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The influence of defects on first-order structural phase transitions

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Abstract. When studying the influence of quenched defects on first-order structural phase transitions, one has to take into account the elastic deformations which are induced by heterogeneous nucleation of the new phase. In the present paper the influence of both 'random local transition temperature' and 'random local field' defects are considered within a first-order perturbation Landau theory. Both the order parameter (o.p.) fluctuations and the defects are treated perturbatively, which is consistent for structural phase transitions close to a tricritical point and for low enough defect concentrations. In such systems the o.p. fluctuations tend to induce the first-order character of the transition, whereas both types of defect play the opposite role and tend to decrease the jump of the o.p. at the transition. Estimates of the concentration of defects which can lead to complete smearing of the transition are given.

1. Introduction

The problem of the influence of defects on first-order phase transitions is not only of academic interest: defects can lead to a smearing of the phase transitions which has strong practical repercussions. The influence of quenched defects on first-order phase transitions has been discussed by Imry and Wortis [1] and the results of their paper have been repeatedly used to interpret experimental data on some structural phase transitions (see, e.g., [2]). However, for structural transitions in solids the applicability of the theory [1] is highly questionable. Indeed, what was discussed in [1] is interplay between the volume and the interface energy for a nucleus of the new phase, but the elastic energy associated with the formation of the nucleus has been completely neglected. Such an approach is quite reasonable for liquids or, maybe, for some magnetic transitions where the striction effects are quite small, but it is not true for structural transitions. The importance of the elastic effects at nucleation in solids has been known for a long time [3] and has been repeatedly emphasized (see, e.g., [4, 5]). The main point is that the elastic energy associated with the nucleation is proportional to the volume of the nucleating centre. That leads to a strong suppression of the nucleation which has to be taken into account.

In this paper we shall take into account the above-mentioned elastic deformations. We restrict ourselves to the case of small defect concentrations when defect contributions are proportional to the concentration. We shall consider an elastically isotropic (but solid) medium and one-component order parameter (o.p.) which has symmetry properties different

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from those of any component of the strain tensor. The latter means that the only possible lower-order coupling of the o.p. and the strain is of the striction type (see (3)), which takes place for any o.p.

For systems without defects it has been shown [6] that the fact that the o.p. fluctuations occur in a solid, i.e. in a medium with a non-zero shear modulus μ, changes essentially the character of the phase transition converting, in most cases, a second-order phase transition into a first-order one. The effect is especially drastic for a tricritical point: the phase transition becomes a first-order one for any number of components of the o.p., the fluctuation-induced first-order transition occurs in a region where the criterion of smallness of the critical fluctuations [7] is still fulfilled [8] and, for structural phase transitions, the discontinuity of the order parameter at the transition is not very small compared with its value at $T = 0$ [9]. We shall consider such phase transitions in this paper and study how relatively small concentrations of the defects change the nature of the transitions. It follows from the above remarks that it is reasonable and consistent to study the effects of both the o.p. fluctuations and the defects within a perturbative approach using the Landau (mean field) thermodynamic potential as a zero approximation. For second-order transitions such an approach to study the effects of defects has been used already (see [10]–[12] and references therein).

2. Random local transition temperature defects

In this section we shall study defects that do not break the symmetry of the symmetrical phase, usually called ‘random local transition temperature’ (RLT) ones. Their presence can be taken into account by assuming that the coefficients of the continuous-medium effective Hamiltonian are space dependent (see, e.g., [10]–[12]). Below we shall restrict ourselves to the space dependence of only one coefficient (that for which it is most important) and assume as well that the defects are weak (see the condition below). This allows us not to take into account the defect form factor but only its ‘strength’ [12]. We shall use the perturbation theory to obtain the terms proportional to the defect concentration. Our consideration is quite similar to that presented in [11] and [12] for second-order transitions. It has been pointed out in [11] (p 87) that in the symmetrical phase the RLT defect induced and the fluctuation induced anomalies are of the opposite sign (within the perturbation theory). For first-order transitions it is reasonable to expect that the effect will be qualitatively the same: a reduction of the effects of fluctuations due to the defects, i.e. a smearing of the first-order transition. We shall show this explicitly.

The effective continuous-medium Hamiltonian that can be used as the starting point for study of both the effects of defects and of the order parameter fluctuations has the form of the Landau thermodynamic potential:

$$\Phi(\eta, u_{ik}) = \int [\varphi_\eta(\eta) + \varphi_u(\eta, u)]\,\mathrm{d}v \tag{1}$$

with

$$\varphi_\eta = \tfrac{1}{2}A\eta^2 + \tfrac{1}{4}B\eta^4 + \tfrac{1}{6}C\eta^6 + \tfrac{1}{2}D(\nabla\eta)^2 + \tfrac{1}{2}\Delta A(\boldsymbol{r})\eta^2 \tag{2}$$

and

$$\varphi_u(\eta, u) = r\eta^2 3u_{jj} + \tfrac{1}{2}Ku_{jj}^2 + \mu(u_{ik} - \tfrac{1}{3}u_{jj}\delta_{ik})^2 \tag{3}$$

where η is the order parameter, u_{ik} is the strain tensor and

$$\Delta A(\boldsymbol{r}) = A_1 N(\boldsymbol{r}) = A_1 \sum_j \delta(\boldsymbol{r} - \boldsymbol{r}_j) \tag{4}$$

describes the contribution of the random temperature defects, $N(\boldsymbol{r})$ is the defect density, $\boldsymbol{r}_j$ is the defect position and A_1 is the defect strength.

The defect contribution can be found neglecting the fluctuations: it has been mentioned above that in the problem in question they are never large. Thus one has to find the minimum of the effective Hamiltonian (1) and to average over the defect distribution, but first it is convenient to minimize over the elastic degrees of freedom making use of the fact that the effective Hamiltonian (1) is quadratic in the strain components. In this minimization one has to discriminate between spatially homogeneous and inhomogeneous strains:

$$u_{ij}(\boldsymbol{r}) = u_{ij}^0 + \frac{1}{2}\sum_{\boldsymbol{k}\neq\boldsymbol{0}}[\mathrm{i}k_j u_i(\boldsymbol{k}) + \mathrm{i}k_i u_j(\boldsymbol{k})]\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}) \tag{5}$$

where u_{ij}^0 is the homogeneous strain and $u_i(\boldsymbol{r})$ is the displacement vector. One has to minimize with respect to u_{ij}^0 and $u_i(\boldsymbol{r})$ separately [6]. As a result one obtains

$$\int \varphi_u(\eta, u)\,\mathrm{d}V = -\frac{r^2}{2K}\left(\sum_{\boldsymbol{k}}\eta_{\boldsymbol{k}}\eta_{-\boldsymbol{k}}\right)^2 - \frac{r^2}{2\lambda}\sum_{\boldsymbol{k}\neq\boldsymbol{0}}\sum_{\boldsymbol{k}_1\boldsymbol{k}_2}\eta_{\boldsymbol{k}_1}\eta_{-\boldsymbol{k}_1-\boldsymbol{k}}\eta_{-\boldsymbol{k}_2}\eta_{\boldsymbol{k}_2+\boldsymbol{k}} \tag{6}$$

where $\lambda = K + 4\mu/3$ and the volume of the system is taken to be equal to unity. Let us emphasize that it is the difference between λ and K that is responsible for the specific features of the problem in solids.

In what follows we shall distinguish two parts in the free energy: Φ_0, which depends on the zero Fourier component of the order parameter (η_0), and Φ_1, which depends on η_0 and $\eta_{\boldsymbol{k}}$. One has

$$\Phi_0 = (\tilde{A}/2)\eta_0^2 + \tfrac{1}{4}\tilde{B}\eta_0^4 + \tfrac{1}{6}C\eta_0^6 \tag{7}$$

where

$$\begin{aligned}\tilde{A} &= A + NA_1\\ \tilde{B} &= B - 2r^2/K.\end{aligned} \tag{8}$$

N is the defect concentration. In Φ_1 one keeps only terms at most quadratic in $\eta_{\boldsymbol{k}}$:

$$\Phi_1 = A_1\eta_0\sum_{\boldsymbol{k}\neq\boldsymbol{0}}N_{\boldsymbol{k}}\eta_{-\boldsymbol{k}} + \frac{1}{2}\sum_{\boldsymbol{k}\neq\boldsymbol{0}}(\tilde{A} + 3B_1\eta_0^2 + 5C\eta_0^4 + Dk^2)\eta_{\boldsymbol{k}}\eta_{-\boldsymbol{k}} \tag{9}$$

where

$$B_1 = B - \tfrac{2}{3}r^2/K - \tfrac{4}{3}r^2/\lambda = \tilde{B} + \tfrac{16}{9}r^2\mu/K_\lambda \equiv \tilde{B} + \Delta. \tag{10}$$

The term NA_1 in (8) describes the shift of the transition temperature due to the defects: within the first-order perturbation theory this shift is naturally proportional to the defect concentration.

It follows from (9) that the equilibrium value of $\eta_{\boldsymbol{k}}$ ($\eta_{\boldsymbol{k}e}$) is:

$$\eta_{\boldsymbol{k}e} = -A_1N_{\boldsymbol{k}}\eta_0/(\tilde{A} + 3B_1\eta_0^2 + 5C\eta_0^4 + Dk^2). \tag{11}$$

The equilibrium value of $\Phi_1(\eta_0)$ is

$$\Phi_1(\eta_0) = -\frac{1}{2}A_1^2\eta_0^2\sum_{\boldsymbol{k}\neq\boldsymbol{0}}\frac{\langle N_{\boldsymbol{k}}N_{-\boldsymbol{k}}\rangle}{\tilde{A} + 3B_1\eta_0^2 + 5C\eta_0^4 + Dk^2} \tag{12}$$

where the angular brackets $\langle\,\rangle$ mean averaging over the defect positions. For randomly distributed defects

$$\langle N_{\boldsymbol{k}}N_{-\boldsymbol{k}}\rangle = N \tag{13}$$

and one finds after integration over $\boldsymbol{k}$

$$\Phi_1(\eta_0) = -\tfrac{1}{2}(A_1^2 N k_{max}/2\pi^2 D)\eta_0^2 + (A_1^2 N/2\pi D^{3/2})\eta_0^2(\tilde{A} + 3B_1\eta_0^2 + 5C\eta_0^4)^{1/2} \tag{14}$$

where k_{max} is the cutoff vector of the problem, which can be estimated to be of the same order of magnitude as the reciprocal lattice vector. It is not important to know it accurately because the first term in the rhs of (14) can be interpreted as a correction to the shift of the transition temperature and can be neglected as long as the 'strength' of the defects is low enough to ensure applicability of our treatment [11, 12].

Now let us consider the o.p. fluctuations. Within the first-order perturbation theory the contributions of the defects and fluctuations are additive and one can use the results of [9] where the fluctuation contribution has been calculated to obtain

$$\begin{aligned}\Phi(\eta_0) = \frac{1}{2}\tilde{A}\eta_0^2 + \frac{1}{4}\tilde{B}\eta_0^4 + \frac{1}{6}C\eta_0^6 + \frac{A_1^2 N}{2\pi D^{3/2}}\eta_0^2(\tilde{A} + 3B_1\eta_0^2 + 5C\eta_0^4)^{1/2}\\ -\frac{T}{12\pi D^{3/2}}(\tilde{A} + 3B_1\eta_0^2 + 5C\eta_0^4)^{3/2}\end{aligned} \tag{15}$$

where it is assumed that $A, B,$ and B_1 correspond to coefficients renormalized by fluctuations.

Let us consider first the case of very small concentrations of defects, i.e. the phase transition is almost the same as in the perfect crystal. Let us assume as well that $B_1 \gg B$, i.e. the phase transition is close to what would be a tricritical point if the effects of the fluctuations and of the defects were negligible. It was shown in [9] that only the second terms in the brackets appearing in the right-hand side of (15) are important close to the first-order phase transition point. (15) can then be written, for this temperature interval, in an approximate form:

$$\Phi(\eta_0) \approx \tfrac{1}{2}\tilde{A}\eta_0^2 + \tfrac{1}{4}\tilde{B}\eta_0^4 + \tfrac{1}{6}C\eta_0^6 + [(3B_1)^{1/2}/2\pi D^{3/2}](A_1^2 N - B_1 T/2)|\eta_0|^3. \tag{16}$$

One sees that the contributions of the defects and of the critical fluctuations to the coefficient of the third-order term are of opposite signs, i.e. the random temperature defects reduce the first-order character of the phase transition which is induced by the o.p. fluctuations, i.e. they 'smear' the transition.

One can estimate the defect concentration necessary to suppress essentially the first-order nature of the transition. As a dimensionless parameter characterizing the defect 'strength' it is natural to use [11, 12] $\varphi = A_1/Dd$ where d is the lattice spacing. The condition $\varphi \ll 1$ is necessary for the applicability of the approach used, otherwise one has to take into account not only the 'strength' A_1 but also the form factor of the defect [11]. The order of magnitude of other relevant quantities can be found in a way that is conventional for displacive transitions [13]. Assuming, e.g., that η is dimensionless, and $\eta_{at} \approx 1$, one obtains

$$D \approx T_{at}d^{-1} \qquad B \approx T_{at}d^{-3}$$

where T_{at} is a typical 'atomic temperature' ($T_{at} \approx 10^4$–10^5 K). Close to the tricritical point, $B \approx 0$ and $B_1 \approx \Delta \approx (\mu/K)(r^2/\lambda)$, B_1 is then expected to be of the order of $B\mu/K \approx T_{at}d^{-3}\mu/K$. One sees that the concentration in question can be estimated as

$$N_c d^3 \approx (\mu/K)(T_0/T_{at})/\varphi^2 \tag{17}$$

where T_0 is the temperature of the transition.

One sees that for a 'strongly displacive' system ($T_0 \ll T_{at}$) and strong enough defects (φ is not too small in spite of the fact that, in any case, it is much less than unity) one would find that $N_c d^3 \ll 1$, i.e. the phase transion becomes essentially smeared at defect concentrations which are much less than the 'atomic' one (d^{-3}). Experimentally, T_0/T_{at} is usually about 10^{-2}–10^{-3} and one sees that even for $\varphi \approx 10^{-1}$, i.e. for fairly strong defects, the concentration of defects producing complete smearing is not too far from the atomic concentrations, but it is quite possible that it is still much less. In this case one of the conditions of applicability of the perturbation theory to study the effects of defects is fulfilled. Let us recall that another condition of applicability of the perturbation theory of the effects of RLT defects is [11, 12] $\varphi^2 N d^2 r_c \ll 1$. Since for the pure crystal the first-order transition occurs fairly far from the points of the loss of stability against inhomogeneous fluctuations where $r_c = \infty$, one can estimate r_c, for displacive systems [13], as $d\ (T_{at}/T_c)^{1/2}$. One can see that the condition (17) does not contradict the condition of applicability of the perturbation theory. Indeed,

$$\varphi^2 N_c d^2 r_c \approx (\mu/K)(T_0/T_{at})^{1/2} \ll 1. \tag{18}$$

This means that the situation when the RLT defects diminish the jumps at the first-order transition, say, twice is well described within the perturbation theory. However, one needs another type of theory to describe the complete smearing of the first-order transition: when the first-order transition occurs close to the points of the loss of stability the perturbation theory both of the defect and of the fluctuation effects is no longer applicable.

3. Random local field defects

The above approach can be easily applied to a crystal with random local field (RLF) defects: instead of the last term in the right-hand side of (2) one has to put $-h(\boldsymbol{r})\eta$ (see [10]–[12]) with

$$h(\boldsymbol{r}) = \sum_j h_j \delta(\boldsymbol{r} - \boldsymbol{r}_j) \tag{19}$$

where $h_j = \pm h_0$, h_0 is a coefficient characterizing the 'strength' of the defect localized at $\boldsymbol{r} = \boldsymbol{r}_j$, and the sign of h_j is random. The defect contribution to the thermodynamic potential is now

$$-\frac{1}{2}\sum_{\boldsymbol{k}} \frac{\langle h_{\boldsymbol{k}} h_{-\boldsymbol{k}}\rangle}{A + 3B_1\eta_0^2 + 5C\eta_0^4 + Dk^2}. \tag{20}$$

For randomly distributed defects

$$\langle h_{\boldsymbol{k}} h_{-\boldsymbol{k}}\rangle = N h_0^2 \tag{21}$$

and the thermodynamic potential with both the RLF defects and the o.p. fluctuations taken into account has the form

$$\begin{aligned}\Phi(\eta_0) = {} & \frac{1}{2}A\eta_0^2 + \frac{1}{4}\tilde{B}\eta_0^4 + \frac{1}{6}C\eta_0^6 + \frac{Nh_0^2}{8\pi D^{3/2}}(A + 3B_1\eta_0^2 + 5C\eta_0^4)^{1/2} \\ & - \frac{T}{12\pi D^{3/2}}(A + 3B_1\eta_0^2 + 5C\eta_0^4)^{3/2}. \end{aligned} \tag{22}$$

The same reservations as those after (15) should be made and, in addition, a temperature- and η_0-independent term is omitted.

Under the same conditions as when writing (16) one obtains

$$\begin{aligned}\Phi(\eta_0) = {} & \tfrac{1}{2}A\eta_0^2 + \tfrac{1}{4}B\eta_0^4 + \tfrac{1}{6}C\eta_0^6 + (Nh_0^2/8\pi D^{3/2})(3B_1)^{1/2}|\eta_0| \\ & - (T/12\pi D^{3/2})(3B_1)^{3/2}|\eta_0|^3. \end{aligned} \tag{23}$$

One sees again that the defect and the fluctuation term compensate each other to some extent.

Let us estimate now the defect concentration (N_c) which smears essentially the fluctuation induced first-order transition. It has to be taken into account that the dimensionless parameter describing the defect strength is [12] $\psi = h_0 B^{1/2}/D^{3/2}$; this parameter should be smaller than unity to make it possible to apply the perturbation theory [12]. Putting η_0 equal to the o.p. jump at the transition which is of the order of magnitude of $\eta_0(T = 0)$ [9], i.e. can be estimated as [13] (T_0/T_{at}), and using the above estimations of the coefficient B_1, one finds

$$N_c d^3 \approx (1/\psi^2)(\mu/K)(T_0/T_{at})^2. \tag{24}$$

Comparing with (17) one sees that, at the same dimensionless 'defect strength', much smaller concentrations of the RLF defects produce essential smearing of first-order transtions: the right-hand side of (24) contains an additional small factor (T_0/T_{at}). This means that one of the conditions of applicability of the perturbation theory remains fulfilled for $N \approx N_c$: $N_c d^3 \ll 1$.

Let us recall that another condition of applicability of the perturbation theory of the effects of RLF defects is [11, 12] $\psi^2 N r_c^3 \ll 1$. Since for the pure crystal the first-order transition occurs fairly far from the points of the loss of stability against inhomogeneous fluctuations where $r_c = \infty$, one can again estimate r_c as $d\ (T_{at}/T_c)^{1/2}$. One sees that the condition (24) does not contradict the condition of applicability of the perturbation theory. This means that the situation when the RLF defects diminish the jumps at the first-order transition, say, twice is well described within the perturbation theory, as well as in the above-discussed case of RLT defects. However, once more, one needs another type of theory to describe the complete smearing of the first-order transition: when the first-order transition occurs close to the points of the loss of stability the perturbation theory both of the defect and of the fluctuation effects is no longer applicable.

4. Conclusions

Apart from the quite expected qualitative result that defects do smear first-order phase transitions the final results of this paper are formulae (15) and (22) for the thermodynamic potential depending on the Landau order parameter, which is, in fact, averaged over the volume (the zero Fourier component, η_0). These formulae differ from the classical (Landau) ones due to the account taken of the defects as well as of o.p. fluctuations. Using these formulae one can obtain the dependence on defect concentration of many characteristics of first-order transitions (the latent heat, the jump of the o.p., etc) as well as of thermodynamic quantities for the system close to the phase transition. We have considered a first-order displacive structural transition in the region of what would be the mean field tricritical point. In a pure crystal such a transition, due to the o.p. fluctuations, is a strongly first-order one; the jump of the o.p. is of the order of magnitude of the order parameter at $T = 0$. Nevertheless, strong enough random field defects were found essentially to smear the transition even when their concentration is much less than the atomic one. Within our approach we were able to consider the first stage of the smearing only: the decreasing of the jumps of various thermodynamic quantities at the phase transition for relatively small defect concentrations.

Let us emphasize once more the difference between our approach and that of Imry and Wortis [1]. Imagine the system to be just at the temperature where the thermodynamic potentials of the two phases are equal, i.e. at the thermodynamic phase transition point. Imagine then that there are local fluctuations of the system parameters, specifically of phase transition temperature, due to defects. How to estimate the possibility that the new phase will appear because of these fluctuations? The ‘textbook’ way is to compare the gain of the bulk energy due to the local fluctuation of the phase transition temperature and the surface energy due to the boundary between the phases. This type of reasoning used by Imry and Wortis is quite correct for a liquid but inapplicable for a solid.

Let us assume, as in the present paper, that the phase transition is associated with a change of mass density and not with a shear deformation. In a liquid, the change of density at nucleation of the new phase does not lead to any consequences for the surrounding liquid beyond the boundary layer, but this is not true for a solid where the surrounding phase proves to be inhomogeneously deformed. The energy of this deformation is, of course, positive and, which is most important, is proportional not to surface but to the volume of the nucleus. This means that an infinitely large nucleus surrounded by the old phase will have a positive energy even if its surface energy can be neglected and if the free system with the parameters of the region inside the nucleus would prefer the new phase (if, of course, the energy gain due to the transformation is not too large). In other words were the nucleation to be possible only inside the old phase there would always be overheating and overcooling at solid-state first-order phase transitions however slowly the temperature changed. In the real life this is the case normally: the surface of the sample may play an important role but that is another story.

A natural question arises as to whether the results of the present paper can be used to interpret the experimental data [2] on the synchrotron radiation scattering in $SrTiO_3$ instead of the results of [1] which are irrelevant to the problem as we hope to have shown. In the experiment two temperature-dependent length scales have been revealed but in our consideration there were not two length scales and thus we have to admit that our results do not explain the experimental results [2] as well as, of course, the results of [1] but due to another factor (see above). However we would like to mention that the occurrence of two length scales is not surprising *per se* if the crystal contains dislocations: it has been known for a long time [11, 14] that near an edge dislocation the nucleation of the new phase takes place and close to the second-order phase transition the radius of the nucleus is much larger than the correlation length. In a more general form this idea has been put forward in [15]. The irrelevance of the theory of Imry and Wortis to the two-length-scale problem has been emphasized as well in [16] from another point of view.

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