HETEROGENEOUS STATES WITH FERROELECTRIC SOLITON-FLUCTUONS

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This paper considers a Hamiltonian of crystal fluctuations as a functional of three kinds of fields, i.e. ordering field, a field corresponding to the distribution of solitons and a field connected with the distribution of coupled electron-fluctuon systems. A thermodynamic potential of soliton-fluctuon system was analysed and the fluctuation theory of this system was developed elsewhere. The present paper analyses basic kinetic equations describing the process of domain wall motion within the framework of the given model.

НЕОДНОРОДНЫЕ СОСТОЯНИЯ С ФЕРРОЭЛЕКТРИЧЕСКИМИ ФЛУКТУОНАМИ СОЛИТОННОГО ТИПА

В работе рассматривается гамильтониан флуктуаций в кристалле как функционал трех типов полей: упорядочивающего поля, поля, соответствующего распределению солитонов и поля, связанного с распределением спаренных электронов и флуктуонов. Проведен анализ термодинамического потенциала системы солитонов и флуктуонов и сформулирована теория флуктуаций этой системы. Кроме того, в рамках данной модели изучаются основные кинетические уравнения, описывающие движение границы домена.

I. INTRODUCTION

Soliton and fluctuation formations in ordered systems have recently become and object of thorough investigations. The behaviour of solitons in ferromagnetics was analysed in [1—3] and that of fluctuons and phasons in ferromagnetics and ferroelectrics was considered in [4—6].

It is known [7] that for any real ferroelectric the polar state is changed for a nonpolar one near the phases transition point T by means of the formation of new phase germs, but not within the whole volume simultaneously. New phase regions appear stochastically. This appearance of an energetically preferable phase does not lead to a complete phase change (temperature hysteresis is $\Delta T \sim 1 \div 2$ K). The

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heterogeneous structure of ferroelectric crystals is stable in the hysteresis region, this assumption being used, e.g., in the diffuse phase transition theory [8].

According to the thermodynamic theory of germ formation at a phase transition their increase is possible only when a certain critical size is exceeded [9]. It is also known that the dependence of the domain wall motion rate on the external field coincides with the dependence of the thermal wall motion on $|T - T_0|$ [10].

We consider the change of structure from a less preferable to a more preferable one as a kinetic phase transition, since its major properties are connected with the dependence on the time structure rearrangement process [11]. A phase transition is connected with strongly diffused fluctuations of the form and position of a boundary between two structures, causing wall motion. The fluctuation theory of phase transitions [12] allows to describe a system having a phase transition by means of the multi-component ordering field $\varphi(r)$.

Its expansion in the Fourier series

$$\varphi(r) = \sum_q \varphi_q e^{iqr} \qquad (1)$$

does not include short-wave harmonics $q = |q| < q_0$. This paper analyses, along with the ordering field $\varphi(r)$, the field $\varphi^s(r)$, which corresponds to a definite distribution of stationary (not moving) solitons in a crystal, and the field $\varphi'(r)$ corresponding to the definite distribution of electrons which form coupled states, i.e. fluctons. We consider the crystal to be broken into a number of small regions. In the case of a given region we have $\varphi^s(r) = 1$, otherwise we have $\varphi^s(r) = 0$. Summing up fields $\varphi(r)$, $\varphi^s(r)$ and $\varphi'(r)$ in a statistical sum with respect to all degrees of freedom we may determine the thermodynamic potential of a system, i.e. the "fluctuation Hamiltonian", which is the functional $H(\varphi, \varphi^s, \varphi')$. In case of zero fields φ^s and φ' we can express $H(\varphi)$ as [9]:

$$H = \int \left[\frac{1}{2} \xi(\nabla\varphi)^2 + \Phi(\varphi) \right] dr, \qquad (2)$$

where $\Phi(\varphi)$ is the nonlinear functional of φ. In the case of $H = H(\varphi, \varphi^s, \varphi')$, we can write

$$H = \int \left[\frac{1}{2} \xi_1(\nabla\varphi')^2 + \frac{1}{2} \xi_2 |\nabla\Psi|^2 n_e + \frac{1}{2} \xi_3 \Psi\varphi n_e + \Phi(\varphi') \right] dr, \qquad (3)$$

where $\varphi' = \varphi + \varphi'$, Ψ is a wave function of electrons, n_e is electron density. The equilibrium meaning φ' and Ψ can be found by linearization of the functional [6]

$$H = H(\varphi, \varphi^s, \varphi') = H(\varphi, \varphi^s, \Psi). \qquad (4)$$

According to [13] the rate of change is proportional to the conjugated ther-

modynamic strength at a small deviation from equilibrium. Assuming the deviation of φ to be small, equation [12] can be written

$$\varphi' = -\lambda \frac{\delta H}{\delta \varphi}, \qquad (5)$$

where λ is a kinetic coefficient. It should be stressed that of the three components of the pseudospin $\sigma = (\sigma^x, \sigma^y, \sigma^z)$ only σ^z characterizing polarization (in our model it describes an ordering field), φ is different from zero. Thus the Landau–Lifshits equation is reduced to equation (5). The components of the pseudospin, following [14] can be presented as

$$\sigma_i^x = [a_s^i a_s^i - a_a^i a_a^i], \quad \sigma_i^y = i[a_s^i a_a^i - a_a^i a_s^i], \quad \sigma_i^z = [a_s^i a_s^i + a_a^i a_a^i],$$

where a_s^i, a_a^i are Fermi or Bose operators of the formation and annihilation for protons and neutrons (i-position, quantum state S or a).

II. FLUCTUATION THEORY OF A SYSTEM WITH SOLITON-FLUCTONS

In the present paper we are not going to consider a domain wall as a continuous medium and investigate thoroughly its structure. Instead we shall analyse the switch-in function $\varphi(r)$ determined on scales $R \gg h$ which equals unity, provided the r point gets into the volume of an energetically more preferable structure, and equals zero otherwise. Analogously we determine the switch-in functions $\varphi^s(r)$ and $\varphi'(r)$ equal to unity if an electron and soliton, respectively, are present in the elementary cell (the Känzig region) [7] and equal to zero if these are not present there.

A microscopically well-founded assumption could be made that the depolarization of an elementary region leads to the disappearance of a soliton there with a corresponding release of its energy into a system. Now in the limits of the proposed model we are going to describe the interaction between elementary regions. The total number of regions is assumed to be N, n of them being in an energetically more preferable phase. Evidently both the state of a system and the kinetics of a phase transition are characterized completely by the function

$x = \dfrac{n}{N}(T)$. The expression for the full number of ways of distributions of n regions with the total number N, taking into account that there are n^s solitons and n^e electrons in the system can be written as

$$P = \frac{N!}{n! \, n^s! \, (N - n - n^s)!} \left(N \frac{dx}{dT} \right)^{n^e}. \qquad (6)$$

Expression (6) assumes the number of coupled states (fluctuons) and, respectively, places for the distribution of electrons to be directly proportional to the derivative dx/dT, which characterizes the number of order inhomogeneities in a system [15]. We designate $x^s = n^s/N^s$, $x^e = n^e$, where N^s is the initial number of solitons in a system. Expression (6) allows to obtain system entropy by means of the Boltzmann formula:

$$S(x, x^s, x^e) = -kN \left[x \ln x + x^s \ln x^s + (1 - x - x^s) \ln (1 - x - x^s) - x^e \ln N - x^e \ln \left(\frac{dx}{dT} \right) \right].$$ (7)

where k is the Boltzmann constant.
The internal energy of the system under consideration equals

$$U = N\Delta\mu x + N\Delta\mu^s x^s + N\Delta\mu^{es} x^s x + N\Delta\mu^e x^e x + N\Delta\mu^e x^e$$ (8)

and the free energy for one region is

$$F = \frac{U}{N} - \frac{TS}{N} = \Delta\mu x + \Delta\mu^s x^s + \Delta\mu^{es} x^s x + \Delta\mu^e x^e x +$$
$$+ \Delta\mu^{es} x^s x^s x + kT \left[x \ln x + x^s \ln x^s + \right.$$
$$\left. + (1 - x - x^s) \ln (1 - x - x^s) - x^e \ln \left(\frac{dx}{dT} \right) - x^e \ln N \right].$$ (9)

In expressions (8)—(9):

$$\Delta\mu = -2P_s E + O(E^3), \quad \Delta\mu^e = \frac{\hbar^2}{2\mu} \int \int |\nabla\Psi(r)|^2 dr,$$

$$\Delta\mu^e = 2g \int P_s \Psi(r) dr, \quad \Delta\mu^s = \frac{16S}{n_0} \text{th} \left(\frac{n}{n_0} \right) P_s,$$ (10)

$$\Delta\mu^{es} = 2g \int P_s (f(r) - 1) \Psi(r) dr,$$

where P_s is the polarization, E is the external field, μ is the electron effective mass, g is the coupling constant, $n_0 = 4\sqrt{\delta}/\alpha$ and δ are constants of the microscopic Hamiltonian [3], $f(r)$ is the soliton distribution function. It should be noted that our model neglects the possibility of the appearance of two electrons or two solitons in one Känzig region; this assumption is quite justified as their concentration is small. The attempt to take into account the interaction of elementary regions leads to the internal energy of the system being a concentration function of x. We expand this function in the series

$$U = N \left[\Delta\mu x + \Delta\mu^s x^s + \Delta\mu^e x^e + \Delta\mu^e x^e x + \Delta\mu^s x^s + \right.$$
$$\left. + \Delta\mu^{es} x^s x - \frac{1}{2} \alpha x^2 + \frac{1}{3} \beta x^3 - \frac{1}{4} \gamma x^4 \right],$$ (11)

where $\alpha > \beta > \gamma > 0$ are corresponding coefficients of expansion. The number of electrons according to the assumption is constant, $n^e = $ const. and hence $x^e = $ const. According to the regions model (KRM) for diffuse phase transitions the phase transition point is determined by the condition $x = 1/2$. Since we assume that depolarization of an elementary region causes the disappearance of a soliton, it is reasonable to draw a conclusion that the phase transition point corresponds to

$$n^e = N^e/2, \quad x^e = \frac{1}{2} N^e/N.$$

Using (11) we may obtain for the free energy

$$F(x, x^s, x^e) = \Delta\mu^s x^e + [\Delta\mu + \Delta\mu^e x^e + \Delta\mu^e x^e +$$
$$+ \Delta\mu^{es} x^s] x - \frac{1}{2} \alpha x^2 + \frac{1}{3} \beta x^3 - \frac{1}{4} \gamma x^4 +$$
$$+ kT \left[x \ln x + x^s \ln x^s + (1 - x - x^s) \ln (1 - x - x^s) - \right.$$
$$\left. - x^e \ln \left(\frac{dx}{dT} \right) - x^e \ln N \right].$$ (12)

In the case of the first order phase transition two values of x_1 and x_2 should exist for which the free energy satisfies the conditions [16]:

$$F(x_1) = F(x_2), \quad F'(x_1) = F'(x_2) = 0,$$
$$F''(x_1) > 0, \quad F''(x_2) > 0.$$ (13)

Therefore we get for the phase transition point T_0

$$T_0 = \frac{1}{k \ln (N - N^s)} \left[\Delta\mu + \Delta\mu^e N^e + \frac{\Delta\mu^s N^s}{2} + \right.$$
$$\left. + \frac{\Delta\mu^{es} N^s N^s}{2} - \frac{\alpha}{2} + \frac{\beta - \gamma}{8} \right].$$ (14)

Now let us consider the second order phase transition at the phase transition point where should exist x_0 for which there holds

$$F'(x_0) = 0, \quad F''(x_0) = 0, \quad F'''(x_0) = 0, \quad F^{IV}(x_0) > 0,$$
$$F'\left(\frac{1}{2}\right) = -\alpha + \beta - \frac{3}{4}\gamma + \frac{4}{kT_0} = 0, \quad F'''\left(\frac{1}{2}\right) = 2\beta - 3\gamma = 0.$$ (15)

Expressions (15) determine the connection between the constants:

$$\beta = 2(\alpha - 4kT_0), \quad \gamma = \frac{4}{3}(\alpha - 4kT_0) = \frac{2}{3}\beta.$$ (16)

of only

It follows from (16) that the internal energy expansion may be expressed by means of only

$$U = N\left[\Delta\mu x + \Delta\mu^e x^e + \Delta\mu^{es} x^e x + \Delta\mu^s x^s x + \Delta\mu^{es} x^e x^s x - \frac{1}{2}\alpha x^2 + \frac{1}{3}(\alpha + 4kT_0)(2x^3 - x^4)\right].$$ (17)

From $F^{IV}\left(\frac{1}{2}\right) > 0$ we obtain $\frac{1}{kT_0} > \frac{8}{\alpha}$.

Now we analyse the system divided by a domain wall into two connected homogeneous regions with the thermodynamic potential density μ_0 and μ_1, respectively. Then, the relation $\Delta\mu = \mu_1 - \mu_0$ holds. This structure corresponds to the ordering the system with two thermodynamic potentials taking into account the ordering fields $\varphi(r)$, $\varphi^s(r)$, $\varphi^e(r)$:

$$H = \int [\mu_0 + \Delta\mu\varphi(r) + \Delta\mu^e\varphi^e(r) + \Delta\mu^e\varphi^e(r)\varphi^e(r) + \Delta\mu^s\varphi^s(r)\varphi(r) + \Delta\mu^{es}\varphi^e(r)\varphi^s(r)\varphi(r)]\,dr + \sigma S(\varphi,\varphi^e,\varphi^s),$$ (18)

where

$$\Delta\mu = -2P_sE, \quad \Delta\mu^e = \frac{h^2}{2\mu}|\nabla\Psi(r)|^2, \quad \Delta\mu^e = 2gP_s\Psi(r),$$

$$\Delta\mu^s = \frac{16S}{h_0}th\left(\frac{n}{n_0}\right)P_s^2, \quad \Delta\mu^{se} = 2gP_s(f(r) - 1)\Psi(r),$$

and $S(\varphi,\varphi^e,\varphi^s)$ is the surface area of phase division, σ is the surface energy density.

We need to formulate more precisely the lattice approach to the considered problem. We assume that the system volume is split into cubic cells with linear size $l(q_0 = \pi/l)$. Thus the complicated coordinates $j = (x, y, z)$ of the cell centres form a simple cubic cell. We regard the z-axis to be the direction of a wall motion and exclude the configuration in which this axis crosses the wall surface more than once (overhanging condition). This leads to the exclusion of the following configuration-s: $\varphi_{j'} = \varphi_{j''} = 0$, $\varphi_{j'} = 1$, $\varphi_{j''} = 0$, where the x and y coordinates are constant, and we can present the expression for energy as

$$\beta H(\varphi,\varphi^e,\varphi^s) = \alpha\sum_j\varphi_j + \alpha^e\sum_j\varphi_j^e + \alpha^e\sum_j\varphi_j^e + \alpha^s\sum_j\varphi_j^s + \alpha^{es}\sum_j\varphi_j^e\varphi_j^s\varphi_j + \frac{x}{2}\sum_{ij}a_{ij}\omega(\varphi_i,\varphi_j) + \frac{x^e}{2}\sum_{ij}a_{ij}\omega(\varphi_i,\varphi_j)\varphi_j^e + \Omega(\varphi,\varphi^e,\varphi^s),$$ (19)

Where $\beta = 1/kT$, $\alpha = \Delta\mu l^3\beta$, $\alpha^e = \Delta\mu^e l^3\beta$, $\alpha' = \Delta\mu' l^3\beta$, $\alpha'' = \Delta\mu^s l^3\beta$, $x = \beta\sigma l^2$, $x^e = x\Delta\mu^e$, and $\Omega(\varphi,\varphi^e,\varphi^s)$ turns into infinity for forbidden configurations, the values a_{ij} are equal to unity for the nearest neighbours and equal to zero for other cases

$$\omega(\varphi_i,\varphi_j) = \begin{cases} 0, & \varphi_i = \varphi_j \\ 1, & \varphi_i \neq \varphi_j \end{cases}, \quad \alpha = \Delta\mu l^3\beta, \quad x^e = x\Delta\mu^e, \quad \alpha^e = \Delta\mu l^3\beta.$$ (20)

The existence of Ω in (19) leads to the realization of only those system configurations where for any x and y in the z-axis direction the parameters φ_i equal unity for crossing the domain wall surface and zero after its crossing. This in its turn leads to $\alpha^e = 0$, since fluctuation states are possible only when there are inhomogeneities in a system. The local position of the domain wall may be therefore described by

$$L_{x,y} = \sum_z\varphi_i,$$ (21)

where $v_{x,y}$ adopts whole values. To calculate the kinetics of the process we have to analyse the distribution function $\varrho(\varphi,t)$. The medium position of the domain wall equals in this case

$$L = \langle L_{x,y}\rangle = l\sum_\varphi\varrho(\varphi,t)v_{x,y}.$$ (22)

We assign the wall thickness and the wall motion rate as follows

$$v = \frac{dL}{dt}, \quad h = 2[\langle L_{x,y}^2\rangle - \langle L_{x,y}\rangle^2]^{1/2}.$$ (23)

III. KINETIC EQUATION

We describe the wall motion as a random sequence of φ_j, φ_j^e, φ_j^s changes. The distribution function $\varrho(\varphi,t)$ giving the probability to reveal the assigned configuration at time t is described by equation [17]:

$$\tau\frac{d\varrho(\varphi,t)}{dt} = \sum_j\left[\sum_{\varphi_j}f(\varphi_j,\varphi_j^s,\varphi_j^e)\varrho(\varphi,t) - \varrho(\varphi,t)\right],$$ (24)

where

$$f(\varphi_j,\varphi_j^s,\varphi_j^e) = \exp(-\beta H(\varphi,\varphi^s,\varphi^e))\left[\sum_{\varphi_j}\exp(-\beta H(\varphi_j,\varphi_j^s,\varphi_j^e))\right]^{-1},$$ (25)

$$\varrho^{(1)}(\varphi_j) = C_n(\varphi),$$ (26)

and $\varphi \equiv \varphi_i$. It follows from the normalization conditions that $C_n(0) = 1 - C_n(1)$.
A two-particle distribution function in the plane (x, y) reads

$$\varrho^{(2)}(\varphi_i, \varphi_i) = B_n(\varphi, \varphi'),$$ (27)

whence we obtain

$$B_n(1, 0) = B_n(0, 1) = C_n(1) - B_n(1, 0),$$
$$B_n(0, 0) = 1 + B_n(1, 1) - 2C_n(1).$$ (28)

A set of equations for $C_n(1)$ and $B_n(1, 1)$ has the form

$$\tau \frac{dC_n(1)}{dt} = (C_n(1) - C_{n+1}(1))(G(1) - 1) + (C_{n-1}(1) - C_n(2))G(0),$$

$$\tau \frac{dB_n(1,1)}{dt} = 2\left[(C_n(1) - C_{n+1}(1))\left(F(1) - \frac{B_n(1,1)}{C_n(1)}\right) + (C_{n-1}(1) - C_n(1))F(0) \right].$$ (29)

where

$$G(\varphi) = \sum_{m=0}^{4} \binom{4}{m} f_m \left[\frac{B_n(\varphi, 1)}{C_n(\varphi)} \right]^m \left[\frac{B_n(\varphi, 0)}{C_n(\varphi)} \right]^{4-m},$$

$$F(\varphi) = \sum_{m=0}^{3} \binom{3}{m} f_{m+1} \left[\frac{B_n(\varphi, 1)}{C_n(\varphi)} \right]^{m+1} \left[\frac{B_n(\varphi, 0)}{C_n(\varphi)} \right]^{3-m},$$

$$f_m = [\exp(-(\alpha + \alpha' n_\delta^s + \alpha'' n_\delta^s n_\delta^s) - 2(x + x'(N' - n'))(m-2)) + 1]^{-1},$$ (30)

$$n_\delta^s = N' \frac{\sum_n (1 - C_n(1))}{\sqrt{N}}, \quad n_\delta^s = N' \frac{\sum_n (1 - C_n(1))}{\sqrt{N}}.$$

The wall-motion rate and its thickness are readily expressed via

$$v = dL/dt = \frac{l}{\tau} \sum_n \frac{dC_n(1)}{dt'}, \quad t' = t/\tau,$$

$$h^2 = 4l^2 \left[\sum_n C_n(1)(1 - C_n(1)) + 2 \sum_n (1 - C_n(1)) \sum_{n>n} C_n(1) \right].$$ (31)

The dimensionless rate $u = \tau v/l = U(\alpha, \alpha', \alpha'', x, x')$ and thickness h oscillate with the period l, which reveals the appearance and increase of prominences from the wall in a plane. Thus the physical sense of the obtained results is presented in the fact that the present formalism describes the influence of the distribution of free electron and self-localized states of the ordering factor on the kinetics of the depolarization process. The soliton and fluctuon fields, as it is seen, effect both the speed and thickness of the domain wall.

IV. CONCLUSIONS

1. The process of depolarization of a ferroelectric with ferrofluctuons in a system as a kinetic phase transition has been analysed in the paper presented. This process is considered to be a kinetic phase transition.
2. The ordering fields of a system have been determined, as well as the fluctuation Hamiltonian. The problem is reduced to the investigation of lattice model kinetics.
3. The main kinetic equation has been formulated and solved. It is shown that taking into account solitons-fluctuons in a system leads to the renormalization of the interaction constants.

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