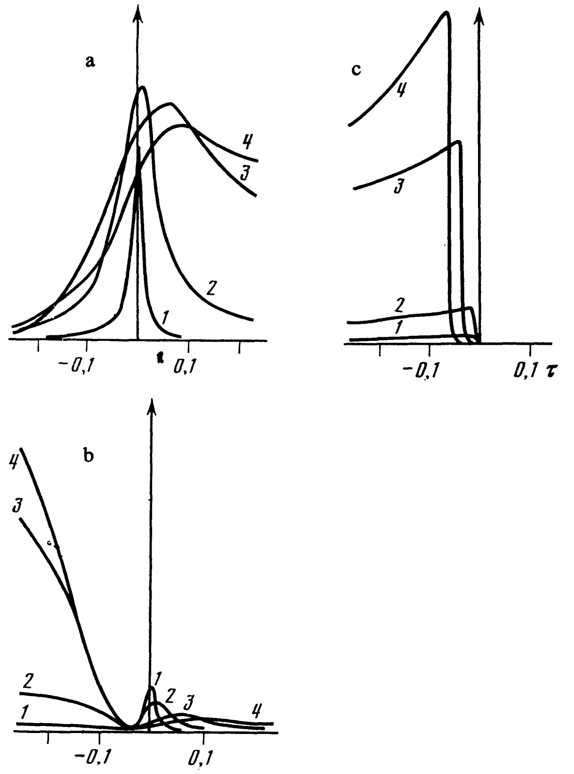

FIG. 3. Temperature dependence of the elastic light-scattering intensity for defects with different "strengths": a) $\eta_0/\eta_{\mathrm{at}}=0.6$, b) 0.2, c) 0.001. Curve 1) $N=10^{17}\,\mathrm{cm}^{-3}$, 2) 10^{18}, 3) $6.4\cdot10^{18}$, 4) 10^{19}. The values of the remaining parameters: $d=10^{-7}$ cm, $D/A_0=10^{-14}\,\mathrm{cm}^2$, $\eta_{0P}/\eta_{\mathrm{at}}=0.001$.

Let us estimate the defect contribution to the light-scattering as compared to the noncritical molecular scattering I_M. By way of estimate we can assume that I_M is governed by density fluctuations. Then at $k=0$ we find

$$\frac{I_d}{I_{\mathrm{M}}}=a^2\langle(\eta^2(0)-\bar{\eta}^2)(\eta^2(\mathbf{r})-\bar{\eta}^2)\rangle\left[\frac{k_BT_c}{\lambda}\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)^2\right]^{-1}, \quad (36)$$

where a is the coefficient in the relation $\varepsilon=\varepsilon_0+a\eta^2$. Taking $\rho\partial\varepsilon/\partial\rho=1$, $T_c=100$ K, $\lambda=10^{11}$–10^{12} erg/cm^3, and $a\eta_{\mathrm{at}}{}^2=10^{-1}$–$10^{-3}$, we obtain $I_d/I_M\sim10^{-2}$–10^3 for the intensity corresponding to the peak in Fig. 3a. Thus we see that, in principle, defects can make a large contribution to the light-scattering even at relatively low concentrations. As we have see, a growth of the defect concentration does not by any means always lead to an increase in the light-scattering intensity.

4. KINETIC COEFFICIENTS

Let us consider the polarized-defect contributions to the anomalies in the kinetic coefficients, using as examples the acoustic damping and the soft-mode damping constant. This topic is of interest inasmuch as defects can give a substantial contribution to these quantities, as was shown in Ref. 5 in the approximation of independent defects.

To allow for the connection between the acoustic strains and the order parameter, we must add to the free energy density (1) the following terms[13,5,6]

$$\frac{r}{2}\eta^2 v+\frac{\lambda}{2}v^2,$$

where $v = u_{ll} = -\Delta\rho/\rho$, and λ is the elastic modulas. For the sake of simplicity let us neglect the crystalline anisotropy and the nonzero value of the shear modulus. The linearized equations of the motion for the strain v in the sound wave and the accompanying oscillations in the order parameter η are of the form

$$\rho\ddot{v}=\lambda\Delta v+r\Delta(\eta_e\eta),\quad -\gamma\dot{\eta}=[\varphi''(\eta_e)+A_d]\eta-D\Delta\eta+r\eta_e v, \tag{37}$$

where $\eta_e = \eta_e(r)$ denotes the equilibrium value of the order parameter in the crystal in the presence of the defects. In solving equations (37), we transform to the Fourier components $v(\mathbf{k},\omega)$ and $\eta(\mathbf{k},\omega)$ and retain terms of lowest order in the fluctuations $\tilde{\eta}_e = \eta_e(r) - \bar{\eta}_e$, as was done in Refs. 5 and 6.

As in the specific-heat calculation, it is fluctuations of the S component tht are important at low defect concentrations [see (26) and (28)]. In this case we find for the renormalized modulus $\tilde{\lambda}(\omega)$

$$\begin{aligned}\tilde{\lambda}(\omega)=\lambda-r^2\bar{\eta}_e^2[\varphi''(\bar{\eta}_e)+NV_1A_1^*+{}^1/_2\,\varphi^{\mathrm{IV}}\langle\eta_e^2\rangle-i\omega\gamma]^{-1}\\ -r^2\left[1-\frac{\varphi''(\bar{\eta}_e)\bar{\eta}_e}{\varphi''(\bar{\eta}_e)+NV_1A_1^*-i\omega\gamma}\right]^2\\ \times\sum_{\mathbf{k}}\frac{\langle|\tilde{\eta}_{e\mathbf{k}}|^2\rangle}{i\omega\gamma+Dk^2+\varphi''(\bar{\eta}_e)+NV_1A_1^*}.\end{aligned} \tag{38}$$

Recognizing that

$$\langle\tilde{\eta}_e^2\rangle=\eta_0^{*2}2\pi Nd^2r_c,$$

and that $\bar{\eta}_e$ is given by expression (24a), we have for the case of low frequencies $\omega\ll\omega_R$, where $\omega_R = (\varphi'' + NV_1A_1^*)/\gamma$ is the inverse relaxation time of the order parameter,

$$\operatorname{Re}\tilde{\lambda}-\lambda=-\frac{t}{3}\Delta\lambda-\pi Nr_c^3\left(\frac{\eta_0^*}{\eta_{\mathrm{at}}}\right)^2\Delta\lambda(1-8t+3t^2), \tag{39}$$

$$\operatorname{Im}\tilde{\lambda}=\frac{t}{3}\frac{\omega}{\omega_R}\Delta\lambda+\frac{1}{2}\pi Nr_c^3\left(\frac{\eta_0^*}{\eta_{\mathrm{at}}}\right)^2\frac{\omega}{\omega_R}\Delta\lambda(1-22t+13t^2). \tag{40}$$

Here $\Delta\lambda = r^2/2B$ is the discontinuity in the modulus λ at the phase transition, and the quantity t is the same as in (24). We note that for $\lambda\gg|\operatorname{Re}\tilde{\lambda}-\lambda|$ the anomalous part of the inverse modulus is related to the specific heat (24c) by the Pippard relation,[14] as, of course, it must be. For the damping of sound in the high-frequency limit $\omega\ll\omega_R$ we have

$$\operatorname{Im}\tilde{\lambda}=\frac{t}{3}\frac{\omega_R}{\omega}\Delta\lambda+4\pi Nr_c^3\left(\frac{\eta_0^*}{\eta_{\mathrm{at}}}\right)^2\frac{\omega_R}{\omega}\Delta\lambda(1-t). \tag{41}$$

The first terms in (39), (40), and (41) represent the contribution due to the Landau-Khalatnikov relaxation mechanism with allowance for the effective field and shift of the transition temperature, while the second terms are due to fluctuations of the defect concentration. As can be seen by comparing formulas (39), (40), and (41) with expression (11), the contributions to $\tilde{\lambda}(\omega)$ due to concentrations fluctuations are proportional to the small parameter of our approximation. This also pertains to the contributions (written out below) of the T and P components. They can be appreciable under the same conditions that apply for the contributions of concentration fluctuations to the thermodynamic quantities (Sect. 2). The corrections to the low-frequency values $\operatorname{Re}\tilde{\lambda}$ and $\operatorname{Im}\tilde{\lambda}$ have the same qualitiative form as the correction to the specific heat (24c) (See Fig. 1). The correction to the Landau-Khalatnikov damping in the high-frequency regions is negative in the asymmetric phases and positive in the symmetric phase, passing through zero at $r_c = r_{c\,\max}$. On the whole, however, the quantity $\operatorname{Im}\tilde{\lambda}$ in the high-frequency region, as given by (41), depends monotonically on the temperature.[2)]

For the contribution of fluctuations of the P and T components to the low-frequency value of $\tilde{\lambda}(\omega)$ we find from (38) and (18)

$$\operatorname{Re}\tilde{\lambda}\approx\lambda-\frac{t}{3}\Delta\lambda+9\pi Nr_c^3\left(\frac{\bar{\eta}_{(\mathrm{I})}\xi^*}{\eta_{\mathrm{aT}}}\right)^2\Delta\lambda-2\pi Nd^2r_c\left(\frac{\eta_{0P}^*}{\eta_{\mathrm{aT}}}\right)^2\Delta\lambda, \tag{42}$$

$$\begin{aligned}\operatorname{Im}\tilde{\lambda}\approx\frac{t}{3}\frac{\omega}{\omega_R}\Delta\lambda+6\pi Nr_c^3\left(\frac{\bar{\eta}_{(\mathrm{I})}\xi^*}{\eta_{\mathrm{aT}}}\right)^2\frac{\omega}{\omega_R}\Delta\lambda\\ +2\pi Nd^2r_c\left(\frac{\eta_{0P}^*}{\eta_{\mathrm{aT}}}\right)^2\frac{\omega}{\omega_R}\Delta\lambda.\end{aligned} \tag{43}$$

We note that if the defects lack an S component the contributions of the T and P components to $\operatorname{Re}\tilde{\lambda}$ and $\operatorname{Im}\tilde{\lambda}$ have the same temperature dependence as do the contributions of thermal fluctuations of the order parameter: $\tau^{-1/2}$ and $\tau^{-3/2}$, respectively. Estimates show that these contributions are completely comparable in order of magnitude as well. There is no substantial contribution to the high-frequency value of the acoustic damping from the T and P components.

Let us now discuss the contribution of polarized defects to the soft-mode damping constant. Several mechanisms are known[5,7,15] to cause anomalous behavior of this constant near the phase transition point in crystals containing defects. The temperature dependence corresponding to the different contributions is qualitatively the same. Let us consider as an example the damping mechanism due to the coupling of the soft mode to the heat-conduction mode. The coupling of these modes is induced by the defects. The equations of motion for the order parameter η with allowance for fluctuations of the temeprature T are of the form[5]

$$\begin{aligned}m\ddot{\eta}+\gamma\dot{\eta}+[\varphi''(\eta_e)+A_d]\eta-D\Delta\eta+\frac{A_0}{T_c}\eta_e T=0,\\ C_\eta\dot{T}-A_0\eta_e\eta=\varkappa\Delta T,\end{aligned} \tag{44}$$

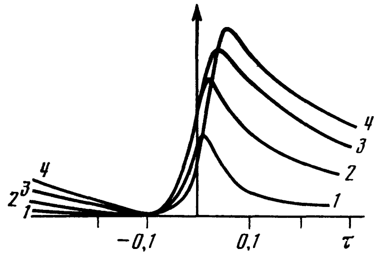

FIG. 4. Temperature dependence of the polarized-defect contribution to the soft-mode damping constant: 1) $N = 10^{17}\ \mathrm{cm}^{-3}$, 2) $4\cdot10^{17}$, 3) $7\cdot10^{17}$, 4) 10^{18}. The values of the remaining parameters: $\eta_0/\eta_{\mathrm{at}} = 0.3$, $\eta_{0P}/\eta_{\mathrm{at}} = 0.05$, $d = 10^{-7}$ cm, $D/A_0 = 10^{-14}\ \mathrm{cm}^2$, $\xi = 1$.

where C_η is the specific heat far from the transition point and $\varkappa$ is the thermal conductivity.

A calculation in the same approximation as for $\tilde{\lambda}(\omega)$ yields the following expression for the defect contribution to the soft-mode damping constant:

$$\gamma_d(\omega) = \frac{A_0^2}{T_c} \sum_{\mathbf{k}} \frac{\langle |\tilde{\eta}_{e\mathbf{k}}|^2 \rangle}{-i\omega C_\eta + \varkappa k^2}. \tag{45}$$

In sufficiently close vicinity of the phase transition, the region of the greatest experimental interest, the soft-mode frequency

$$\omega_0^2 = [\varphi''(\eta_e) + NV_1A_1^*]/m$$

is substantially greater than the characteristic frequency $\Omega_r = \varkappa/C_\eta r_c^2$ of the heat-conduction mode. In this frequency region ($\omega_0 \gg \omega_r$) we have

$$\gamma_d(\omega\sim\omega_0) = \frac{\pi\sqrt{2}A_0^2(\eta_0^* - \xi^*\bar{\eta}_{(\mathrm{I})})^2 d^2}{\varkappa T_c} \times Nr_c^3\left(\frac{\omega_0}{\Omega_r}\right)^{-3/2} + \frac{4\pi}{3}\frac{\sqrt{2}A_0^2\eta_{0P}^{*2}d^2}{\varkappa T_c} \times Nd^2 r_c\left(\frac{\omega_0}{\Omega_r}\right)^{-1/2}. \tag{46}$$

This formula differs from the expression obtained in the isolated-defect approximation only in that the quantities r_c and $\eta_{(\mathrm{I})}$ appearing in it are re-evaluated with allowance for the shift in T_c and for the effective field. At low defect contributions [conditions (26) and (27)] the leading contribution to (46) is due to the S components of the defects, and the temperature dependence has a peak near T_c (Fig. 4). At high defect concentrations the monotonically decaying contribution of the T component is dominant. Here there may also be an noticeable peak due to the P component.

CONCLUSION

Let us at last discuss the question of the relation of our theory with experiment. Of greatest interest in this regard are the effects due to fluctuations in the defect concentration, i.e., to the randomness of the defect distribution. These effects are manifested most clearly in the scattering of neutrons, light, and x rays. The essential point is that the very nature of the temperature dependence and concentration dependence of the scattering intensity depend strongly on the parameters of the defects. This raises the hope of determining the defect parameters from scattering experiments. It would then be of interest to use these parameters in calculating theoretical curves for the thermodynamic quantities and kinetic coefficients and to compare then with the experimental observations. In carrying out such a program one would encounter a number of serious difficulties in both the experimental and theoretical procedures. The first of these difficulties concerns primarily the preparation of samples with uniformly distributed impurities of a given kind and a known, sufficiently small ($Nd^3 \ll 1$) concentration. In the theory one would still have to take into account the role of elastic distortions around the defect (this is particularly important near a tricritical point), the nonuniformity of the defect distribution in the sample, the multicomponent nature of the order parameter, and so forth. It should be noted that in doing this, one need not consider separately the problem of an incompletely polarized defect system: The results given above can be applied in this case as well; one need only introduce a depolarization factor in the expression for the effective field in formula (20).

We wish to thank A. I. Morozov for helpful discussions of this study in various stages of its progres, N. A. Makarov for doing the computer calculations, and A. A. Sobyanin for a discussion of the results.

1) The temperature curves given in Figs. 2–4 were obtained by computer calculations of the corresponding formulas. The calculations were done by N. A. Makarov.

2) The text of Ref. 6 leaves the impression that owing to the presence of defects, the damping coefficient for high-frequency sound will have a maximum at the phase-transition point. For the case of random defects this conclusion would go beyond the accuracy of the approximation on account of the contribution from the Landau-Khalatnikov damping mechanism. As to the case of polarized defects, the temperature dependence of the absorption coefficient, as we mentioned earlier, is monotonic.

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