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Abstract
A theoretical approach to the crystalline-to-amorphous phase transformation during self-irradiation is proposed. The different stages of the amorphization process (metamictization) are shown to correspond to internally stressed equilibrium states, associated with the minima of a free-energy functional, in which the elastic distortions induced by the creation of amorphous material are compensated by an internal stress-field, deriving from a unique stress-field potential. The transformation dynamics is described in terms of a nucleation and growth mechanism of amorphous regions involving a critical size.

1. Introduction

In some minerals radioactive elements such as uranium and thorium decay spontaneously by α-radiation, creating heavy recoil nuclei that undergo ballistic collisions with the atoms of the surrounding crystalline matrix, displacing them from their equilibrium positions. Over time the long-range order of the original crystal is affected undergoing a gradual transformation to an amorphous state called the *metamict* state [1, 2]. Such transformation is known to occur naturally in mineral phases such as zircon [3], titanite [4], the apatites [5], pyrochlores [6] and others [2]. Intensive experimental efforts have been made to clarify the amorphization process leading to the metamict state by investigating either minerals of known age and U and Th contents, or obtained by actinide doping, or by ion beam irradiation. These efforts have been strongly motivated by the increasing importance of finding alternative ways to store the nuclear waste produced world wide. One of the options being considered [8] is the immobilization with specially formulated ceramic forms. The evaluation of the effect of radiation on their crystalline phases and the resulting damages, as well as the correlated changes in their physical and chemical properties, are some of the critical concerns for their application. In this respect

zircon has recently become a model host matrix for studying encapsulating of plutonium waste in ceramic materials [8].

Comprehensive studies on zircon ($ZrSiO_4$) have revealed that the metamictization process can be decomposed into three successive stages [7].

(1) At low degree of damage the material is essentially crystalline: α-particles produce isolated defects, and α-recoil nuclei produce a few isolated amorphous embryos surrounded by the crystalline matrix, which is expanded as a consequence of the deformation produced by the defects.

(2) In an intermediate regime there is an increasing content of aperiodic regions in the material with amorphous zones forming an interconnected network, and the volume swelling reaches its maximum value.

(3) In the last stage of the amorphization process the long-range order is lost, but a short-range order persists, with nanometric crystalline domains embedded in an amorphous matrix.

Different models have been proposed for describing the metamictization process, either as due to an accumulation of point defects [9], or as an interface controlled effect [10], or as resulting from a direct amorphization in the simple or multiple cascades following the recoil of the nuclei [11, 12]. The three-stage evolution was recently interpreted in terms of successive percolation regimes and transitions [13]. Although the combined picture resulting from these models seems to be consistent with the experimental observations, it does not provide a clear insight into the internal forces acting at the crystalline–metamict transformation and on its thermodynamic nature. The aim of the present work is to describe a possible mechanism for this transformation in the framework of an extension of the Lifshitz–Dzialoshinskii approach to transitions to inhomogeneous crystal phases [14, 15]. The new feature in this extension is that the free-energy density associated with the crystalline–metamict transformation has a *local gauge structure*, which is required to compensate the spatial variation of the translational symmetry in the partially or fully amorphized structure. In this context we propose

(1) that the different stages of the radiation induced transformation correspond to *equilibrium internally stressed states*, i.e., are associated with minima of a free-energy functional, in which the elastic distortions induced via the creation of amorphous regions are compensated by an internal stress-field deriving from a unique stress-field potential, and

(2) that the transformation dynamics can be depicted in terms of nucleation of amorphous embryos with a growth of the amorphous regions via a merging process involving a specific critical size.

2. Theoretical approach to the crystal-to-metamict transformation

Let us formulate our approach considering for simplicity that the order parameter associated with the crystal–metamict transformation has two components $\eta = \rho\, e^{i\varphi}$ and $\eta^* = \rho\, e^{-i\varphi}$ associated with the two-branch star of a critical wavevector $\pm\mathbf{k}_c$, where the amplitude ρ and phase φ depend on the space coordinates x_i ($i = 1, 2, 3$). Following the Lifshitz–Dzialoshinskii model [14, 15], summarized in the appendix, the equilibrium distribution of the order-parameter should correspond to the minima of a free-energy functional $F = \int \phi\, dV$ where the sum runs over the volume of the sample, the free-energy density ϕ depending on the order-parameter components and their spatial derivatives. In the standard model, which aims to describe an aperiodic modulation of the crystal lattice, the order-parameter usually transforms as a single irreducible representation of the crystal space-group, associated

with a constant critical wavevector $\mathbf{k}_c = \mathbf{k}_o$. At variance, we will assume that *the critical wavevector associated with the transition to the metamict state is space dependent* $\mathbf{k}_c = \mathbf{k}(x_i)$ and may vary continuously or discontinuously from one to another region of the damaged crystalline structure. Such an assumption is well adapted to the different stages of the metamictization process. It allows us to describe the last stage of the process in which a short-range order persists [16], meaning that one has a preservation of the translation order in localized nanometre-sized crystallites, which may vary from one crystallite to another. In the intermediate stage, where the crystalline and amorphous regions percolate, a spatial dependence of the wavevector is able to reflect the increasing content of aperiodic regions, having different densities, which vary as a function of the self-irradiation dose [17], and the formation of interconnected networks of amorphous regions. In the initial stage, the wavevector remains constant in the crystalline matrix with localized discontinuities at the forming amorphous domains. It has to be emphasized that in contrast to phase transitions to homogeneous phases where the spectra of values of the order-parameter components and critical wavevectors are discrete, at transitions to inhomogeneous phases the equilibrium values of $\eta(x_i)$ and $\mathbf{k}(x_i)$ form continuous spectra, as is seen for example in the vibrational spectra of titanite [18]. Therefore, neighbouring localized regions are induced by an infinite continuous set of irreducible representations of the 'parent' crystal space-group, which are indexed by the continuous spectrum of $\mathbf{k}(x_i)$.

Under the effect of the primitive translations $\mathbf{t}_j$ ($j = 1, 2, 3$) of the crystalline phase the order-parameter components transform as

$$\widehat{t}_j\eta = e^{i\mathbf{k}\cdot\mathbf{t}_j}\eta \qquad \text{and} \qquad \widehat{t}_j\eta^* = e^{-i\mathbf{k}\cdot\mathbf{t}_j}\eta^* \tag{1}$$

where the $\widehat{t}_j$ are the primitive translation operators. Writing $\mathbf{k}(x_i) = \mu_j(x_i)\mathbf{t}_j^*$, where the $\mathbf{t}_j^*$ are the reciprocal lattice translations, one has $\mathbf{k} \cdot \mathbf{t}_j = 2\pi\mu_j$ and equation (1) becomes

$$\widehat{t}_j\eta = e^{2i\pi\mu_j}\eta \qquad \text{and} \qquad \widehat{t}_j\eta^* = e^{-2i\pi\mu_j}\eta^*. \tag{2}$$

In contrast to η and η^*, the spatial derivatives $\frac{\partial\eta}{\partial x_i}$ and $\frac{\partial\eta*}{\partial x_i}$ *are not* eigenfunctions of the translation operators since one has $\widehat{t}_j\left(\frac{\partial\eta}{\partial x_i}\right) = 2i\pi\frac{\partial\mu_j}{\partial x_i}e^{2i\pi\mu_j}\eta + e^{2i\pi\mu_j}\frac{\partial\eta}{\partial x_i}$ and $\widehat{t}_j\left(\frac{\partial\eta^*}{\partial x_i}\right) = -2i\pi\frac{\partial\mu_j}{\partial x_i}e^{-2i\pi\mu_j}\eta^* + e^{-2i\pi\mu_j}\frac{\partial\eta^*}{\partial x_i}$. Diagonalizing these expressions one can introduce as eigenfunctions of the $\widehat{t}_j$ the covariant derivatives:

$$D_i\eta = \frac{\partial\eta}{\partial x_i} - i\gamma\sum_j A_{ji}\eta \qquad \text{and} \qquad D_i^\bullet\eta^* = \frac{\partial\eta^*}{\partial x_i} + i\gamma\sum_j A_{ji}\eta^* \tag{3}$$

where the γA_{ji} are the components of a second rank tensor representing a compensating field potential (a *gauge-field potential* in the sense given by Bogoliubov and Shirokov [19]), which transforms under the $\hat{t}_j$ as

$$\widehat{t}_j(\gamma A_{ji}) = \gamma A_{ji} + 2\pi\frac{\partial\mu_j}{\partial x_i} \qquad \text{and} \qquad \widehat{t}_j(A_{lj}) = \gamma A_{lj} \qquad (l \neq j) \tag{4}$$

with γ representing a coupling constant between the order parameter and the compensating field components. One can verify that $\widehat{t}_j D_i\eta = e^{2i\pi\mu_j}D_i\eta$ and $\widehat{t}_j D_i^\bullet\eta^* = e^{-2i\pi\mu_j}D_i^\bullet\eta^*$. Accordingly, the free-energy density ϕ has a *local gauge structure* defined by $\eta \rightarrow \eta\,e^{2i\pi\mu_j}$, $\eta^* \rightarrow \eta^*e^{-2i\pi\mu_j}$, and can be constructed from the gauge invariant quantities $\eta\eta^*, \eta D_i^\bullet\eta^*, \eta^* D_i\eta$, and $D_i\eta D_k^\bullet\eta^*$. The equilibrium states are obtained by minimizing the free energy F. Since one has from equation (3) $\frac{\partial\phi}{\partial D_i\eta} = \frac{\partial\phi}{\partial\left(\frac{\partial\eta}{\partial x_i}\right)}$, $\frac{\partial\phi}{\partial D_i^\bullet\eta^*} = \frac{\partial\phi}{\partial\left(\frac{\partial\eta^*}{\partial x_i}\right)}$ and

$\frac{\partial \phi}{\partial A_{ji}} = \frac{\partial \phi}{\partial D_i \eta}(-i\eta) + \frac{\partial \phi}{\partial D_i^* \eta^*}(i\eta^*)$, the minimization of F can be performed independently with respect to the A_{ji} components. The corresponding Euler–Lagrange equations read

$$\frac{\partial \phi}{\partial A_{ji}} = \varepsilon_{ikl} \frac{\partial}{\partial x_k}\left(\frac{\partial \phi}{\partial \sigma_{jl}}\right) \tag{5}$$

where ε_{ikl} is the antisymmetric permutation tensor and

$$\sigma_{jl} = -\varepsilon_{ikl} \frac{\partial A_{jl}}{\partial x_k}. \tag{6}$$

On the other hand, since ϕ is invariant by the $\widehat{t}_j$ operators, it follows from Noether's theorem that the first integrals

$$\alpha_{ji} = i\eta^* \frac{\partial \phi}{\partial \left(\frac{\partial \eta^*}{\partial x_i}\right)} - i\eta \frac{\partial \phi}{\partial \left(\frac{\partial \eta}{\partial x_i}\right)} = \frac{\partial \phi}{\partial A_{ji}} \tag{7}$$

are conserved and obey the continuity equation:

$$\frac{\partial \alpha_{ji}}{\partial x_i} = 0. \tag{8}$$

Introducing the conjugate quantity $\omega_{jl} = \frac{\partial \phi}{\partial \sigma_{jl}}$, the equation of state (5) takes the form

$$\alpha_{ji} = \varepsilon_{ikl} \frac{\partial \omega_{jl}}{\partial x_k} \tag{9}$$

which can be identified as the Kröner equation [20] relating the dislocation density tensor components α_{ji} and the components of the distortion tensor [21] ω_{jl}. Therefore, the conjugate components σ_{ji} correspond to the internal stress tensor [22], and one can infer from equation (6) the physical meaning of the compensating field potential, which represents an *internal stress-field potential*. In summary, when assuming a space dependence of the order parameter *and* of the critical wavevector associated with the transition from a crystalline state to a state having no long-range order but a local translational order, the minimization of the transition free energy F with respect to the internal stress-field results in an equation of state relating the stationary density of dislocations and the distortion field. The corresponding equilibrium state equation (8) expresses the conservation of the dislocation flux, and from equation (6) one has $\frac{\partial \sigma_{ji}}{\partial x_i} = 0$, which corresponds to the zero divergence of the internal stress-field. Accordingly, the different stages of the metamictization process are interpreted as a succession of internally stressed equilibrium states where the internal stress-field compensates the space dependence of the order-parameter gradient and critical wavevector. The amorphous domains (in the early stage), localized crystallites (in the final stage), or the assembly of crystalline and amorphous regions (in the intermediate regime), create a distortion field and a dislocation field which are locally balanced by a stress-field deriving from a unique stress-field potential. The description of the transformation as a *reversible* succession of equilibrium states is consistent with the 'memory effect' reported by Geisler [23] on partially metamict zircon, which is found to recover epitaxially its crystalline form by isothermal annealing, in a three stage recovery process. Note that this is not in contradiction with the intuitive idea that irradiation drives the initial system far from its initial equilibrium crystalline state, i.e., the amorphization can be viewed as a slowly driven system due to stress accumulation, the successive equilibrium states undergoing a continuous evolution towards lower minima of the free energy. Our results show that *local equilibrium* is realized in each stage of the metamictization process. Along the same line the dislocation field assumed in our description may be observable in localized regions of the sample, a property which is not contradicted by the low dislocation densities observed at the microscopic scale in zircon and other metamict materials.

3. The nucleation and growth process

The preceding picture provides only a continuum description of the internal forces acting in the system, and it has to be complemented by a description of the nucleation and growth process of the amorphous regions in the crystalline matrix. Referring to the observations in zircon [13, 17, 24, 25] of amorphous nanodomains (embryos) created within the cascades caused by α-recoil nuclei, one can assume that these embryos serve for a swelling of the amorphous regions via a merging and impingement process of the recoil-produced cascades. Note that this mechanism is different from the conventional growth process described, for example, at martensitic transformations, in which a fraction of the martensitic embryos increase cooperatively in size on approaching the critical temperature. The free energy of the defected crystal has the general form [26]

$$F(\eta, \eta^*, u_{ik}) = \int [\phi(\eta, \eta^*, D_i\eta, D_i^\bullet\eta^*) + a(\eta\eta^*)^2 u_{ii} + f_{el}(u_{ik})]\,dV \qquad (10)$$

where ϕ is the free-energy density discussed above, the u_{ik} are the strain components induced by the embryos, $(\eta\eta^*)^2\, u_{ii}$ is the striction coupling between the order parameter and the dilatation produced by the embryos, and $f_{el}(u_{ik}) = C_{iklm}u_{ik}u_{lm}/2$ is the corresponding elastic energy. For the purpose of our demonstration one can express ϕ in the simple form[1]

$$\phi = \frac{\alpha}{2}\eta\eta^* + \frac{\beta}{4}(\eta\eta^*)^2 + \cdots + \frac{\delta}{2}D_i\eta D_i^\bullet\eta^* \qquad (11)$$

where $\beta, \delta\ldots$ are constant coefficients, and $\alpha = \alpha_0(c_0 - c)$, c representing the concentration of amorphous embryos and c_0 a critical concentration. Replacing $D_i\eta$ and $D_i^\bullet\eta*$ by their expressions and using the notation $\eta_1 = \rho\cos\varphi$, $\eta_2 = \rho\sin\varphi$, ϕ takes the form

$$\phi = \frac{\tilde\alpha}{2}(\eta_1^2 + \eta_2^2) + \frac{\beta}{4}(\eta_1^2 + \eta_2^2)^2 + \cdots + \frac{\delta}{2}[(\nabla\eta_1)^2 + (\nabla\eta_2)^2] \qquad (12)$$

with $\tilde\alpha = \alpha - \delta\gamma\sum_j A_{ji}\frac{\partial\varphi}{\partial x_j} + \frac{\delta}{2}\big(\gamma\sum_j A_{ji}\big)^2$. The equations minimizing F are

$$\delta\Delta\eta_i - (\tilde\alpha + 2au_{ii})\eta_i = \beta\eta_i(\eta_1^2 + \eta_2^2) + \cdots \qquad (i = 1, 2), \qquad \text{and} \qquad \frac{\partial\sigma_{ik}}{\partial x_k} = 0 \quad (13)$$

where $\sigma_{ik} = C_{iklm}u_{lm} + a(\eta_1^2 + \eta_2^2)\delta_{ik}$ is the internal stress-tensor. For each isolated embryo the system of equations (13) possesses two solutions: (1) $\eta_1 = \eta_2 = 0$, $u_{ij} = u_{ij}^0(x_k)$,s where the strain-field induced by the embryo obeys the condition $u_{ij}^0(\infty) = 0$; (2) $\eta_1 = \eta_2 = \eta(x_k) \neq 0$, $u_{ij} = u_{ij}^0(x_k) + O(\eta^2)$, with $\eta(\infty) = 0$. The two solutions describe stable states for different *positive* values of $\tilde\alpha$. In order to determine the value $\tilde\alpha^*$ corresponding to the bifurcation from one solution to another [27] one has to linearize equations (13) around $\eta = 0$. This yields the auxiliary Schrödinger-type equation:

$$\delta\Delta\psi_* = [\tilde\alpha^* + 2au_{ii}^0(x_k)]\psi_* \qquad (14)$$

where ψ_* is the eigenfunction associated with the eigenvalue $\tilde\alpha^*$ of its basic state. For $\tilde\alpha \leqslant \tilde\alpha^*$ the second solution branches off the first one and one can write [26, 27]

$$\eta = \xi\psi_*(x_k) + O(\xi^2), \qquad \text{and}$$

$$u_{ij}(x_k) = u_{ij}^0(x_k) + \frac{a\xi^2}{(2\pi)^3}\int G_{ik}(\mathbf{k})k_jk_k Q(\mathbf{k})\,e^{i\mathbf{k}\cdot\mathbf{r}}\,d^3k \qquad (15)$$

where ξ is a normalized amplitude. The last term in equation (15) is a Fourier transform in $\mathbf{k}$-space, representing the contribution of the strain induced by the embryo. $G_{ik}(\mathbf{k})$ is the Fourier

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[1] Other invariants such as $\eta D_i^\bullet\eta^* \pm \eta^* D_i\eta$, or $(D_i\eta D_i^\bullet\eta^*)^2$ are neglected.

transform of the elastic Green function, and $Q(\mathbf{k}) = \int [\psi_*(x_k)]^2 \, e^{-i\mathbf{k}\cdot\mathbf{r}} \, d^3r$. The asymptotic solution of equations (15) for the order parameter is $\eta \approx \xi \, e^{-\frac{r}{r_N}}$, where r_N is the *radius* of the amorphous region given by

$$r_N = \left(\frac{\delta}{\tilde{\alpha}^*}\right)^{1/2} + r_0 = \left(\frac{\delta}{\alpha_0}\right)^{1/2} (c_c - c_N)^{-1/2} + r_0 \qquad (16)$$

where r_0 is the initial radius of the amorphous embryo, c_N is the concentration at which the embryo nucleates, and $c_c = c_0 + \frac{\delta\gamma}{\alpha_0} \sum_j A_{ji} \frac{\partial\varphi}{\partial x_j} - \frac{\delta}{2\alpha_0}(\gamma \sum_j A_{ji})^2$ is a critical concentration, depending on the internal stress-field potential, *at which the merging of the amorphous embryos occurs critically*. Equation (16) shows that for large values of $c_N - c_c$, i.e. for a small concentration of isolated embryos, the radius of the amorphous regions remains almost equal to r_0, whereas when c_N approaches c_c r_N tends to diverge with the critical exponent $-1/2$. The existence of a critical concentration of defects initiating the swelling of the amorphous embryos was reported by Weber *et al* in ion-beam irradiated zircon [28]. The explicit form of the local dilatation, created by the embryos above the critical concentration c_c, is given by the trace $u_{ii}(x_k)$, which can be worked out using the second equation (15) with $j = i$. It depends on the shape assumed for the amorphous embryos and on the symmetry of the surrounding crystal lattice [29].

4. Summary and conclusion

In summary, it has been shown that the phenomenological description of the crystalline-to-metamict transformation can be inserted into an generalization of the Lifshitz–Dzialoshinskii approach to the formation of inhomogeneous crystal phases. The description provides an insight into the forces which ensure the equilibrium of the damaged crystal in the different stages of the amorphization process, and into the nucleation and growth mechanism of the amorphous regions. Only one critical regime is found in this mechanism, which corresponds to the first percolation point described for the metamictization process in zircon [30]. The absence of a second critical regime in our description is due to the simplified form assumed for ϕ in equation (11). Including in ϕ the gauge invariant term $\eta D_i^{\bullet} \eta^* - \eta^* D_i \eta$ gives rise, at higher concentration $c_1 > c_c$ of amorphous embryos, to a *second* transition, analogous to the lock-in transition found in incommensurate systems [15], that should correspond to the second percolation point reported for the metamictization of zircon [30]. Note that the flexibility of our approach is compatible with a variety of processes leading to the metamict state, in agreement with the different behaviours reported in materials other than zircon. In particular, the influence of the temperature T can be taken into account by assuming a linear dependence of α_0 on T. The application of external stresses results in a renormalization of the elastic constants in $f_{el}(u_{ik})$ and of the striction coupling in equation (10).

In conclusion, let us stress that the theoretical formalism introduced in the present work applies more generally to any transformation leading to a state in which the long-range order is lost but a local translational order is preserved. One should note the formal analogy existing between

(1) our description of the metamict state, as resulting from the breaking of the translational symmetry of the crystalline phase, in which the local spatial dependence of the order-parameter gradient is compensated by an internal stress-field potential, and
(2) the phenomenological description of the superconducting state [31], resulting from the breaking of the gauge symmetry of the normal phase, where the local time-dependence of the order-parameter phase gradient is compensated by the electromagnetic potential.

Thus, under the condition that the time and space variables are exchanged, the gauge invariance of the free-energy density ϕ associated with the crystalline–metamict transformation is equivalent to the gauge invariance of the Lagrangian density in the action formalism of superconductivity [32]. A number of properties of the metamict state, which are beyond the scope of the present article, can be inferred from this analogy, for example the existence of elastic vortices in the dislocation flux [33], reflected by the curl-like structure of equation (9), that should induce density fluctuations in the amorphous material, as observed in zircon [24].

Acknowledgments

Appendix

In the Landau theory of phase transitions the increment of the probability density describing the change in the spatial distribution of atoms in a crystal undergoing a phase transition reads

$$\delta\rho^k(\vec{r}) = \sum_{i=1}^{p} \eta_i^k(\vec{r})\Psi_i^k(\vec{r}) \tag{A.1}$$

where the $\Psi_i^k(\vec{r})$ are normalized functions transforming as the basis of an irreducible representation (IR) of the parent high-symmetry space group G_0, the k-vector corresponding to a point of the Brillouin zone of the parent structure. The stable states of the crystal are characterized by definite values η_i^k of the order-parameter components associated with an absolute minimum of the transition free energy $F(\eta_i^k)$, expressed as a polynomial expansion of the η_i^k. Lifshitz has noted [14] that when the transition leads to an inhomogeneous low-symmetry structure, the preceding condition does not test completely the stability of the phases at the macroscopic level, as F has also to be minimum with respect to variations of the η_i^k which vary from one point to another in the low-symmetry phase. As a consequence, instead of a homogeneous polynomial free energy F one has to consider a functional $F = \int \phi(\eta_i^k(\vec{r}))\,d\vec{r}$, where the sum runs over the volume of the system, the free-energy density ϕ depending on the η_i^k and on their spatial derivatives. If the η_i^k vary slowly at the microscopic level Lifshitz shows that to determine the eventual stability of an inhomogeneous structure it is sufficient to consider antisymmetric gradient invariants of the form $\eta_i^k \frac{\partial \eta_j^k}{\partial x_l} - \eta_j^k \frac{\partial \eta_i^k}{\partial x_l}$, with $i \neq j$, $(x_l = x, y, z)$. If such terms (called Lifshitz invariants) are allowed by the parent symmetry, an inhomogeneous phase may stabilize across a second-order transition from the parent phase. These ideas were used by Dzialoshinskii [15] in the description of modulated (incommensurate) structures. In these structures the transition wavevector k varies with temperature (or concentration or pressure) within an interval Δk, and therefore one has to consider the variation of $\delta\rho^k(\vec{r})$ in this interval. Dzialoshinskii proposed to fix the value of $k = k_c$ where k_c represents a rational vector associated with the lock-in transition, which generally takes place below the range of stability of the incommensurate phase. The physical assumption underlying this choice is that, within the incommensurate phase, the system is asymptotically governed by a periodic potential possessing the symmetry of the lock-in phase, i.e., the incommensurate phase is treated as a spatial modulation of the lock-in structure. As a consequence the Ψ_i^k functions form the basis of a single IR associated with k_c, but the order-parameter components $\eta_i^{k_c}$ appear

as slowly modulated functions of the actual order-parameter components η_i^k. As a result the free energy F is a continuous function of k through the sequence of parent-incommensurate-lock-in phases and is written as the sum over the volume of the system of a local density which depends on the $\eta_i^{k_c}$ and on their derivatives with respect to the space coordinates.

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