Landau Model for Commensurate-Commensurate Phase Transitions in Uniaxial Improper Ferroelectric Crystals

M. Latković and A. Bjeliš
Department of Theoretical Physics, Faculty of Science
University of Zagreb, Bijenička 32, 10000 Zagreb, Croatia

V. Dananić
Department of Physics, Faculty of Chemical Engineering and Technology
University of Zagreb, Marulićev trg 19, 10000 Zagreb, Croatia

We propose the Landau model for lock-in phase transitions in uniaxially modulated improper ferroelectric incommensurate-commensurate systems of class I. It includes Umklapp terms of third and fourth order and secondary order parameter representing the local polarization. The corresponding phase diagram has the structure of harmless staircase, with the allowed wave numbers obeying the Farey tree algorithm. Among the stable commensurate phases only those with periods equal to odd number of lattice constants have finite macroscopic polarizations. These results are in excellent agreement with experimental findings in some A_2BX_4 compounds.

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Many uniaxial systems undergo a sequences of incommensurate-commensurate (IC-C) or commensurate-commensurate (C-C) phase transitions 1,2. It is generally accepted that the main cause of such behavior comes from the mode softening induced by competing interactions between neighboring sites in the crystal lattice 3,4. The mode softening occurring at an arbitrary point in the Brillouin zone characterizes the so called class I of IC-C systems, in contrast to the examples of the class II for which the wave vectors of soft modes are at (or close to) the center or the border of the Brillouin zone 3.

The IC-C and C-C phase transitions are mostly of the first order, and are accompanied with hysteresis and memory effects. Also, depending on the wave number of modulation, some commensurate phases have a uniform (e.g. ferroelectric) component. Recent experiments show that the modulation of commensurate phases is usually domain like, suggesting that multisoliton configurations that are usually present in the incommensurate states, are to some extent frozen in lock-in phases as more or less dense soliton lattices 5-7.

Theoretical considerations of the class I of IC-C systems mostly start either from discrete microscopic models of competing interactions or from phenomenological expansions of the free energy functional. In the former cases the sequence of phase transitions is usually characterized by complete or incomplete devil’s staircases for the wave number of ordering 8,10. The crucial importance of discreteness in these models comes from the assumption that the couplings between neighbouring atoms or molecules are very strong. However, as argued below, this assumption usually does not suit the microscopic properties of real materials.

The latter approach is based on the Landau theory of phase transitions, and is generally appropriate for weakly coupled systems. The justification for continuous Landau models comes from many experimental indications, e.g. from the neutron scattering data 11-14, showing a well-defined dispersion curves for collective modes with distinct soft-mode minima. The basic Landau model, commonly applied to various systems of the class I, includes one Umklapp term and the Lifshitz invariant. Within the mean field approximation it leads to the sine-Gordon problem 15,17 which has one isolated phase transition of the second order to the unique commensurate phase. However, in such systems one usually encounters more commensurate phases, sometimes with the order of commensurability much higher than three or four.

The phases with high order commensurabilities can be explained only by some extension of the sine-Gordon model. One widely explored way is to introduce an additional set of high order Umklapp terms 18-20, more precisely to take into account as many terms as there are stable commensurate states in the experimental phase diagram. This approach is however not free from serious disadvantages. At first, one introduces terms with powers of the order parameter amplitude which are twice the order of commensurability, and as such are beyond the standard (minimal) Landau scheme that includes only those higher (usually fourth) order terms which necessarily guarantee the boundness of the free energy density from below. Further on and even more important, the microscopic analyses, which are usually avoided in such approaches, show that such terms are as a rule very small, and as such physically irrelevant in weakly coupled systems.

The model which is, in contrast to those of Refs. 18-20, formulated within the standard Landau scheme, and still explains the stabilization of high order commensurate phases, is proposed recently by two of us 21,22. In this model
we take into considerations two Umklapp terms of the lowest possible orders in the free energy expansion, i.e. those of the third and fourth order, and so remain strictly within the Landau theory \[23\]. The similar starting point, but with Umklapp terms of orders higher than four, was proposed earlier in Ref. \[21\]. In Refs. \[21,22\] we show that, due to the competition between these two Umklapp terms, the periods of ordering follow harmless staircase with the values obeying a Farey tree algorithm, while the corresponding commensurate configurations have the soliton lattice-like forms. Also, the hysteresis and memory effects which are usually observed in particular materials, are within the present model interpreted as an intrinsic property, i.e. as consequences of the free energy barriers that appear due to the nonintegrability of the Landau functional. These barriers prevent smooth transitions between neighbouring thermodynamically stable configurations \[23,27,21,22\].

The Landau model from Refs. \[21,22\] contains only one order parameter, and as such does not possess all ingredients needed for the explanation of phase transitions in uniaxial improper ferroelectric materials. For such materials it is necessary to include a secondary order parameter, that responsible for the net polarization of particular modulation structures. Although the necessity for such extension was put forward already by Iizumi et al. \[11\], the subsequent studies of phase transitions in improper ferroelectrics were developed mostly within the simple sine-Gordon model with one Umklapp term \[21,22\]. These analyses were concentrated on one isolated IC-C phase transition, allowing at most for its first order nature.

Our aim is to show that the model of Iizumi et al. \[11\], extended with the additional Umklapp term which provides the mechanism for sequences of C-C phase transitions \[21,22\], enables the understanding of the complex phase diagrams in \(A_2BX_4\) compounds. In particular we explain several features of C-C phase transitions, like harmless staircase behaviour of the wave number of ordering, the first order nature of phase transitions, and the polarization and modulation properties of commensurate phases.

We start from the assumption that the quadratic soft mode contribution to the Landau expansion has minima at wave numbers \((\pm q_c, -q_c)\), where \(q_4 < q_c < q_3\), with \(q_4 = 2\pi/4\) and \(q_3 = 2\pi/3\) (the unit length is taken equal to the lattice constant). The distances of \(q_c\) from \(q_3\) and \(q_4\) are denoted by \(\delta_3\) and \(\delta_4\) respectively, with \(\delta_3 + \delta_4 = \pi/6\), which gives the first independent control parameter, \(\delta_4\). The order parameter is complex, \(\rho e^{i\phi}\). Limiting the further analysis to the temperature range well below the critical temperature for the transition from the normal (disordered) to the incommensurate phase, we also make the usual approximation of space independent amplitude \(\rho\), and keep only the phase-dependent part of the free energy density.

To allow for a finite polarization along some direction, we introduce a secondary order parameter \(u\), and assume that it is coupled to the third-order Umklapp term, in agreement with the symmetry requirements for \(A_2BX_4\) crystal lattice \[11\]. In order to stabilize the free energy with respect to changes in \(u\), we also introduce the terms proportional to \(u^2\) and \(u^2\). The complete free energy density reads

\[
f(\phi, u, x) = \frac{1}{2} \phi'^2 + \frac{1}{2} u'^2 + \frac{1}{2} \lambda u^2 + Bu \cos [3\phi + 3(\frac{\pi}{6} - \delta_4)]x + C \cos (4\phi - 4\delta_4)x,
\]

(1)

where primes denote spatial derivation. The coefficients \(\lambda\), \(B\) and \(C\) in Eq. (1) are rescaled in order to simplify equations. Thus \(f(\phi, u, x)\) is the original free energy density divided by \(\rho^2\). Note also that after this rescaling the coefficients \(B\) and \(C\) in Eq. (1) are linear and quadratic in the order parameter amplitude \(\rho\) respectively. Altogether, there are three control parameters, namely \(\delta_4\), \(B\) and \(C\), while the parameter \(\lambda\) just defines a scale for the polarization \(u\). Among the elastic terms in Eq. (1), the first one \((\phi'-\text{dependent})\) favors the incommensurate sinusoidal ordering with the wave number \(q_c\). The last two Umklapp terms cause the harmless staircase behaviour of the wave number of ordering \[21,22\]. The above Landau expansion follows, after spatial continuation, from microscopic models like e.g. those of Refs. \[30,31\], that take into account local interactions presumably responsible for the mode softening (and for the coupling to the secondary order parameter), and start from the discrete presentation along the uniaxial direction.

The mean-field approximation for free energy \(f(\phi, u, x)\) leads to the Euler-Lagrange (EL) equations:

\[
\phi'' + 3Bu \sin [3\phi + 3(\frac{\pi}{6} - \delta_4)]x + 4C \sin (4\phi - 4\delta_4)x = 0,
\]

\[
u'' - \lambda u - B \cos [3\phi + 3(\frac{\pi}{6} - \delta_4)]x = 0.
\]

(2)

We are interested only in those solutions of EL equations which participate in the thermodynamic phase diagram of the model \[1\]. These solutions have, for given fixed values of the control parameters, the lowest values of the averaged free energy

\[
\langle F \rangle = \frac{1}{L} \int dx f(\phi(x), u(x), x).
\]

(3)
\( L \) is the macroscopic length of the system.

Before calculating such solutions, let us make few remarks on EL equations (2). They can be considered as Lagrange equations for an equivalent classical mechanical problem. Note however that general ("classical mechanical") solutions of these equations are not bounded, since \( \lambda \) is positive. The only bounded solutions are periodic. From one side, obviously only such solutions may participate in the thermodynamic phase diagram. From the other side, the unboundedness of all other solutions makes the analysis of the present model more complicated than that of the model without the secondary order parameter \( u \) [21,22], for which the mechanical phase portrait is bounded, although chaotic [22].

It is clear from above remarks that periodic solutions are orbitally unstable. Therefore they cannot be calculated by a direct numerical integration of EL equations (2). Before embarking into another suitable numerical method, it is useful to establish, by extending straightforwardly the previous treatment [22], necessary analytic conditions for these periodic solutions. We start by looking for allowed periods, after taking into account that periodic solutions have to obey the relations

\[
\phi(x + P) = \phi(x) + \phi_P, \\
u(x + P) = \nu(x),
\]

where \( P \) is the period and \( \phi_P \) is the phase increment per period. From EL equations (2) it follows that allowed periods and phase increments are

\[
P = 4k + 3l, \quad \phi_P = \delta_4P - l\frac{\pi}{2},
\]

where \( k \) and \( l \) are integers. The corresponding values of the total wave number (measured from the origin of Brillouin zone) are then

\[
2\pi q \equiv q_e = \phi_P = 2\pi \frac{k + l}{4k + 3l}.
\]

The values of \( q \) given by Eq. (1) form a Farey tree structure, shown in Fig. 1 for wave numbers between \( q = 1/3 \) \((k = 0, l = 1 \text{ and } P = 3)\) and \( q = 1/4 \) \((k = 1, l = 0 \text{ and } P = 4)\). The periodic solutions with \( q = 1/3 \) and \( q = 1/4 \) are the basic commensurate configurations, favoured by the Umklapp terms of third and fourth order respectively. A higher order commensurate configurations are represented by periodic solutions with wave numbers \( q \) positioned between \( q = 1/3 \) and \( q = 1/4 \) in the Farey tree. The values of integers \( k \) and \( l \) are positive for all these configurations. We emphasize that the Farey tree structure for \( q \) is not the characteristic of usual continuous Landau models for IC-C transitions [15–17]. Here it is imposed through the competition between two Umklapp terms in the first of EL equations (2).

For numerical purposes it appears convenient to eliminate the explicit \( x \)-dependence from one of Umklapp terms in the EL equations (2), by passing from the variable \( \phi(x) \) to

\[
\psi(x) = \phi(x) + (\frac{\pi}{6} - \delta_4)x.
\]

The EL equations (2) now read

\[
\psi'' + 3Bu \sin (3\psi) + 4C \sin (4\psi - \frac{2\pi}{3}x) = 0 \\
u'' - \lambda \nu - B \cos (3\psi) = 0.
\]

The corresponding free energy acquires a \( \delta_4 \)-dependent term in the form of Lifshitz invariant,

\[
F = \int dx \left\{ \frac{1}{2} [\psi' - (\frac{\pi}{6} - \delta_4)]^2 + \frac{1}{2} u'^2 + \frac{1}{2} \lambda u^2 + Bu \cos (3\psi) + C \cos (4\psi - \frac{2\pi}{3}x) \right\},
\]

which considerably simplifies the calculation of the \( \delta_4 \)-dependence of the averaged free energy (3) for particular periodic solutions of the EL equations (2). Note that these equations do not contain the parameter \( \delta_4 \), so that it is sufficient to find solutions in variable \( \psi(x) \) for some parameter values of \( B \) and \( C \).

Once the analytic conditions (3) are established, the orbitally unstable periodic solutions of Eq. (3) can be systematically calculated by treating Eqs. (3) as a boundary value problem, and using an appropriate numerical algorithm suitable for nonlinear differential equations, like the finite difference method. The boundary conditions have to be
specified on the left and right end points of the integration, i.e. for \( x = x_0 \) and \( x = x_0 + P \). It is convenient to put \( x = x_0 \) at one of the inflection points of \( \psi(x) \). To complete the boundary conditions we still have to find out values of \( \psi(x_0) \) and one of the values \( u(x_0) \) or \( u'(x_0) \), as will be discussed in detail elsewhere. Tab. I contains choices of \( x_0, \psi(x_0) \) and \( u(x_0) \) or \( u'(x_0) \) that specify completely the appropriate boundary conditions. As previously, we find two independent periodic solutions for given values of \( k \) and \( l \) (i.e. for a given wave number \( q \)). They differ by symmetry and have distinct average free energies. These solutions are denoted as type A (antisymmetric in \( \psi \), symmetric in \( u \)) and type B (without particular symmetry). These solutions are uniquely determined once the parameters \( (k, l) \) are chosen. Note that they do not depend continuously on the parameter \( \delta_q \). The additional minimization of the free energy (1) provides only the \( \delta_q \)-dependence of ranges of stability for each pair \( (k, l) \).

Three low order periodic solutions of Eq. (8), with wave numbers \( q = 1/3, 1/4 \) and \( 2/7 \), are shown in Fig. 2. For each of these values the phase of the primary order parameter, \( \phi(x) \), shows qualitatively different behavior. A linear \( x \)-dependence of \( \psi \) (i.e. a simple sinusoidal modulation of the primary order parameter) is realized for \( q = 1/4 \). For \( q = 1/3 \) \( \psi \) has an additional sinusoidal variation, while for \( q = 2/7 \) one encounters the periodic alternation of short domains with \( q = 1/4 \) and \( q = 1/3 \) modulations, or, in other words, the formation of a dense soliton lattice. The tendency towards more and more dilute soliton lattices strengthens as one goes down along the Farey tree from Fig. 1. The secondary order parameter \( u(x) \), i.e. the local polarization, either changes periodically in space with alternate positive and negative values forming an antiferroelectric lattice (type B solution from Fig. 2a, and type A solution from Fig. 2b) or has everywhere a ferroelectric space dependence (type B solution from Fig. 2c).

From the macroscopic point of view we are interested in spatially averaged polarizations, given by

\[
\langle u \rangle = \frac{1}{P} \int_{x_0}^{x_0+P} u(x) \, dx
\]

for periodic solutions. The values of \( \langle u \rangle \) for solutions with various values of the wave number \( q \) are listed in Tab. I. Note that the averaged polarization \( \langle u \rangle \) vanishes for all solutions with even values, and all type B solutions with odd values, of the period \( P \).

The above boundary value method enables the calculation of thermodynamically stable solutions for rather high values of the parameters \( k \) and \( l \) in Eq. (1). Here we limit the analysis of the phase diagram by keeping up to the tenth row in the Farey tree from Fig. 1. Also, in order to facilitate the further discussion, we fix the value of the parameter \( B \), and calculate the phase diagram in the reduced parametric space \( (C, \delta_q) \), as shown in Fig. 3. Other choices of the cross sections in the parameter space lead to the qualitatively same conclusions.

As is seen in Fig. 3, only the lowest order commensurate phases, namely 1/3, 1/4, 2/7, and 3/10 are present for large values of \( C \). By decreasing \( C \), i.e. by moving towards the sine-Gordon limit \( (C \to 0) \), more and more higher order commensurate phases start to participate in the phase diagram. E.g., for \( C \approx 0.05 \) new phases with the commensurabilities 3/11, 4/13, 5/16 are present.

The phases which have nonzero average polarizations are denoted by the letter \( F \) in Fig. 3. As numerical calculations show, all phases with odd periods, e.g. those with wave numbers 1/3, 2/7 and 3/11, turn out to be ferroelectric. In other words, configurations with odd periods that have lowest averaged free energies, all belong to the type A solutions from Tab. I. Note also that all lines in Fig. 3 represent phase transitions of the first order.

The phase diagram from Fig. 3 is in qualitative agreement with experimental phase diagrams for some members of the \( A_2BX_4 \) family. As an example we take rubidium tetrabromozincate, \( \text{Rb}_2\text{ZnBr}_4 \), for which there is a variety of data on the temperature and pressure variation of the modulation wave number. Our phase diagram is in qualitative agreement with high pressure measurements collected in Fig. 9 of Ref. [24] and in Fig. 8 of Ref. [26]. In particular, the experimentally observed high order commensurate phases with \( q \) equal to 2/7, 3/10, 3/11, 4/15, 5/17 and 7/24 are all present in Fig. 3. Note also that our expression (1) (and Fig. 1), which is obtained as an inherent property of the competition between two Umklapp terms, represents the theoretical explanation of the phenomenological hint on the Farey tree structure of experimentally observed commensurabilities. Regarding the macroscopic polarization associated with various commensurate phases with odd periods, up to now the measurements are reported only for that with the lowest commensurability \( (1/3) \). It shows a finite ferroelectric order, in agreement with our results. The experimental investigations of phases with higher commensurabilities, for which our results suggest presence or absence of a finite ferroelectric component, are highly desirable.

The phase is ferroelectric, the mixture of phases 2/7 and 1/4 shows a weak ferroelectricity, and some
antiferroelectric order is observed in the nonpolar phase 1/4. This is in agreement with our results in Fig. 2. Indeed, the local polarization $u(x)$ for type A solution is constant for phase 1/3, it is nonpolar and has an antiferroelectric modulation for phase 1/4, while for the phase 2/7 we expect a weak ferroelectricity since the solution $u(x)$ of type A, although almost antiferromagnetically modulated, still has a nonzero averaged value $\langle u \rangle$.

The last example, ammonium hydrogen selenate $\text{NH}_4\text{HSeO}_4$ (together with its deuterated version $\text{ND}_4\text{DSeO}_4$), does not belong to the $A_2\text{BX}_4$ family. The sequence of phase transitions shown in Fig. 8 of Ref. [41], namely 1/3 (F), 3/10, 2/7 (F) and 1/4, can be also reproduced by going along a particular path in Fig. 3. Let us also note that the present model is not directly applicable to betaine-calciumchloride-dihydrate (BCCD), a system with a well-known rich sequence of IC-C and C-C phase transitions [12-13]. In this material, usually considered as a member of class II, one very probably encounters a competition of the first (ferroelectric, $q = 0$) and some higher order (probably $q = 1/4$) commensurabilities.

Let us finally shortly focus on the form of the commensurate modulations. As already mentioned, multisoliton structures are sometimes frozen in commensurate phases well below the IC-C transitions [2, 3]. Our numerical analysis shows that the stable solutions of higher orders (like that with $q = 2/7$ shown in Fig. 3) have the properties of dense, and not dilute, soliton lattices. It also suggests that particular solutions keep this form well below lock-in phase transitions, and that the polarization closely follows such behaviour of the dominant order parameter by forming the modulated ferroelectric or antiferroelectric patterns.

In conclusion, by extending the basic model [21, 22] with the secondary order parameter representing the ferroelectric polarization, we obtain the phase diagram for the series of commensurate phases which is in qualitative and quantitative agreement with experimental phase diagrams for some $A_2\text{BX}_4$ compounds [4-8, 9, 10], as well as for ammonium hydrogen selenate [11]. The main results of the present analysis are as follows. The wave number of ordering is given by the harmless staircase obeying the Farey tree algorithm. The commensurate phases are mainly characterized by dense soliton lattice modulations. The phase transitions between successive lock-in phases are of the first order. Finally, we establish the selection rule, by which only those among the commensurate phases which have odd periods are accompanied by finite macroscopic electric polarizations.

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TABLE I. Boundary conditions for periodic solutions of EL equations (8).

| k   | l   | Type | $x_0$ | $\psi_0$ | $u_0$ | $u_0'$ |
|-----|-----|------|-------|----------|-------|--------|
| odd | odd | A 0  | 0     | 0        | 0     | 0      |
|     |     | B 1  | $\pi/6$ | 0        | 0      | 0      |
| even| odd | A 0  | 0     | 0        | 0     | 0      |
|     |     | B 1  | $\pi/6$ | 0        | 0      | 0      |
| odd | even| A 0  | 0     | 0        | 0     | 0      |
|     |     | B 3/2 | 0     | 0        | 0      | 0      |

TABLE II. Polarization properties of commensurate phases with wave number $q$ (Eq. (6)). Letter F indicates that particular solution have nonzero average polarization (Eq. (10)).

| k   | l   | $q$  | Type | $\langle u \rangle$ |
|-----|-----|------|------|---------------------|
| odd | odd | even | A    | F                   |
|     |     | odd  | B    | 0                   |
| even| odd | odd  | A    | F                   |
|     |     | odd  | B    | 0                   |
| odd | even| odd  | A    | 0                   |
|     |     | even | B    | 0                   |

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FIG. 1. Farey tree for wave numbers $q$ defined by Eq. (6).

FIG. 2. Solutions of EL equations (8) $\psi(x)$ (phase of the primary order parameter) and $u(x)$ (local polarization): $k = 0$, $l = 1, q = 1/3$ (a), $k = 1, l = 0, q = 1/4$ (b) and $k = 1, l = 1, q = 2/7$ (c). Other parameters are $B = 0.1, C = 0.1, \lambda = 1$. 
FIG. 3. Phase diagram in the \((C, \delta_4)\) plane for \(B = 0.3\) and \(\lambda = 1\). The numbers in the figure represents wave numbers of ordering \(q\) of commensurate phases. Phases with nonzero average polarizations \(\langle u \rangle\) are marked by the letter F.