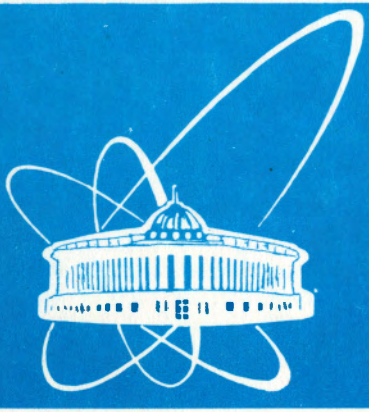

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KINETIC THEORY OF PHASE TRANSITIONS WITH MECHANISM OF AN ORDER-DISORDER IN FERROELECTRICS

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Кинетическая теория фазовых переходов
с механизмом порядок–беспорядок в сегнетоэлектриках

Исследованы кинетические уравнения в виде обобщенного уравнения Ландау–Халатникова вблизи фазового перехода. Определены температурная и частотная зависимости динамической восприимчивости. Обсуждена проблема центрального пика.

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Kinetic Theory of Phase Transitions
with Mechanism of an Order–Disorder in Ferroelectrics

The paper presents the investigations of kinetic equations introduced as generalization of Landau–Halatnikov equation near the phase transition. Temperature and frequency dependencies of dynamical susceptibility have been determined. The problem of central peak of the crystals with the order–disorder mechanism of phase transition has been discussed.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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1. INTRODUCTION

The phenomenological kinetic theory of the phase transitions with mechanism of an order- disorder has been developed in the present research.

In the terms of Landau's inverse problem [1], the phenomenological theory of the phase transitions in the crystals with transition mechanism of an order- disorder may be formulated by the following way. Suppose that the space groups of symmetry of high symmetry G_0 and low symmetry G_1 phases are known and G_1 is the subgroup of G_0 group. Then the matrix group L, defining the transformational properties of the order parameter, may be determined. The basic confirmation is that in the crystals with the order-disorder mechanism of the phase transition the major order parameter is transformed by L matrix group and has the meaning of probability [2]. The connection of the major order parameter with the secondary one in accordance with their physical meanings may be determined from the symmetrical arguments. Such symmetrical consideration provides the consistent composition of the thermodynamical potential, which knowledge allows to describe the static and kinetic properties of the system near the critical point [1, 2, 3].

In the theoretical description of the phase transition in the crystals with the order-disorder mechanism the physical parameters as polarization, displacement and so on are always the secondary order parameters. It is the essential difference from the crystals with the transitions of displacement type. The possible changes of the symmetry in the system are definitively determined by the evident form of the matrix group L. However, besides this, L group also determines the transformational properties of the order parameter. The situation when the major and secondary order parameters have the same transformational properties is possible. In an investigation of the kinetic properties of the system the roles of major and secondary order parameters are marked. At the theoretical consideration of the dynamical processes in the crystals with the order-disorder transition mechanism it is necessary to take into account that the space description of the equalprobable atoms' states may have a complex structure. Obviously at that, considering the symmetry of the structure we always have in mind the average states of the atoms and it is not essential the averaging has been made over the discrete or continuous aggregations of the states. Besides, as the average value of the order parameter over the crystal volume represents a linear combination of the coordinates of the system particles in any real system the movements corresponding one freedom degree are connected with the movements corresponding all others [3].

When the defects are presented in the system we may expect that in the systems with the order-disorder mechanism of the phase transitions an evolution of the disorder parameter will take place with the losses, meaning that not all states of the system are realized. Besides, when the system is relaxed to the equilibrium state the presence of the structure inhomogeneities produces the change of the order parameter by means the smoothing or the acquiring of the additional states. In such situation for the transitions with order-disorder mechanism it seems characteristic that the evolution process of the order parameter is accompanied by the nonlocal processes. So a generalization of the dynamical equation for the order parameter by means introduction of the nonlocal function is seemed to be necessary.

2. DYNAMICAL EQUATIONS FOR THE ORDER PARAMETER IN LANDAU—HALATNIKOV FORMALISM

Since in the phase transition of the crystals with the order-disorder mechanism the order parameter defines the change of symmetry and has the meaning of probability, so in such approach the total number of equalprobable states in the structure is arbitrary in principle. However for the reduction of symmetry it is necessary that a narrowing of the set of the equalprobable states would take place as a result of the phase transition. During the relaxation of the order parameter an indefinitivity of the equilibrium state option connecting with the presence of a set of the stationary states may induce the nonlocal processes due to the structure inhomogeneities. Introduce the generalized Landau-Halatnikov equation in the following form :

$$\gamma\eta(t) = -\int_0^t K(t-\tau)\frac{\delta g[\eta,P]}{\delta\eta}d\tau,$$

$$0 = \frac{\delta g[\eta,P]}{\delta P}. \qquad (1)$$

If the order parameter η and the polarization P have the same transformational properties in the thermodynamical potential $g[\eta,P]$, the term describing the interaction of the lowest order will be the linear function of both the order parameter η and the polarization P. In this case [2] in accordance with the symmetrical arguments, the thermodynamical potential is presented by the expression:

$$g = \frac{1}{2}\alpha\eta^2 + \frac{\beta}{4}\eta^4 + \frac{\omega_0^2}{2}P^2 - \xi\eta P - PE, \qquad (2)$$

where $\alpha = \alpha_0(T - T_0)$, E – the external electric field. At $K(t-\tau) \equiv 1$ by means the simple differentiation it may be shown that the equation (1) coincides with Landau-Halatnikov equation (2). If $K(t-\tau) = \delta(t-\tau)$ the equation (1) takes the form:

$$\gamma\eta = -\alpha\eta + \beta\eta^3 - \xi P,$$
$$\omega_0^2 P - \xi\eta - E = 0. \qquad (3)$$

It is followed that due the renormalization of the coefficient of a square invariant the shift of the critical temperature $T_c = T_0 + \gamma/\alpha_0$ takes place. In this case the equation (3) determines the equilibrium state of the system with the critical temperature T_c.

Therefore the introduction of $K(\tau)$ function characterizes the influence of disorder causing the change of the generalized force $\delta g[\eta,P,...]/\delta\eta$, which action produces the dynamics of the order parameter. Physically, the nonlocal function $K(\tau)$ describes the influence of the nonhomogeneities of the potential energy caused by

the presence of the structure defects, on the relaxation of the order parameter to the equilibrium state. At the definite distributions of the structure defects the potential energy can contain a set of the shallow minimums corresponding the metastable phases. So if the system initially is in the definite state then before the coming to the final state, it accomplishes the series of the transitions between the neighboring minimums. The set of the shallow minimums of the potential energy is induced by the geometry of the fractal distribution of the defects what is closely connected with the presence of the complex interaction in the structure.

The classification of $K(\tau)$ curves may be made by Hausdorf-Bezikovich dimension of d_c [4]. Further consider the influence $K(\tau)$ with $d_c < 1$, $d_c = 1$ and $d_c > 1$ on the process of relaxation of the order parameter to the equilibrium state near the phase transition in the crystals with the order- disorder mechanism.

Consider the intergro-differential equation (1) describing an evolution of the structure transferred by leap to the nonequilibrium state and define the nonlocal function $K(\tau)$ in Cantor's fractal set [4].

$$K(t-\tau) = \frac{1}{t(2\xi)^N}\sum_{m=1}^{2^N}\Theta(t_m^{(N)} < t < t_{m+1}^{(N)}). \qquad (4)$$

Here $\xi(\xi < 1/2)$ is the parameter of selfsimilarity, $t_m^{(N)}$ are the points' coordinates, $1/(t(2\xi)^N)$ is the density of the remained states at N-stage of the construction of Cantor's set, $1/(t(2\xi)^N)$ is the step function.

Using substitution $f(t) = \delta\Phi/\delta\varphi$ one can get

$$J(t) = \int_0^t K(t-\tau)f(\tau)d\tau \qquad (5)$$

and applying Laplas transform we obtain:

$$J(t)_\bullet =^\bullet \Phi(p) = \frac{F(p)}{\xi^N pt}[1 - \exp(-pt\xi^N)]Q_N(z), \qquad (6)$$

where the following designations have been introduced:

$$Q_N(z) = \frac{1}{2^N}\prod_{n=0}^{N-1}(1 + \exp(-z\xi^N)), \qquad z = pt(1-\xi). \qquad (7)$$

The last expression allows to obtain for $Q_N(z)$ the following recurrent relationship

$$Q_N(\frac{z}{\xi}) = \frac{1}{2}(1 + \exp(-\frac{z}{\xi}))Q_{N-1}(z). \qquad (8)$$

In the stationary point:

$$\Phi^*(\frac{z}{\xi}) = \frac{1}{2}\Phi^*(z), \qquad (9)$$

which solution has the form:

$$\Phi^*(z) = A_\nu z^{-\nu} \qquad (10)$$

where $\nu = \ln 2/\ln(1/\xi)$ is the dimension of Cantor's fractal set. Consequently,

$$\Phi(p) = A_\nu (pt)^{-\nu}(1-\xi)^{-\nu}F(p). \tag{11}$$

The representation of $K(p)$ function has the form:

$$K(p) = A_\nu (pt)^{-\nu}(1-\xi)^{-\nu}. \tag{12}$$

With a help of Mellin's transformation we it is found

$$K(t-\tau) = \frac{A_\nu}{\Gamma(\nu)}(1-\xi)^{-\nu}t^{-\nu}, \tag{13}$$

where $\Gamma(\nu)$ is Eiler's gamma function. Therefore, $J(t)$ function is defined in the following form

$$J(t) = A_\nu(1-\xi)^{-\nu}t^{-\nu}\frac{1}{\Gamma(\nu)}\int_0^t (t-\tau)^{\nu-1}f(\tau)d\tau = A_\nu(1-\xi)^{-\nu}t^{-\nu}D^{-\nu}f. \tag{14}$$

Introducing the nondimensional variable $u = \tau/t$ with the help of the commutative properties of the fractional derivative, eq. (1) may be presented as

$$\begin{cases} \frac{\chi}{t}\frac{\partial^\nu \eta(t)}{\partial u^\nu} &= -B_\nu \frac{\delta g[\eta,P]}{\delta \eta}, \\ \frac{\delta g[\eta,P]}{\delta P} &= 0 \end{cases} \tag{15}$$

The solution of the equations' system (15) will be found in the following form:

$$\begin{aligned} \eta(t) &= \eta_0 + \eta e^{i\omega t u}, \\ P(t) &= P_0 + P e^{i\omega t u}, \\ E &= E_0 e^{i\omega t u}. \end{aligned} \tag{16}$$

The static solutions of the system (15) are determined from the equations:

$$\begin{aligned} \omega_0^2 P_0 - \xi\eta_0 &= 0, \\ -\alpha\eta_0 - \beta\eta_0^3 + \xi P_0 &= 0. \end{aligned} \tag{17}$$

After the linearization of the eqs. (15) close to the solutions (17) it is obtained for η and P fields, correspondingly:

$$\eta = \frac{1}{\xi}(\omega_0^2 P - E)$$

$$\eta = \frac{B_\nu P \xi}{\gamma(i\omega t)^\nu/t + B_\nu\alpha + 3B_\nu\beta\eta_0^2}. \tag{18}$$

The dynamical susceptibility $\chi = \lim_{E\to 0}\partial P/\partial E$ defining the system's response on the external electric field is presented by expression

$$\chi(\omega,T) = \left(\omega_0^2 - \frac{\xi^2 B_\nu}{\gamma(i\omega t)^\nu/t + B_\nu\alpha + 3B_\nu\beta\eta^2}\right)^{-1} \tag{19}$$

At $\omega \to 0$ $\chi(0,T)$, coincides with the result (2) for the static dielectric constant.

In the paraphase $\eta_0^2 = 0$ and consequently

$$\chi_{para}(\omega,T) = \frac{1}{\omega_0^2}\frac{\gamma(i\omega t)^\nu/t + B_\nu\alpha_0(T-T_0)}{\gamma(i\omega t)^\nu/t + B_\nu\alpha_0(T-T_c)} \tag{20}$$

In the ferroelectric phase

$$\chi_{fer}(\omega,T) = \frac{1}{\omega_0^2}\frac{\gamma(i\omega t)^\nu/(B_\nu t) + \alpha_0(T-T_0) - 3\alpha_0(T-T_c)}{\gamma(i\omega t)^\nu/(B_\nu t) - 2\alpha_0(T-T_c)}, \tag{21}$$

where $T_c = T_0 + \xi^2/\omega_0^2$.

Consider the case $d_c = 1$, when a smearing of $K(t) = \delta(t)$ function to a bell-like dependence defined by the exponential function with the width being specified by the time interval τ_0, occurs. Therefore $K(t-\tau) = \exp(-b(t-\tau)/\tau_0)$ and suppose that the characteristic time interval $\tau_0 = 1$. In this case the kinetic equation is written in the form:

$$\gamma\eta(t) = -\int_0^t \exp(-b(t-\tau))\frac{\delta g[\eta,P]}{\delta\eta}d\tau$$

$$0 = \frac{\delta g[\eta,P]}{\delta P}. \tag{22}$$

Using the definition of the fractional derivative

$$D^\nu f(t) = \frac{1}{\Gamma(-\nu)}\int_0^t (t-\tau)^{1-\nu}f(\tau)d\tau, \tag{23}$$

where $-\infty < \nu < 0$, and the expansion of the exponential function into Tailor's series, we may write

$$\gamma(b + \frac{\partial}{\partial t})\eta(t) = -\frac{\delta g[\eta,P]}{\delta\eta}$$

$$0 = \frac{\delta g[\eta,P]}{\delta P} \tag{24}$$

As $g[\eta,P]$ involves the square invariant on $g[\eta,P]$ with coefficient $\alpha = \alpha_0(T-T_0)$, so the system of eqs. (24) may be written in the form:

$$\gamma\frac{\partial}{\partial t}\eta(t) = -\frac{\delta g[\eta,P]}{\delta\eta}$$

$$0 = \frac{\delta g[\eta,P]}{\delta P} \tag{25}$$

where the critical temperature is defined as

$$T_c^0 = T_0 - \frac{b\gamma}{\alpha_0}.$$

The solution of eqs'. system (25) will be sought in the form (16). The static solutions of the system (25) are defined from eqs.

$$\alpha_0(T-T_c^0)\eta_0+\beta\eta_0^3-\xi P_0=0$$

$$\omega_0^2 P_0-\xi\eta_0=0. \tag{26}$$

After the linearization of the eqs. (25) near the static solutions for η and P fields, it is obtained, consequently:

$$\eta=\frac{1}{\xi}(\omega_0^2 P-E)$$

$$\eta=\frac{\xi P}{i\gamma\omega+\alpha_0(T-T_c^0)+3\beta\eta_0^2}. \tag{27}$$

The dynamical susceptibility $\chi=\chi(\omega)$ for the system with an exponential memory is presented by expression

$$\chi(\omega)=\frac{1}{\omega_0^2}\frac{i\gamma\omega+\alpha_0(T-T_c^0)+3\beta\eta_0^2}{i\gamma\omega+\alpha_0(T-T_c')+3\beta\eta_0^2}, \tag{28}$$

where

$$T_c'=T_c^0+\frac{\xi^2}{\omega_0^2}=T_0-\frac{b\gamma}{\alpha_0}+\frac{\xi^2}{\omega_0^2}.$$

At $\omega\to 0$ and $b\to 0$, $\chi(0,T)$ coincides with the result [2] for the static dynamical permeability.

In the paraelectric phase $\eta_0^2=0$ and hence the dynamical susceptibility takes the form

$$\chi_{para}(\omega)=\frac{1}{\omega_0^2}\frac{i\gamma\omega+\alpha_0(T-T_c^0)}{i\gamma\omega+\alpha_0(T-T_c')}. \tag{29}$$

In the ferroelectric phase $\eta_0^2=(\alpha_0/\beta)(T-T_c')$ and hence the dynamical susceptibility is defined as:

$$\chi_{fer}(\omega)=\frac{1}{\omega_0^2}\frac{i\gamma\omega+\alpha_0(T-T_c^0)+3\alpha_0(T-T_c')}{i\gamma\omega-2\alpha_0(T-T_c')}. \tag{30}$$

Consider integro-differential equation (1) where the nonlocal function $K(\tau)$ has been specified as the fractal curve with dimension $1<d_c<2$. An example of the continuous but nowhere differentiable curve of $d_c>1$ dimension is Weershtrasse function. However, consider further the curve $\Psi(t)$ closely connecting with Weershtrasse function in the form

$$K(t-\tau)=\Psi(t-\tau)=\frac{1-a}{a}\sum_{n=0}^{\infty}a^n b^n\exp(-b^n(t-\tau)), \tag{31}$$

where $a<1$, $b>a$.

It is not difficult to show that $\Psi(t)$ function at high temperatures is the homogeneous function. Indeed,

$$ab\Psi(bt)=\Psi(t)-(1-a)b\exp(-bt),$$

and it follows, that

$$\Psi(t)=ab\Psi(bt)$$

Using the definition of the fractional derivative (2), it may be shown that in the case $K(\tau)\equiv\Psi(\tau)$ eq. (1) is put down in the form

$$\gamma\eta(t)=-\frac{1-a}{a}\sum_{n=0}^{\infty}\frac{a^n}{1+D/b^n}\frac{\delta g[\eta,P,]}{\delta\eta}$$

$$0=\frac{\delta g[\eta,P]}{\delta P}. \tag{32}$$

In this case the shift of the critical temperature $T_c''=T_0-a\gamma/(\alpha_0(1-a))$ takes place. The eq. (32) may be presented in the form:

$$\ldots+\gamma_4\frac{\partial^4}{\partial t^4}\eta(t)+\gamma_3\frac{\partial^3}{\partial t^3}\eta(t)+\gamma_2\frac{\partial^2}{\partial t^2}\eta(t)+\gamma_2\frac{\partial^2}{\partial t^2}\eta(t)=-\frac{\delta g[\eta,P]}{\delta P}$$

$$0=\frac{\delta g[\eta,P]}{\delta P}, \tag{33}$$

where γ_i coefficients are $1/b^i$ order infinitesimal.

The solution of the eqs . system (33) will be sought in the form (16). The static eqs. of the system (33) are determined from eq. (26), where $T_c^0\equiv T_c''$. After the linearization of eqs. (33) near the static solutions for η and P fields it is obtained

$$\eta=\frac{1}{\xi}(\omega_0^2 P-E)$$

$$\sum_{n=1}^{\infty}\gamma_n(i\omega)^n\eta=-\alpha\eta-3\beta\eta_0^2\eta+\xi P. \tag{34}$$

In this case, using (34) and the definition of the dynamical susceptibility it is found

$$\chi(\omega)=\left(\omega_0^2-\frac{\xi^2}{\alpha+3\beta\eta_0^2+\sum_{n=1}^{\infty}\gamma_n(i\omega)^n}\right)^{-1}. \tag{35}$$

Taking account of $\eta_0=P_0=0$ for in the paraelectric phase $\chi=\chi(\omega)$, it is obtained

$$\chi_{para}(\omega)=\left(\omega_0^2-\frac{\xi^2}{\alpha+\sum_{n=1}^{\infty}\gamma_n(i\omega)^n}\right)^{-1}. \tag{36}$$

In the ferroelectric phase

$$\eta_0^2=\frac{\alpha_0}{\beta}(T-T_c''),$$

where

$$T_c'''=T_0-\frac{a}{\alpha_0(1-a)}\gamma+\frac{\xi^2}{\omega_0^2},$$

For the dynamical susceptibility it is obtained:

$$\chi_{fer}(\omega) = \frac{1}{\omega_0^2} \frac{\alpha_0(T - T_c') + 3\alpha_0(T - T_c''') + \sum_{n=1}^{\infty} \gamma_n (i\omega)^n}{-2\alpha_0(T - T_c''') + \sum_{n=1}^{\infty} \gamma_n (i\omega)^n}. \quad (37)$$

Consider the intrinsic ferroelectrics describing by the thermodynamical potential in the form:

$$g = \frac{\alpha}{2} P^2 + \frac{\beta}{4} P^4 - PE \quad (38)$$

In this case it may be shown that the kinetic eq. with the nonlocal function of $d_c < 1$ dimension in Landau- Halatnikov formalism is written in the form:

$$\frac{\gamma}{t} \frac{\partial^\nu}{\partial u^\nu} P = -B_\nu \frac{\delta g[\eta, P]}{\delta P}. \quad (39)$$

An equilibrium value of the spontaneous polarization P_0 is determined by the expression

$$\alpha P_0 + \beta P_0^3 = 0. \quad (40)$$

Correspondingly, for the polarization P it is obtained

$$P = \frac{E}{\alpha + 3\beta P_0^2 + \frac{\gamma}{tB_\nu}(i\omega t)^\nu}. \quad (41)$$

Using the definition of the dynamical susceptibility it is found

$$\chi(\omega) = \left(\alpha + 3\beta P_0^2 + \frac{\gamma}{tB_\nu}(i\omega t)^\nu\right)^{-1}. \quad (42)$$

Taking account of the equilibrium value P_0 in the high- and low- symmetrical phases for the dynamical susceptibility, it is obtained

$$\chi_{para}(\omega) = \left(\alpha + \frac{\gamma}{tB_\nu}(i\omega t)^\nu\right)^{-1}, \quad (43)$$

$$\chi_{para}(\omega) = \left(-2\alpha + \frac{\gamma}{tB_\nu}(i\omega t)^\nu\right)^{-1}. \quad (44)$$

At $\nu \to 1$ ($B_\nu \to 1$) the derived result coincides with the conclusion of Landau theory for the intrinsic ferroelectrics.

3. THE CENTRAL PEAK IN THE DYNAMICAL FORM-FACTOR IN THE CRYSTALS WITH THE ORDER-DISORDER MECHANISM OF TRANSITION

Within the scope of Landau theory the change of symmetry is defined by the major order parameter. At the dynamical consideration the presence of the remaining freedom degrees composes the medium characterized by the kinetic coefficient γ, where the relaxation of the major order parameter takes place.

Physically the dynamical form-factor is proportional to the spectral density of the fluctuations of the major and secondary order parameters, the latter takes part in the scattering process, i.e. interact with the radiation falling on the crystal. In the quantitative aspect the dynamical form-factor defines the frequency dependence of the intensity and the energy spectrum of neutrons scattered by the crystal [8, 9].

The relation between the dynamical form-factor and the dynamical susceptibility is determined by the fluctuation- dissipative theorem

$$S(\omega) \sim \frac{T}{\omega} Im\chi(\omega). \quad (45)$$

The analysis shows that the dynamical form-factor involves the sharply increasing maximum at $\omega \to 0$ and $T \to T_c$ in $S(\omega \to 0) \sim (T - T_c)^{-1}$ form named as the central peak.

Hence the appearance of the central peak is connected with the appearance of the order parameter distinguished from zero. Besides, the analysis of (19) and (37) shows that at the finite frequencies the dynamical form-factor involves the additional maximums connected with the presence of the soft and nonlinear modes accompanying the phase transition. The appearance of the nonlinear modes is connected with the presence of the slight order in the structure and characterizes the particular features of the order parameters' relaxation in the structure with the defects.

4. DISCUSSION

Unlike the displacement transitions in the transitions with the order-disorder mechanism the major order parameter has the meaning of probability and the physical values as polarization, displacement and so on are the secondary order parameters. In this sense if even the transformational properties of the secondary order parameters coincide with the same for the major order parameter the transition has the pseudointrinsic nature. The features of the dynamics of the order parameter are connected with the presence of the set of the equalprobable states to which the order parameter may relax. In such systems the influence of the different distributions of the defects on the relaxation of the order parameter may induce the nonlocal processes. The generalization of Landau-Halatnikov eq. by means the introduction of the nonlocal function $K(\tau)$, allows to take into account an influence of the inhomogeneities of the potential energy caused by the presence of the defects in the structure. In such approach if not all of the states are realized during the relaxation of the order parameter, the nonlocal function may be determined in Cantor's fractal set. In this case the dynamical equation is the equation of the fractional derivatives. The response of the system on the external disturbance is defined by formula (19) and the system has the property of a glass. If the system displays the exponential memory, the dynamical equations have Markov's form. However, it is necessary to take into account that in this case the presence of the defects shifts the critical temperature to the low temperature side. If the system during the relaxation process acquires the additional states the dynamical equations involve the time derivatives of the arbitrary high order. In this case the dynamical susceptibility involves the

additional maximums depending on the temperature what indicates on the presence of the nonlocal modes accompanying the phase transition. In the case of the intrinsic ferroelectrical transitions where the principle parameter is the polarization, the dynamical susceptibility is determined by expression (42). The difference between the relaxation processes in the intrinsic and pseudointrinsic transitions is expressed by the corresponding response functions defined by eqs. (42) and (19), correspondingly.

So the account of the influence of the slight disorder (the latter means that due the presence of such disorder the time independent solutions coincide with the solutions corresponding the ideal crystal) changes the relief of the potential energy of the system and essentially alters the physical properties of the system. The knowledge of the imaginary part allows to determine the frequency dependence of the dynamical form-factor. Depending on the nature of the defects' distribution in the structure the dynamical form-factor of the crystal apart from the central peak involves the additional maximums depending on the temperature what indicates the presence of the soft nonlinear modes accompanying the phase transition.

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