Elastic anomalies in cubic cyrstals in phase transitions with a three-component order parameter

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Elastic anomalies in cubic crystals at structural transitions describable by a three-component order parameter are considered in the framework of the renormalization-group method. It is shown that for small striction constants and certain ratios between the interaction constants there exists a region of temperatures in which the elastic compliances and thermal-expansion coefficients have power-law temperature dependences. The results obtained agree well with experimental data on the propagation of sound in the cubic phase of $RbCdF_3$ and $TlCdF_3$.

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As shown in Refs. 1 and 2, the interaction of the order parameter with the elastic degrees of freedom leads to the result that the phase transition changes from second-order to first-order. However, experimental studies indicate that the transition remains almost second-order and is accompanied by an appreciable increase of the elastic compliances. In agreement with this, for the example of a phase transition with a one-component order parameter in a cubic crystal it has been established[3] that near the transition there exist, in the general case, several regions in which the elastic constants display scaling behavior, with different critical indices in each of these regions. In the present paper we consider phase transitions describable by a three-component order parameter in cubic crystals. The general analysis of the elastic properties in this case is extremely complicated. However, if the striction constants are sufficiently small, then, as will be seen from the following, the critical indices for the compliances in the first scaling region (the furthest from the transition point) can be calculated with neglect of the influence of the elastic degrees of freedom on the fluctuations of the order parameter.

In all cases in which the order parameter φ_i transforms according to a three-component irreducible representation satisfying the Landau condition,[4] the part of the thermodynamic potential that depends only on φ_i can be written in the form

$$\Phi = {}^1/_2\alpha\varphi_i\varphi_i + {}^1/_4\beta_1(\varphi_i\varphi_i)^2 + {}^1/_4\beta_2(\varphi_1^4+\varphi_2^4+\varphi_3^4).$$

Following Ref. 5 we obtain for Φ the renormalization-group (RG) equations in first order in ε:

$$\begin{aligned}\dot\beta_1 &= (\varepsilon - 11\beta_1 - 6\beta_2)\beta_1,\\ \dot\beta_2 &= (\varepsilon - 12\beta_1 - 9\beta_2)\beta_2.\end{aligned} \qquad (1)$$

The integral curves of these equations are represented in the Figure. For $\beta_1 + 3\beta_2 > 0$ and $\beta_1 > 0$ they converge to a stable fixed point, and, consequently, in this region of values of β_1 and β_2, in the absence of coupling with the elastic degrees of freedom, the transition will be second-order with critical indices determined by the coordinates $\beta_1^* = \varepsilon/11$ and $\beta_2^* = 0$ of this fixed point (point b in the Figure). Here, the case $\beta_2 > 0$ corresponds to a transition to a rhombohedral phase ($\varphi_1 = \varphi_2 = \varphi_3$), and the case $\beta_2 < 0$ corresponds to a transition to a tetragonal phase ($\varphi_1 = \varphi_2 = 0$, $\varphi_3 \neq 0$).

In a clamped crystal (u_{ij} = const) the strictional coupling of the order parameter with the elastic degrees of freedom leads to the appearance in the thermodynamic potential of an interaction of fourth order in φ_j, corresponding to exchange of an acoustic phonon. Because of the anisotropy of the elastic properties, the tensor constant v_{ijkl} of this interaction is found to depend on the direction of the momentum transfer. As was shown in Ref. 2, for an isotropic n-component model with an anisotropic compressible lattice, in the presence of such an interaction the order of the transition changes from second to first if $n < 4$. This result follows from the fact that the solution of the RG equations for the complete vertex $\Gamma = \beta + v$ has, for sufficiently large values of the correlation length ξ, a pole singularity, with $\Gamma(\xi) < 0$ near the pole. The divergence of Γ also implies that the complete system of equations for β and v does not have stable fixed points. In particular, the isotropic fixed point $\beta^* = \varepsilon/(n+8)$, $v^* = 0$ for $n < 4$ turns out to be unstable with respect to deviations of v from its fixed-point value.[2]

For the potential considered in the present paper, with cubic anisotropy in the φ_i, it is possible, as in Ref. 2 (with certain complications associated with the tensor character of v_{ijkl}), to show that the fixed points with $v^*_{ijkl} = 0$ and with β_1^* equal to the corresponding coordinates of one of the fixed points of Eqs. (1) are also unstable against arbitrary anisotropic perturbations. Evidently, a change in the order of the transition from second to first, analogous to that studied in Ref. 2, also occurs in this case. Moreover, these unstable points are simple singular points of the complete

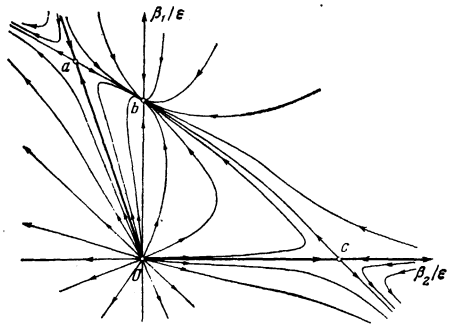

FIG. 1. Integral curves of the renormalization-group equations: a) (1/9, −1/27); b) (1/11, 0); c) (0, 1/9).

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system of equations. Therefore, for small bare values of the components v_{ijkl} (i.e., for small striction constants q_i, $v_{max} \sim q_{max}^2 S_{max} \ll \beta_i$), the integral curves of this system in the space of the parameters β_i, v_{ijkl} lie close to the (β_1, β_2)-plane right up to the nearest (in the direction of increase of ξ) fixed point and differ little from the integral curves of Eqs. (1). Consequently, for not-too-large values of the correlation length [such that the $\beta_i(\xi)$ do not reach the indicated points], we can put the v_{ijkl} approximately equal to zero, and describe the dependence of the β_i on ξ by Eqs. (1).

Everything we have said above about the behavior of the solutions is also valid for a free crystal, since the long-range interaction that arises through uniform deformations in this case reduces to a self-consistent renormalization of the coefficient α and the form of the RG equations does not change. Generally speaking, such a renormalization can also cause the transition to become first-order if C_v is sufficiently large near T_0, since for large C_v the renormalized coefficient α' becomes a nonunique function of α.[1] However, the estimates given in Ref. 2 show that this branching arises at values of ξ of the same order as for the runaway due to the anisotropic interaction via the acoustic phonons. Thus, for small q_i and in the free crystal there exists a region in which we can neglect the influence of the elastic degrees of freedom on the order-parameter fluctuations. In the present paper we shall confine ourselves to considering precisely this region.

Anomalies in the elastic properties arise as a result of the interaction of uniform deformations with the order parameter; in the case under consideration this interaction has the form

$$q_1 e_1 \varphi_i \varphi_i + q_2 \left[e_2 (\varphi_1^2 - \varphi_2^2) + \frac{1}{3^{1/2}} e_3 (\varphi_1^2 + \varphi_2^2 - 2\varphi_3^2) \right] + q_3 \sum_{i \neq j} u_{ij} \varphi_i \varphi_j, \quad (2)$$

where e_1, e_2, and e_3 are those linear combinations of uniform deformations that diagonalize the matrix of the elastic constants in the cubic phase:

$$e_1 = \frac{1}{3^{1/2}} u_{ii}, \quad e_2 = \frac{1}{2^{1/2}} (u_{11} - u_{22}), \quad e_3 = \frac{1}{6^{1/2}} (u_{11} + u_{22} - 2u_{33}).$$

In the expression (2) q_3 is nonzero only when the symmetric square of the representation according to which the φ_i transform contains the representation according to which the shear components u_{ij} $(i \neq j)$ of the deformation tensor transform.

In the cubic phase the elastic compliances are expressed in terms of correlation functions of the e_i and u_{ij} as follows:

$$\begin{gathered} S_{11} + 2S_{12} = \langle e_1^2 \rangle - \langle e_1 \rangle^2, \\ S_{11} - S_{12} = \langle e_2^2 \rangle - \langle e_2 \rangle^2 = \langle e_3^2 \rangle - \langle e_3 \rangle^2, \\ S_{44} = \langle u_{ij}^2 \rangle - \langle u_{ij} \rangle^2, \quad i \neq j. \end{gathered}$$

Because of the coupling (2) these expressions contain singular parts proportional to correlators of the corresponding quadratic forms of components of the order parameter, so that the scaling dimensions of the deformations coincide with the scaling dimensions of these quadratic forms, viz.,

$$\begin{gathered} \Delta_{e_1} = \Delta_{\varphi_i \varphi_i}, \quad \Delta_{e_2} = \Delta_{\varphi_1^2 - \varphi_2^2}, \\ \Delta_{e_3} = \Delta_{\varphi_1^2 + \varphi_2^2 - 2\varphi_3^2}, \quad \Delta_{u_{ij}} = \Delta_{\varphi_i \varphi_j}, \quad i \neq j. \end{gathered} \quad (3)$$

But the dimensions of $\varphi_i \varphi_i$, $\varphi_1^2 - \varphi_2^2$, etc. can be determined with the aid of the RG equations for the fields λ_1, λ_2, λ_3, and λ_{ij} thermodynamically conjugate to these quantities, putting $v_{ijkl} = 0$, as already indicated.

In first order in ε these equations have the form

$$\begin{gathered} \dot{\lambda}_1 = (2 - 5\beta_1 - 3\beta_2)\lambda_1, \\ \dot{\lambda}_{2,3} = (2 - 2\beta_1 - 3\beta_2)\lambda_{2,3}, \\ \dot{\lambda}_{ij} = (2 - 2\beta_1)\lambda_{ij}. \end{gathered} \quad (4)$$

For bare values of β_i lying in the vicinity of the lines $\beta_1 + 3\beta_2 = 0$, $\beta_2 = 0$, or $\beta_1 = 0$, the solutions of Eqs. (1) in the considered region of values of the correlation length tend exponentially to the fixed points a, b, and c, respectively. Therefore, in these cases the β_i in (4) can be replaced by their values at the corresponding fixed point. The resulting coefficients of the λ_i in the right-hand sides of Eqs. (4) will be the dimensions of these fields, which are connected with the dimensions of interest to us by the simple relations $\Delta_{\lambda_1} + \Delta_{\varphi_i \varphi_i} = d$, etc.[5]

Finally, we obtain

1) $\beta_1 \approx -3\beta_2$, $\beta_2 < 0$:

$$\Delta_{e_1} = 2 - {}^5/_9 \varepsilon, \quad \Delta_{e_2} = \Delta_{e_3} = 2 - {}^8/_9 \varepsilon, \quad \Delta_{u_{ij}} = 2 - {}^7/_9 \varepsilon; \quad (5)$$

2) $\beta_2 \approx 0$, $\beta_1 > 0$:

$$\Delta_{e_1} = 2 - {}^6/_{11} \varepsilon, \quad \Delta_{e_2} = \Delta_{e_3} = \Delta_{u_{ij}} = 2 - {}^9/_{11} \varepsilon; \quad (6)$$

3) $\beta_1 \approx 0$, $\beta_2 > 0$:

$$\Delta_{e_1} = \Delta_{e_2} = \Delta_{e_3} = 2 - {}^2/_3 \varepsilon, \quad \Delta_{u_{ij}} = 2 - \varepsilon. \quad (7)$$

Thus, for values of the β_i belonging to the neighborhood of the lines indicated, the elastic compliances and also the thermal-expansion coefficients α_{ij} in the cubic phase display power-law dependences, the exponents of which can be found without difficulty from (5) – (7).[5]

1) $\beta_1 \approx -3\beta_2$, $\beta_2 < 0$ (on the boundary with the tetragonal phase):

$$\gamma_{\alpha_{11}} = \gamma_{s_{11} + 2s_{12}} = {}^1/_{18}, \quad \gamma_{s_{11} - s_{12}} = {}^7/_{18}, \quad \gamma_{s_{44}} = {}^5/_{18};$$

2) $\beta_2 \approx 0$, $\beta_1 > 0$ (the vicinity of the triple point):

$$\gamma_{\alpha_{ii}} = \gamma_{s_{11} + 2s_{12}} = {}^1/_{22}, \quad \gamma_{s_{11} - s_{12}} = \gamma_{s_{44}} = {}^7/_{22};$$

3) $\beta_1 \approx 0$, $\beta_2 > 0$ (on the boundary with the rhombohedral phase):

$$\gamma_{\alpha_{ii}} = \gamma_{s_{11} + 2s_{12}} = \gamma_{s_{11} - s_{12}} = {}^1/_6, \quad \gamma_{s_{44}} = {}^1/_2.$$

It is necessary to note also that the critical temperature T_c in the power dependences under consideration is determined by the condition $2\alpha + 5\beta_1 + 3\beta_2 = 0$ and differs from the transition temperature T_0. Since the scaling dimensions (5) – (7) have the same values below the transition,[6] we can also find the critical indices in the low-symmetry phases. Determining the expressions for the pair correlators of the quantities e_i and u_{ij} in terms of the compliances with allowance for the form of the matrices $S_{\mu\nu}$ in these phases, we obtain, e.g., for $\beta_2 \approx 0$.

1) tetragonal phase:

$$\begin{gathered} \gamma_{2(s_{11} + s_{12}) + 4s_{13} + s_{33}} = {}^1/_{22}, \\ \gamma_{s_{11} - s_{12}} = \gamma_{s_{44}} = \gamma_{s_{66}} = \gamma_{s_{11} + s_{12} - 4s_{13} + 2s_{33}} = {}^7/_{22}, \\ \gamma_{(s_{11} - s_{12} + s_{13} - s_{33}} = {}^2/_{11}, \\ \gamma_{2\alpha_{11} + \alpha_{33}} = {}^1/_{22}, \quad \gamma_{\alpha_{11} - \alpha_{33}} = {}^2/_{11}; \end{gathered}$$

2) rhombohedral phase (in the axes of the original cubic phase):

$\gamma_{S_{11}+2S_{12}}=1/22$, $\gamma_{S_{11}-S_{12}}=\gamma_{S_{44}}=\gamma_{S_{66}}=\gamma_{S_{14}-S_{24}}=7/22$,
$\gamma_{S_{14}+2S_{34}}=2/11$, $\gamma_{\alpha_{11}}=1/22$, $\gamma_{\alpha_{12}}=2/11$.

The results obtained have been compared with the experimental data on the propagation of sound in $RbCdF_3$ and $TlCdF_3$ crystals undergoing a phase transition from the cubic (O_h^1) to the tetragonal (D_{4h}^{18}) phase at the temperatures $T_0 = 124$ K and 192 K, respectively.[7] The order parameter transforms according to a three-dimensional irreducible representation of the group O_h^1, associated with the vector $\mathbf{k} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. In this case, $q_3 \neq 0$ in (2).

In the cubic phase the elastic compliances calculated from the sound velocities have, in the vicinity of the transition, temperature dependences of the following form:

1) $RbCdF_3$

$$2\rho(S_{11}-S_{12}) = [1190.2+70.9\tau+85.4(1/\tau)^{0.35}]\cdot 10^{-4} \text{ (sec/km)}^2$$

for $11° < T-T_0 < 66°$, $T_c = 130.8$ K;

$$\rho S_{44} = [2431.8-33.1\tau+34.1(1/\tau)^{0.29}]\cdot 10^{-4} \text{ (sec/km)}^2$$

for $11° < T-T_0 < 56°$, $T_c = 130.0$ K;

2) $TlCdF_3$

$$2\rho(S_{11}-S_{12}) = [1978.6+172.7(1/\tau)^{0.36}]\cdot 10^{-4} \text{ (sec/km)}^2$$

for $9° < T-T_0 < 38°$, $T_c = 196.8$ K.

Here $\tau = (T-T_c)/T_c$. The terms linear in τ in the expressions for the compliances of $RbCdF_3$ are temperature dependences far from the transition, extrapolated into the critical region. The formulas given for $S_{11}-S_{12}$ in both crystals give values of the sound velocities that differ (in the indicated intervals of $T-T_0$) from the experimental values by not more than $\pm 3.5\times 10^{-3}$ km/sec, while for S_{44} the difference is not more than $\pm 1.4\times 10^{-3}$ km/sec.

The values of the critical indices for $RbCdF_3$ ($\gamma_{S_{11}-S_{12}} = 0.35$ and $\gamma_{S_{44}} = 0.29$) are in satisfactory agreement with the theoretical values for a transition to the tetragonal phase with $\beta_1 \approx -3\beta_2$ ($\gamma_{S_{11}-S_{12}} = 7/18 \approx 0.39$ and $\gamma_{S_{44}} = 5/18 \approx 0.28$). Evidently, the same ratio between the β_i is also realized in $TlCdF_3$ ($\gamma_{S_{11}-S_{12}} = 0.36$). Unfortunately, the small number of experimental points for S_{44} in the critical region does not permit us to establish the value of $\gamma_{S_{44}}$ reliably in this case.

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