SOLITON REGIME IN THE MODEL WITH NO LIFSHITZ INVARIANT

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В рамках феноменологической модели без инварианта Лифшица описаны нелинейные свойства модулированной волны параметра порядка в таких системах как тиомочевина. Показано, что для некоторого набора параметров термодинамического потенциала запирающий переход в система типа II является непрерывным. В окрестности этого перехода появляется солитонная решетка.

INTRODUCTION

When describing the incommensurate (IC) phases in the ferroelectrics of so called type II [1] the phenomenological approach proposed by Y.Ishibashi and H.Shiba [2] is widely used [3].

In according to this model, the system thermodynamic potential can be written in the form [4]:

$$\Phi = \Phi_0 \frac{1}{L} \int_0^L \left\{ (\varphi'')^2 + g(\varphi\varphi')^2 - \gamma(\varphi')^2 \right.$$
$$\left. + q\varphi^2 + p\varphi^4 + h\varphi^6 \right\} dx, \qquad (1)$$

where $\varphi(x)$ is a one-dimensional order parameter (e.g., a component $P_y(x)$ of the spontaneous polarization); $\varphi'(x) \equiv \partial\varphi(x)/\partial x$, L is a crystal length in the direction of spatial modulation of the order parameter.

In the expression (1) the scale transformation is made [4] in order to emphasize the physically relevant material parameters g, q, p ($h = 0,1$; $\gamma = 1$ - like in the [4] we remain the notation 'γ' to indicate the contribution of invariant $(\varphi')^2$ which favors the appearance of IC state). For all known ferroelectrics of type II (sodium nitrite $NaNO_2$ [5], thiourea $SC(NH_2)_2$ [2], $Sn_2P_2Se_6$ [6] and betaine calcium chloride dehydrate (BCCD) [7]) the parameter g is negative: $g < 0$. For sodium nitrite and $Sn_2P_2Se_6$ the direct (virtual) disordered-to-commensurate phase transition is regarded to be of the first order, thereby $p < 0$ and $h = 1$ for $NaNO_2$ [8] and $Sn_2P_2Se_6$ [6]. In the case of thiourea and BCCD the parameter $p > 0$ and $h = 0$. It is usually assumed that the only parameter q is dependent on temperature T: $q = q_0(T - T_0)$, where q_0 and T_0 are some constants.

The model (1) fairly well describes a lot of properties of the IC phase in the ferroelectrics of type II [3]. One of such properties is a predominantly sinusoidal character of the order parameter modulation wave. Experimental studies indicate that for the type II ferroelectrics higher order satellites are of low intensity even in the close vicinity of the lock-in transition (see for review [9]). This circumstance is a reason why the one-harmonic approximation is often used when considering the IC order parameter configuration in, e.g., sodium nitrite or $Sn_2P_2Se_6$ [2,5,6,7]

$$\varphi(x) = a\sin(bx). \qquad (2)$$

On the other hand, the recent experiments [10.11] reveal that for thiourea the dependence of order parameter on position x contains relatively large contribution of the higher harmonics and can not be regarded as pure sinusoidal.

The solitonic properties of the modulation functions in thiourea are explained in [12]. In order to describe nonlinear features of the IC order parameter configuration the authors of [12] make use another than (1) phenomenological approach [13].

The approach [13] is similar to the model developed for the systems with two-component order parameter for which the Lifshitz invariant can be introduced [14]. Examples of such systems are compounds of the A_2BX_4 family, e.g., Rb_2ZnCl_4 [9]. Although in the case of type II ferroelectrics the order parameter is usually considered as one-dimensional, the role of second component is performed by some other normal coordinate $\xi(x)$ (e.g., xy-component of the elastic strain tensor) [13]. The function $\xi(x)$ transforms like the first derivative of the order parameter: $\xi(x) \sim \varphi'(x)$. These transformation properties of $\xi(x)$ allow to construct the thermodynamic potential with a term analogous to Lifshitz invariant [13].

The approach [13] is expected to be more appropriate than the model (1) when interpreting nonsinusoidal configurations of the order parameter [12]. To some extent, these expectations are grounded on the analogy between the approach [13] and the theory developed in [14]. Really, the latter constitutes a powerful tool for description of the soliton structures in the compounds of A_2BX_4 family [15].

In the present paper we show that the nonlinear contribution to the modulation wave observed in the experiments for thiourea can be explained in the framework of the model (1) as well.

Our consideration is based on the nonlinear approximation for the IC order parameter configuration proposed in [4]. Using it we obtain that if the system material parameters g and p are so that $gp^{-1} \approx -6.0$ (

$h = 0$, $p > 0$) then the ratio a_3 / a_1 for the amplitudes of third (a_3) and fundamental (a_1) harmonics of the modulation wave is equal to $a_3 / a_1 \approx 0.1$. This result completely agrees with the experimental data for thiourea [10, 11].

Moreover, it is found that for some values of the parameters g and p (for systems with $h = 0$, $p > 0$ these values satisfy the relationship $\gamma g p^{-1} \approx -16$) the transition from the IC phase into the commensurate state is continuous. The order parameter configuration is domain-like in the proximity of such transition.

The structure of present paper is following. In section 2 we formulate main features of the order parameter approximation proposed in [4]. In section 3 we consider the nonlinear configurations of order parameter in the case of thiourea. The estimations of nonlinear properties for other known ferroelectrics of type II are given as well. The principal possibility of existence of the strong soliton regime in the type II systems is investigated in section 4. The comparison of the obtained results with the properties of type I systems is presented in section 5.

SN-APPROXIMATION FOR THE IC ORDER PARAMETER CONFIGURATION

In according with the analysis made in [4], the equilibrium configuration of order parameter in the IC phase for the type II systems (1) can be approximated as

$$\varphi(x) = a \cdot sn(bx, k), \qquad (3)$$

where $sn(x, k)$ is the Jacobi elliptic sinus [16]. In (3) the amplitude a, the wave number b and the elliptic modulus k ($0 \leq k \leq 1$) are defined by the minimization of the thermodynamic potential (1) in respect to a, b, k [4].

In contrast to the approach (2), the approximation (3) allows to consider not only the linear regime of the IC phase but the nonlinear configurations of order parameter as well.

Really, if the elliptic modulus k is small ($k \approx 0$) the function (3) is closed to the dependence (2}): $sn(x, k \approx 0) \approx sin(x)$ [16]. But when $k \to 0$ the spatial behavior of elliptic sinus becomes domain-like: the wide regions with almost constant values $\varphi(x) \approx \pm \varphi_0$ are separated by the narrow regions where the function (3) changes abruptly.

In the model (1), (3) the elliptic modulus k is equal to zero at the point $q_I = \frac{1}{3}\gamma^2$ of the disordered-to-incommensurate phase transition [4]. With decreasing temperature the elliptic modulus k grows and takes its largest value k_c at the lock-in transition point q_c. The preliminary investigations show that k_c can be close to unity for some values of the material parameters. For example, if $g = -10$, $p = 1$, $h = 0$ then $k_c = 0.965$ [4].

Another important property of the order parameter approximation (3) is an additional (in comparison with the approach (2)) mechanism causing the change of modulation period. Another important property of the order parameter approximation (3) is an additional (in comparison with the approach (2)) mechanism causing the change of modulation period.

The continuous dependence of modulation period P on temperature is one of the most characteristical features of the IC phases [3]. In the framework of approach (2) the period of IC structure is equal to $P = 2\pi / b$. The wave number b depends on temperature only when the $(\varphi \varphi')^2$ - invariant is present in the expansion of the thermodynamic potential (1) [2, 5]:

$$b^2 = \frac{1}{2}\gamma^2 + \frac{1}{8}ga^2. \qquad (4)$$

And if the material parameter g is negative then the period P increases with decreasing temperature as it is observed in experiments [9].

The period of IC order parameter configuration (3) is defined as $P = 4K(k) / b$ ($K(k)$ is the complete elliptic integral of the first kind [16]). It depends not only on the value of b (as it takes place in the one-harmonic approach (2)) but also on the elliptic modulus k which characterizes the degree of nonlinearity of the modulation wave. As consequence, the approximation (3) imposes less strong requirements on the material parameters, in particular, on g (e.g., the period P grows even when $g = 0$ [4, 17]). Moreover, if the elliptic modulus k is close to unity $k \to 1$ (i.e. $K(k) \to \infty$ [16]) then the nonlinear mechanism of the increase of modulation period becomes dominant and P can be very large: $P \to \infty$.

The numerical investigation of variational equation for the functional (1) [18] shows that the approximation (3) correctly reproduces nonlinear properties of the equilibrium configuration of order parameter in the IC phase.

NONLINEAR CONFIGURATIONS OF THE ORDER PARAMETER IN THIOUREA

Now let us to apply the model (1), (3) to thiourea.

As it has been mentioned above, in the case of thiourea the material parameter $h = 0$. For the sake of simplicity we also assume that $p = 1$.

Numerical minimization of the thermodynamic potential (1), (3) in respect to a, b, k shows that if the material parameter g is equal to $g = -6.0$ then the elliptic modulus takes the value $k_c \approx 0.923$ at the point $q_c = -0.290$ of the lock-in transition. Using the formulae for the Fourier expansion of elliptic sinus [16] or the procedures of fast Fourier transformation (FFT), one can find that for this k_c the ratio of amplitudes of the third (a_3) and the fundamental (a_1) harmonics of the modulation wave (3) is equal to $a_3 / a_1 \approx 0.104$ and also $a_5 / a_1 \approx 0.012$ (a_5 is an amplitude of the fifth harmon-

ic). The spatial dependence of the order parameter $\varphi(x)$ at the temperature q_c is shown in figure (full line). The obtained results are in a good agreement with the experimental data [10, 11].
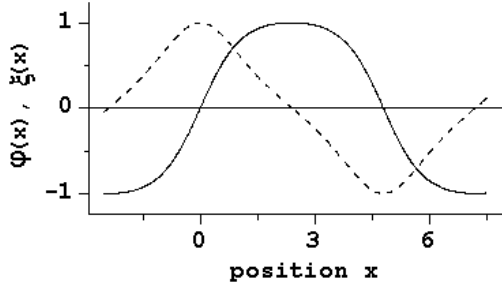
The order parameter $\varphi(x)$ (full line) and the modulation function $\xi(x)$ (broken line) as functions of position x at the temperature $q_c = -0.29$ (the lock-in transition point). The material parameters are the following: $g = -0.6$, $p = 1$, $h = 0$. The amplitudes of order parameter $\varphi(x)$ and function $\xi(x)$ are arbitrary

Besides the order parameter $\varphi(x)$, some other modulation functions are also discussed in the case of thiourea [12]. If such function $\xi(x)$ is coupled with the order parameter $\varphi(x) = a \cdot sn(bx, k) \equiv a \cdot sin[\theta(x)]$ by the relation $\xi(x) \sim a \cdot cos[\theta(x)]$ then the dependence of $\xi(x)$ on position x has the form depicted in figure by broken line (cf. with figure 3 in [12]).

Of course, the complete description of $\xi(x)$ and other modulation functions requires a modification of the thermodynamic potential (1) by including additional invariants which correspond to interactions of $\xi(x)$,... with each other and with the order parameter. Such an analysis is, however, beyond the scope of the present paper.

The nonlinear properties of the IC modulation wave in thiourea essentially differ from ones for sodium nitrite. Taking for estimations the material parameters given in [5] (in our notations they correspond to $g = -9.51$, $p = -0.651$, $h = 1$) we find that for NaNO$_2$ the lock-in value of elliptic modulus is $k_c \approx 0.589$, $a_3 / a_1 \approx 0.026$ and $a_5 / a_1 \approx 0.0007$. Note that these results are very close to ones obtained analytically in [19].

As for other ferroelectrics of type II, estimations reveal that for Sn$_2$P$_2$Se$_6$ ($g = -1.37$, $p = -0.19$, $h = 1$ [6]) the characteristics of modulation wave at the lock-in transition point are the following: $k_c \approx 0.706$, $a_3 / a_1 \approx 0.041$ and $a_5 / a_1 \approx 0.0018$; and for BCCD ($g = -8.0$, $p = 2$, $h = 0$ [7]) $k_c \approx 0.887$ $a_3 / a_1 \approx 0.086$ and $a_5 / a_1 \approx 0.0082$.

SOLITON REGIME IN THE TYPE II SYSTEMS

Results given in the preceding section describe the nonlinear properties of the IC modulation in four known

compounds belonging to the type II ferroelectrics. However, these estimations do not answer on more general question: to what extent the soliton regime can develop in the model (1), (3) in principle. In order to clarify the situation we have investigated the behavior of systems (1}), (3) in the limit $k \to 1$ in more detail.

When $k \to 1$ the thermodynamic potential (1), (3) acquires the form [4]:

$$\Phi \approx a^2\left[q\left(1 + \frac{1}{2}k'^2\right) - \Lambda^{-1}\left(q + \frac{2}{3}\gamma b^2 - \frac{8}{15}b^4\right) - \frac{1}{2}k'^2\Lambda^{-1}\left(q + \frac{8}{15}b^4\right)\right]$$
$$+ a^4\left[\frac{1}{2}p\left(1 + k'^2\right) - \Lambda^{-1}\left(\frac{2}{3}p + \frac{2}{15}gb^2\right) - k'^2\Lambda^{-1}\left(\frac{2}{3}p + \frac{1}{15}gb^2\right)\right]$$
$$+ a^6 \cdot \frac{1}{3}h\left[1 + \frac{3}{2}k'^2 - \frac{23}{15}\Lambda^{-1} - \frac{23}{10}k'^2\Lambda^{-1}\right], \quad (5)$$

where $k'^2 = 1 - k^2$, $\Lambda = ln(4/k')$; $\Lambda^{-1} \to 0$ if $k \to 1$.

The equilibrium wave number b can be found from the equation $\partial\Phi / \partial b = 0$ what yields (cf. with (4)):

$$b^2 = \frac{5}{8}\gamma\left(1 + \frac{1}{2}k'^2\right) + \frac{1}{8}ga^2\left(1 + k'^2\right). \quad (6)$$

Taking into account (6) the thermodynamic potential (5) can be rewritten as

$$\Phi \approx a^2\left(1 + \frac{1}{2}k'^2\right)\left[q - \Lambda^{-1}\left(q + \frac{5}{24}\gamma^2\right)\right]$$
$$+ a^4\left(1 + k'^2\right)\left[\frac{1}{2}p - \Lambda^{-1}\left(\frac{2}{3}p + \frac{1}{12}\gamma g\right)\right]$$
$$+ a^6\left(1 + \frac{3}{2}k'^2\right)\left[\frac{1}{3}h - \Lambda^{-1}\left(\frac{23}{45}h + \frac{1}{120}g^2\right)\right]. \quad (7)$$

The function Λ^{-1} changes much slower then k'^2 approaches to zero (e.g., for $k'^2 = 0.1$ the value of Λ^{-1} is 0.394, for $k'^2 = 4 \cdot 10^{-8}$ - $\Lambda^{-1} \approx 0.1$). Hence, it is reasonable to omit the terms proportional to k'^2 in (7).

The further analysis depends on the value of material parameter h. For the systems with $h = 0$, $p > 0$ the results are as follows.

The equation $\partial\Phi / \partial a = 0$ defines the equilibrium amplitude a. Substituting its solution into the expression (7) and comparing the result with the thermodynamic potential for the commensurate state $\Phi_c = -q^2 / (2p)$, we find the effective temperature q_c of the lock-in transition:

$$q_c = 5g^{-2}p^2\left[\gamma gp^{-1} - 4 + 4\left(1 - \frac{1}{2}\gamma gp^{-1}\right)^{1/2}\right]$$
$$\approx \frac{5}{2}\gamma^2\left[\gamma gp^{-1} - 4\right]. \quad (8)$$

In (8) the approximate (right hand side) formula is derived for the case when the term proportional to a^6 is

neglected in (7). This expression clearly demonstrates main features of the dependence of q_c on gp^{-1}.

Now we consider the conditions under which the elliptic modulus can be equal to unity at the lock-in transition point q_c.

The thermodynamic potential (7) depends on the elliptic modulus k only through the function Λ^{-1} (remember we omit the terms proportional to k'^2 in (7)). Thereby it is convenient to formulate the variational task for k in terms of Λ^{-1}. In these terms the condition $k_c = 1$ means $\Lambda\left(k_c\right)^{-1} = 0$, and the equation $\partial\Phi / \partial k = 0$ is equivalent to $\partial\Phi / \partial\left(\Lambda^{-1}\right) = 0$. Solving the latter at the temperature q_c (8), we find that the function $\Lambda\left(k_c\right)^{-1}$ is equal to zero if the material parameters satisfy the relation:.

$$\gamma gp^{-1} = -16.\qquad(9)$$

Therefore, for the systems with material parameters g and p related with each other in according to (9) ($h = 0$), the elliptic modulus of the order parameter modulation wave (3) is equal to unity at the point of lock-in transition. It means that for such systems the transition from the IC phase into the commensurate state is continuous. Close to this transition the order parameter spatial configuration is domain-like, and the soliton density $n_S = \pi / \left[2K\left(k\right)\right]$ (see [12] and references therein) approaches to zero with decreasing temperature q to its lock-in value q_c.

Numerical calculations confirm the results of analytical investigation (see table).

For the case $gp^{-1} = -16$ the elliptic modulus k_c has approached to unity, but we have stopped calculations at the value $k_c = 0.9999$ The properties of elliptic functions change abruptly at the point $k = 1$ [16], and there are some difficulties to reproduce the point $k = 1$ numerically.

Characteristics of the order parameter modulation wave in the type II systems (the case $h = 0$, $p > 0$) at the point q_c of lock-in transition for different values of the material parameters g and p: k_c is a lock-in value of the elliptic modulus, a_3 / a_1 is a ratio of the third and first harmonics, n_S is a soliton density

g/p	q_c	k_c	a_3/a_1	n_S
-1.	-0.690	0.768	0.052	0.81
-10.	-0.212	0.065	0.136	0.57
-16.	-0.156	0.9999	0.262	0.27
-20.	-0.134	0.970	0.142	0.56
-100.	-0.048	0.766	0.052	0.81

As it follows from table the dependence of k_c on gp^{-1} has a maximum when $gp^{-1} = -16$. For small ($gp^{-1} \sim 1$) and large ($gp^{-1} \sim 100$) values of the parameter combination gp^{-1} the contribution of higher harmonics in the modulation wave is relatively small.

The analysis for the case of systems with $h = 1$, $p < 0$ can be made in the similar manner. Here we point out only the following.

When $h = 1$, $p < 0$ the direct disordered-to-commensurate phase transition is of the first order. As consequence, for large enough values of the parameter $|p|$ the range of IC phase stability can be relatively small like it takes place for $NaNO_2$ ($q_c \approx +0.07$) [5]. Nevertheless, for any p there exists some g for which $k_c = 1$. For example, for $p = -0.65$ (the case of sodium nitrite [4, 5]) the lock-in value k_c of the elliptic modulus of the modulation wave (3) equals to 1 if the material parameter g is $g \approx -0.45$.

However, due to the presence of term proportional to a^6 in (1), i.e. due to $h = 1$, the material parameters g and p are not so correlated as in the case of $h = 0$ (in fact, when $h = 0$ the ratio g / p is relevant rather than the parameters g and p themselves). As consequence, for the systems with $h = 1$, $p < 0$ the dependence $g(p)$ providing $k_c = 1$ is more complex than (9).
square well and oscillator potentials.

DISCUSSION

In the present paper we have shown that in the framework of the phenomenological model with no Lifshitz invariant [2, 8] different nonlinear configurations of the IC order parameter can be described: almost sinusoidal one as in the case of sodium nitrite ($a_3 / a_1 \leq 0.03$); one with more large contribution of higher harmonics as in thiourea ($a_3 / a_1 \approx 0.01$); strong soliton regimes when the lock-in transition is continuous (such compound is unknown at the moment). Nonlinear properties of the concrete system are defined by values of the material parameters g and p (see, e.g., table).

The specific role of $\left(\phi\phi'\right)^2$ - invariant should be emphasized when discussing the nonlinear features of the IC\ order parameter configurations. If this term is not included in the thermodynamic potential (1) the soliton structure does not develop (see also [4, 17]). From this point of view, $\left(\phi\phi'\right)^2$ - term is analogous to the Umklapp invariant of relatively low order (the anisotropy invariant) which favors the appearance of domain-like structures in the type I ferroelectrics [3, 14]. The difference between $\left(\phi\phi'\right)^2$ - term in (1) and the anisotropy invariant is that the former is a part of the gradient gain of the thermodynamic potential and can not influence on characteristics of the commensurate phase. On the contrary, in the type I ferroelectrics the behavior of IC and commensurate phases is correlated due to the anisotropy invariant (it belongs to the local interactions). In the case of type II systems analogous

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correlation appears only for some specific values of the material parameters g and p, i.e. only for some correlated actions of the invariants $(\varphi \varphi')^2$ and φ^4 which define the low-temperature behavior of IC and commensurate phases.

The existence of IC state in the type I systems is caused by symmetric properties (the Lifshitz condition is not fulfilled) [3]. For the systems of type II such global reasons are absent and the spatial modulation of the order parameter is a consequence of specific features of the interatomic interactions [3].

As result, in the type II ferroelectrics the appearance of soliton regime has no systematic character, in contrast to the situation which takes place for, e.g., compounds of the A_2BX_4 family.

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СОЛИТОННЫЙ РЕЖИМ В МОДЕЛЯХ БЕЗ ИНВАРИАНТА ЛИФШИЦА

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В рамках феноменологической модели без инварианта Лифшица описаны нелинейные свойства модулированной волны параметра порядка в таких системах как тиомочевина. Показано, что для некоторого набора параметров термодинамического потенциала запирающий переход в система типа II является непрерывным. В окрестности этого перехода появляется солитонная решетка.

СОЛІТОННИЙ РЕЖИМ В МОДЕЛЯХ БЕЗ ІНВАРИАНТА ЛІФШИЦЯ

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В рамках феноменологічної моделі без інваріанта Ліфшиця описано нелінійні властивості модульованої хвилі параметра порядку в таких системах як тіомочевина. Показано, що для деякого набору параметрів

термодинамічного потенциалу замикаючий перехід в система типу II виявляється неперервним. Поблизу цього переходу з'являється солітонна гратка.

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