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On the connection between the appearance of two scales and the central peak in the scattering spectra near phase transitions points in crystals

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The paper proposes a mechanism for the formation of a dynamic central peak observed in scattering spectra as a result of an increase in the size of nuclei on dislocations as the phase transition point is approached. An equation of motion of the interphase boundary for the nucleus is derived, and from its solution an estimate is obtained for the frequency range of the central peak.

Keywords: phase transition, central peak, temperature gradient of a local phase transition, equation of motion for a nonstationary phase boundary.

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1. Introduction

As it is known, even in the late 60s to explain the universality of the critical behavior of materials near points of second kind phase transitions (PTs), the thermodynamic scaling hypothesis was formulated. Its main predictions were soon supported by theoretical model calculations performed within the framework of ε and 1/N expansions of the renormalization group method. The physical clarity of the scaling pattern of PT and the experimental confirmation of the predicted relationships between the values of the critical indices of thermodynamic variables (susceptibility, order parameter, heat capacity, etc.) greatly strengthened confidence in the correctness of the scaling hypothesis. The existence of critical correlation length of thermal fluctuations of the order parameter, growing indefinitely at the point of 2-nd kind PT, began to seem not only expected, but also even simply a self-evident phenomenon. The most popular way to observe it at that time was through experiments on the scattering of thermal neutrons, X-rays and light. The subsequent application of scaling arguments to describe the dynamics of fluctuations of the order parameter resulted in the prediction of the existence of a characteristic time scale associated with an unlimitedly growing radius of thermal fluctuations and also diverging at the transition point („critical slowing-down").

Near the critical points of liquids both of these tendencies manifested themselves quite clearly in experiments. However, for a number of PTs in solids the picture unexpectedly turned out to be more complex. The first experiments where a qualitative difference of the pattern of the excitation spectrum from the expected one were experiments on inelastic scattering of thermal neutrons in the vicinity of a structural PT of the displacement type in $SrTiO_3$ (see [1] and cited there publications). In addition to the expected „side" phonon peak, the frequency of which decreased as the temperature approached the PT point (but, as it turned out, actually reached saturation in its immediate vicinity), a quasi-static „central peak" (CP) was discovered, the intensity of which increased much faster, and this growth continued just up to the PT point. The same was observed in subsequent experiments on Raman scattering, carried out both on $SrTiO_3$ and other crystals from the perovskite family ($RbCaF_3$, $LaAlO_3$, $KMnF_3$, $LiTaO_3$, $LiNbO_3$) [1–7], as well as in numerous experiments on inelastic neutron scattering, which revealed the occurrence of CP with a similar temperature dependence.

In addition to the presence of two time scales, greatly different in magnitude (at least by 4 orders of magnitude), later, during the scattering of neutrons and X-rays, the existence of two spatial lengths was discovered, the smaller of which could be associated with the thermal correlation radius, and the origin of the larger one, having the order of microns, remained unknown (see [2,8] and the publications cited there). It is appropriate to note here that in addition to these experiments, back in 1956 at $\alpha-\beta$ PT in quartz a very strong increase in the light intensity was observed (approximately by 10^4 times compared to the intensity at room temperature), scattered at an angle of 90° to the optical axis, on large-scale (with a characteristic size $\sim 30\,\mu$m) optical inhomogeneities arising in narrow vicinity of temperatures (~ 0.1 K) [9]. Later, the intensity of light scattering at small angles to the optical axis was measured, and it was found that its relative increase was is even greater ($\sim 10^6$) [10,11].

The situation became even more serious when the occurrence of two characteristic scales was discovered not only for structural PTs, but also in high-resolution experiments on

X-ray and neutron scattering for magnetic PTs in a number of metals and their compounds (Tb, Ho, UO_2, NpAs, USb, UP, UP_2Al_3) (see review [2], and [12–21]), and then in invar alloys $Fe_{1-x}Ni_x$ [22–25].

Thus, incongruities appeared: in almost every crystal, experiments performed with high spatial or temporal resolution demonstrated an pattern of PT universal, but qualitatively contradictory to the results of both static and dynamic generally accepted renormalization group theory!

Of course, starting from the publication of the first unexpected experimental results [26,27], a number of considerations were made about their physical interpretation. First, the possible role of defects was pointed out. Experimental evidence indicated that they undoubtedly played some role, but it was unclear how big this role was. Attempts to purposefully introduce defects or studies with crystals of higher quality did not significantly change the results: PT demonstrated „critical" behavior, did not blur, and the values of the effective indices did not change noticeably. Secondly, a scenario was proposed in which PT mechanism would include not only a soft phonon mode, but also a mode describing the collective movement of dynamic clusters (with a rather high amplitude of the order parameter) as a whole. This intuitively attractive, non-defective mechanism has received some support in two-dimensional numerical experiments [1]. However, no corresponding analytical theory was created. Besides, it was experimentally shown that X-ray scattering from the larger length is not induced in a significant part of the sample volume.

In turn, numerous subsequent works on Raman scattering of light in quartz (see [3,28] and the publications cited there), $LiNbO_3$ (see [3,5,7] and cited publications) and $LiTaO_3$(see [3,4,6] and cited publications) showed that the intensity and half-width of the CP strongly depend on the growth conditions of these crystals, and in addition to abnormally growing, narrowing CPs near PT points, the presence of CPs (possibly having a different mechanism of origin [5]) was discovered far from these points. A relationship was also established between the characteristics of CP and the degree of defectivity of crystals, which are especially sensitive to the concentration of extended defects, such as dislocation threads and loops [7].

Thanks to all these facts, a general oppinion gradually emerged in the literature that it is defects that are the main cause of the occurence of CPs in real crystals.

From the theoretical point of view, the problem was that standard models of weakly disordered crystals, processed using the so-called replica trick within the framework of the general renormalization group method, implied the dominance of one (maximum) correlation length and the corresponding one (also maximum) time scale — the influence of all smaller scales and times turned out to be insignificant compared to large-scale long-lived thermal fluctuations. At the same time, the experiment clearly indicated the existence of two lengths, and the larger of them (as well as the larger of the two times) had a mysterious character! This seemed more strange since, as a rule, the most carefully grown crystals were selected for experiments, and subsequent processing of the samples was also carried out in such a way that they contained the minimum possible number of defects.

Where did the generally accepted scaling hypothesis and the renormalization group theory based on it miss the possibility of the existence of such a global PT scenario? Please note that in the usual renormalization group calculation scheme it is assumed that when approaching PT point the properties of defects do not change. In particular, their effective sizes are considered to be the same as those far from the PT point.

It is clear that the qualitative difference between these ideas and the results of the above scattering experiments means their physical inadequacy. In particular, remarkable optical experiments in NH_4Br clearly showed the occurrence of nuclei of a new phase near dislocations, with their subsequent growth as they approach the global PT point [29]. Besides, additional electron microscopy [14], as well as X-ray experiments [19,20], carried out in the same $SrTiO_3$, also demonstrated that this scenario, based on the effect of local PT near extended defects (dislocations, their clusters, cracks, etc.) is actually realized in real crystals.

These facts suggest a physical reason for the inadequacy of the standard renormalization group calculation scheme: the effective size of the most powerful defects does not remain constant, but can increase, moreover, faster than the radius of thermal fluctuations changes. Thus, the radius of thermal fluctuations turns out to be already not the largest spatial scale; therefore, the entire spatial picture of the PT process development can also qualitatively change.

For the first time, the possibility of a nonclassical scenario of magnetic PT of the 2nd kind in crystal model with postulated large-scale fluctuations of the Curie temperature was indicated in [30]. Then, in [31], the almost inevitable presence of dislocations in actual crystals was pointed out as a specific physical reason for such fluctuations occurrence. The analysis carried out in [31] showed that in dislocation crystals, depending on the parameters of PT and the dislocation ensemble, both the „percolation" PT scenario [30] and PT are possible. when the macroscopic order parameter is initially localized on the „framework" of dislocations. Later, in [32] the crossover mechanism was traced in the global spatial nature of PT process when the dispersion parameters and the correlation scale of the local PT temperature field changed. The results obtained were then used to solve the „problem of two lengths" discovered in experiments on the scattering of X-rays and thermal neutrons in dislocation crystals [21,33].

In particular, the general results obtained in theoretical papers [30–33] were fully confirmed in a series of subsequent experimental studies on small-angle neutron scattering and depolarization of neutron beams in Invar alloys $Fe_{1-x}Ni_x$ [22–25], in a comprehensive study, including small-angle X-ray scattering, in the alloy

V_2H [16,17,21], as well as in local X-ray scattering measurements for $SrTiO_3$ [19,20].

Thus, the „problem of two lengths“ in experiments on elastic scattering near PT points in actual crystals can now be considered solved in principle. However, the origin of the 2 time scales remains unclear. In particular, within the framework of phenomenological models such as two coupled oscillators or oscillator interacting with low-frequency relaxator, the key factor remains unknown — the physical origin of the low-frequency mode associated with the supposed conditionally soft mode. As a result, the corresponding results of calculation of the intensity and half-width of the dynamic CP observed in the spectra during inelastic scattering depend on several fitting parameters (at least three). Therefore, even good quantitative agreement between the results of such calculations and experimental data, obtained for sets of „optimal“ values of such parameters for specific materials, does not mean that CP problem was solved. Note that the CP problem solution cannot be considered complete until it is clarified what properties of the crystal and features of the phase transition are associated with both the very existence of the additional low-frequency mode and its characteristic frequency, half-width and intensity.

In addition to the unresolved question of CP physical nature, the question of the possible existence of relationship between abnormally large spatial and temporal scales also remains open.

This statement proposes a new approach to these issues solution.

2. Process of nucleation of new phase in the elastic field of ensemble of dislocations

In this Section we briefly recall how the process of nucleation of new phase in crystal with dislocations and its further spatial development as the temperature approaches the PT point is theoretically described. At the initial stage of this process, relatively far from PT point, the appearance of nuclei occurs near dislocation lines, in regions where, due to large elastic deformations, the local PT temperature increases, such that $T_c(r) > T$. At this stage, when the size of the occurred nucleus is minimal, to calculate $T_c(r)$ it is sufficient to take into account only the dominant contribution to the local deformation from the dislocation closest to the given nucleus. Since the result does not depend on the structure of the entire dislocation ensemble, the calculation is reduced to solving the model problem of the new phase nucleation on a single dislocation.

For simplicity, we will assume that the order parameter (OP) of PT η is single-component, the dislocation is rectilinear and purely edge, and the crystal 1s elastically isotropic. The free energy density in such model can be written as an expansion in powers of OP:

$$\Phi = \frac{1}{2}g(\nabla\eta)^2 + \frac{1}{2}\alpha_o(T - T_o)\eta^2 + \frac{1}{4}B\eta^4 + \frac{1}{4}D\eta^6 + \frac{1}{2}A\eta^2\big(\varepsilon_{ii} + \varepsilon^d_{ii}(r)\big) + \frac{K}{2}\varepsilon^2_{ii} + \mu\left(\varepsilon_{ik} - \frac{1}{3}\delta_{ik}\varepsilon_{ll}\right)^2, \tag{1}$$

Where coefficient $B > 0$ for 2nd kind PT and $B < 0$ for 1st kind PT, A is strictive coefficient, K and μ — bulk and shear modulus, respectivtly, ε_{ik} — tensor of elastic deformations, and $\varepsilon^d_{ii}(r)$ — contribution to dilatational deformation due to edge dislocation dislocation, recorded in polar coordinates [34]:

$$\varepsilon^d_{ii}(r) = \frac{b\mu\sin\varphi}{\pi\big(K + (4/3)\cdot\mu\big)r}. \tag{2}$$

After eliminating the field of elastic deformations and minimizing the free energy, we obtain an equation for the OP field distribution in the vicinity of the dislocation

$$-g\Delta\eta + \alpha_o\left(T - T_o + \frac{A}{\alpha_o}\varepsilon^d_{ii}(r)\right)\eta + B^*\eta^3 + D\eta^5 = 0, \tag{3}$$

where B^* — coefficient renormalized by striction consideration.

From (3) it is obvious that the dislocation presence changes the local temperature of PT $T_c(r)$, and the ratio

$$\frac{A}{\alpha_o} \approx \left(\frac{K}{T_o}\right)\left(\frac{\partial T_c}{\partial p}\right)$$

(where T_c — PT temperature in dislocation-free crystal, p — pressure), and from (2) it follows that for any sign of the coefficient A there is a region in which $T_c(r) > T$.

In the case of 2nd (or weak 1st) kind PT, the value of the temperature at which the nucleus appears T_n and its size R_n can be found exactly (or approximately) as the bifurcation point of the linearized differential equation (3) [35]. For our purposes, it is enough to have estimates

$$T_n - T_c \approx \frac{1}{\alpha_o g}(Ab)^2,$$

$$R_n \approx \frac{g}{Ab} \approx \sqrt{\frac{g}{\alpha_o(T_n - T_c)}} = r_c(T = T_n). \tag{4}$$

They are obtained from interpretation of the linearized equation as the Schrödinger equation for a particle having a state localized in a potential well.

It is obvious that with temperature decreasing the size of the nucleus $R_*(T - T_c)$ will increase. It is not difficult to obtain its temperature dependence if we note that the term $g\Delta\eta$ is important only within the interphase boundary, which has a thickness equal to about the correlation radius $r_c \sim (T - T_c)^{-1/2}$. Assuming that $R_* \gg r_c$, from (2), (3) we obtain

$$R_*(T) \approx \frac{bK\left(\frac{\partial T_c}{\partial p}\right)}{2\pi(T - T_c)}, \tag{5}$$

i.e., the size of the nucleus actually increases as it approaches PT point faster than the correlation radius. Thus, estimate (5) suggests that thermal fluctuations may not play a dominant role in PT process in crystals with dislocations.

To make sure that PT scenario other than that dictated by large-scale thermal fluctuations is possible, it is necessary to find out how a non-zero average value of the macroscopic PT can arise on a set of nuclei that initially have have OPs of different signs. The answer to this question depends on the spatial structure of the dislocation ensemble (DE) of the sample.

The latter is determined by the conditions of crystal growth and its subsequent treatment (mechanical and chemical) necessary for experimental measurements and further operation. Accordingly, DE structures can vary greatly.

For the model case of an absolutely chaotic arrangement of dislocations with a given average density $n_d = R_d^{-2}$ (R_d — average distance between dislocations) in [31] it was shown that at temperature of nucleation

$$T_n > T_c + K\left(\frac{\partial T_c}{dp}\right)\varepsilon_{ii}^{d}\ (r = R_d)$$

the occurrence of macroscopic OP corresponds to a set of cylindrical nuclei with characteristic radius $R \ll R_d$, i.e. the PT occurs on „framework“ of DE. In this case, the total volume fraction of the new phase $x(T)$ is small, so that the PT is experimentally observed as blurred. When the sign of the inequality is reversed, the PT temperature corresponds to the occurrence of a cluster of new phase, which has the same sign of the local OP values for all nuclei, the merge of which forms this cluster, penetrating the entire sample. The spatial structure of this cluster is close to the geometric structure of the „infinite cluster“ in the classical percolation theory. In this case, the total fraction of the new phase $x(T)$ is not small and PT is experimentally perceived as not blurred, and the behavior of thermodynamic variables is described by critical indices.

3. Manifestation of heterophase spatial structure in experiments by inelastic scattering

The results of observations of elastic scattering (neutrons, X-rays and light) are determined by the equilibrium configuration of the nuclei, i.e. the equilibrium position of their IPB. As is known, the IPB is exposed to a thermodynamic force P (actually, pressure), directed along the normal and equal to the difference in the free energies of the phases per unit area. The magnitude of this force depends on the proximity of the crystal to PT point. $P = P(T - T_c)$. As was established in the previous Section that DE creates a spatially inhomogeneous field of elastic deformations and, due to its strictive interaction with the OP, the associated inhomogeneous distribution of local PT temperatures $T_c(r)$. In thermodynamic equilibrium IPBs are located so that the force acting on them is $P\big(T - T_c(R)\big) = 0$.

It is obvious that with a small deviation by δR from the equilibrium configuration the restoring force appears acting on IPB

$$\delta P = -\frac{dP}{dT}\,\nabla T_c \delta R. \qquad (6)$$

Considering it the IPB movement equation is

$$M(\delta\ddot{R}) + \gamma\delta\dot{R} + \beta\delta R = f(t), \qquad (7)$$

where the point above denotes the time derivative, M — mass of unit area of IPB, γ — coefficient of viscous friction force, $f(t)$ — force acting on IPB, including its random component and $\beta \equiv -\frac{dP}{dT}\nabla T_c$.

Let us now consider the dynamics of the nucleus IPB displacement in the temperature range where the shift of the local PT temperature $T_c(r)$ from dislocations not closest to the given nucleus can be neglected. In this case we can use (2), from where value ∇T_c on IPB (at $r = R_*$, where R_* — size of nucleus) is equal to

$$\nabla T_c\big(r = R_*(T)\big) = \frac{\mu}{\pi\big(K + \frac{4}{3}\mu\big)}\,\frac{b}{R_*^2}\left(\frac{\partial T_c}{\partial p}\right). \qquad (8)$$

In (8) for $R_*(T)$ formula (5) is valid, and since $R_* \gg b$, it is clear that the variable ∇T_c is „universally“ (i.e., regardless of the properties of the dislocation-free material and the type of phase transition) small. Accordingly, the restoring force in (7) is also small, decreasing as the temperature approaches the PT point. For displacement-type PT an additional small factor that reduces the value of ∇T_c is the multiplier — $\frac{dP}{dT_c} \approx \Delta S \ll 1$, where ΔS — the entropy jump. The slowdown of the IPB dynamics is also facilitated by the rather large mass value $M \approx m r_c$, since $r_c \approx \sqrt{\frac{g}{\alpha_o(T - T_c)}}$. Since the value of the viscosity coefficient γ strongly depends on the specific material experiencing a particular PT, it is advisable to use equation (7) to estimate the characteristic time of the IPB dynamics τ_{cp} in two limiting cases

$$\tau_{cp} \approx \left(\Delta S\,\frac{1}{r_c R_*^2}\right)^{-1/2} \quad (\gamma^2 \ll \beta M), \qquad (9)$$

$$\tau_{cp} \approx \left(\Delta S\,\frac{1}{R_*^2}\right)^{-1} \quad (\gamma^2 \gg \beta M). \qquad (10)$$

When writing estimates (9), (10), we used atomic normalization, in which the units of time, length, energy are equal to 10^{-13} s, 10^{-8} cm, $1\,\mathrm{eV} = 1.1 \cdot 10^4$ K, respectively, and the unit of mass is equal to the mass of the atom.

To understand how small the CP frequencies corresponding to cases (9), (10) are compared to normal phonon frequencies, let us make a simple numerical estimate for the size of the nucleus R_* in the temperature range $0.1 < T - T_c < 10$ K, commonly used in experiments. Taking into account that the values of the derivative $\frac{dT_c}{dp} \sim (1{-}10)$ deg/kbar, from (5) we obtain $R \sim \big((10{-}1){-}(0.1{-}0.01)\big)\,\mu$m, respectively, near and far from the temperature T_c. These values are close to those observed experimentally for elastic scattering.

To estimate the ratio of CP width $\Delta\Omega$ to the frequency of normal phonon modes ω, we choose the displacement-type PT, since until now it was difficult to explain CP appearance specifically with this type of PT (in contrast to order-disorder type PTs, where the appearance of CPs is expected due to uncorrelated hops of atoms over potential barriers inside cells) [1,36]). Substituting dimensionless values $\Delta S \sim 10^{-2}$, $r_c \sim 10^2$, $R \sim 10^5$, we get in case (9) $\frac{\Delta\Omega}{\omega} \sim 10^{-5}$, and in case (10) $\frac{\Delta\Omega}{\omega} \sim 10^{-7}$. Accordingly, CP line width is $\Delta\Omega \sim 10^8\,\mathrm{s}^{-1}$ and $\Delta\Omega \sim 10^6\,\mathrm{s}^{-1}$ for small and large attenuation coefficients γ.

The estimates obtained explain the small value of the observed CP width, which by 4−5 orders of magnitude is smaller than the characteristic phonon frequencies [37]. They also help to understand why, at a sufficiently high resolution $\sim (10^6-10^7)\,\mathrm{s}^{-1}$, experiments for some materials can resolve the CP linewidth, while in most others CP linewidth remains unresolvable.

4. Discussion

Based on the assumption of the universal role of the IPB dynamics of nuclei appearing on dislocations already in the symmetric phase, the paper obtained estimates for the width of experimentally observed CPs in inelastic scattering spectra. Note that the dislocation density $n_d = R_d^{-2}$ in most of the high-quality crystals selected for scattering experiments was small, so that $R_d \gg R_*$ (up to the actually achievable vicinity of PT point $\sim 0.1\,\mathrm{K}$). Therefore, the contribution to the deformation field on the given nucleus from distant dislocations can be neglected in comparison with the field of the dislocation closest to it, which was used in calculating the CP width.

In addition to the numerical results, two general conclusions can be immediately drawn from the expressions for the characteristic size of the nucleus (5) and the time associated with the dynamics of CP (9), (10). Firstly, it is obvious that especially narrow CPs shall be observed near tricritical points (at which $\Delta S \to 0$). This prediction is in agreement with experimental observations. Secondly, combining (5) and (9), we find the temperature dependence of CP half-width $\gamma_R \sim |T-T_c|$, and combining (5) and (10) we obtain $\gamma_R \sim |T-T_c|^2$. Both of these dependences were observed for different crystals [5,6,37].

It is especially interesting that the dependence $\gamma_R \sim |T-T_c|$ appears for the case of IPB dynamics with relatively low damping (9), which is typical for displacement-type PTs. Within the framework of the phenomenological description of CP in the traditional Ginzburg−Landau dynamic scheme, such dependence of the „critical slowing down“, on the contrary, is unambiguously interpreted as evidence of the order-disorder type PT presence. It is possible that the above difference explains the solution to the long-standing question about the type of PT in such, for example, crystals as $LiNbO_3$ and $LiTaO_3$.

Despite the universality of the proposed mechanism of CP formation and the agreement of the obtained estimates with experimental data, note that they were obtained within the framework of a simplified elastic-isotropic model with a scalar OP, which does not describe all possible scenarios for the evolution of the heterophase structure when approaching the PT point.

Indeed, the anisotropy of the long-range elastic field upon sufficient approach to PT point, can lead to the formation of correlated structures of nuclei of the solid-state nematic type [38] (this is apparently the origin of large-scale „columns“ of α-phases responsible for the appearance of abnormal small-angle light scattering during $\beta-\alpha$ PT in quartz [9]) or regular heterophase superstructures [39] (observed during PT in crystals Hg_2Cl_2 [40], $BaTiO_3$ [41,42] and DKDP [43]). Besides, in the model used above it was implicitly assumed that dislocations do not change their shape, are not displaced, and are not generated during PT. That is, actually PTs were considered either in brittle materials with high Peierls barriers or in materials with locked dislocations (for example, pinned by the defect atmospheres of Cottrell, Snook and Suzuki). In plastic materials that experience PT of the 1st kind, the initial DE can lose stability and transform into the vicinity of the PT [44–47].

Analysis of such situations, as well as of specific effects caused by the polarization properties of scattered particles or light, scattering geometry, spatial inhomogeneity of DE, etc., requires special consideration for specific materials using more complex models with multicomponent OP and its close connection with data from relevant experiments.

Conflict of interest

The author declares that he has no conflict of interest.

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