

Nonlinear elasticity of the phase-field crystal model from the renormalization group

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(Received 15 October 2009; published 30 December 2009)

The rotationally covariant renormalization group equations of motion for the density wave amplitudes in the phase field crystal model are shown to follow from a dynamical equation driven by an effective free energy density that we derive. We show that this free energy can be written purely as a function of the strain tensor and thence derive the corresponding equations governing the nonlinear elastic response.

DOI: [10.1103/PhysRevE.80.065105](https://doi.org/10.1103/PhysRevE.80.065105)

PACS number(s): 62.20.F-, 05.10.Cc, 61.72.Cc, 81.15.Aa

Multiple scale approaches to materials pattern formation are essential to account for the variety of material structures that emerge on different scales, and the dependence of materials properties on structures at more than one scale [1]. However, a coarse-grained description describing the pattern dynamics is not in itself sufficient, for example, in fracture mechanics, one would like to be able to predict the response of real, heterogeneous materials to deformations, and to couple the resulting elasticity theory to discrete, atomistic descriptions at small scales. In short, the challenge of materials modeling requires not just a coarse-grained description of the underlying atomistic dynamics, but a multiresolution approach that can locally adapt to capture the appropriate fast space and time scales.

Recently, we presented a proof of concept realization of such a calculation [2–4], using a minimal model of a crystalline material called the phase field crystal (PFC) model [5–7]. The PFC model evolves the local density field through a conservation law governed by a free-energy functional that penalizes departures from a perfectly periodic ground state [8]. The resulting density field retains the crystallographic and elastic properties of the material, in contrast to conventional phase field model descriptions of materials, and appears to give a realistic account of a variety of phenomena, including multicrystalline solidification [6], elasticity [5,6,9], defect dynamics [6,10,11], epitaxial growth, as well as crack and fracture dynamics [6]. Our proof-of-concept calculation consisted of two distinct steps: (i) a renormalization-group calculation for the rotationally covariant equation of motion for the amplitudes of density waves corresponding to the lattice periodicity, and (ii) the numerical solution of the resulting equations using adaptive mesh refinement techniques [4]. In two dimensions, this calculation was three orders of magnitude faster than straightforward integration of the PFC equations.

In this Rapid Communication, we show that an additional level of coarse-graining can be performed on the complex amplitude representation, leading to a theory of nonlinear elasticity valid at system wide or macroscopic scales. Our construction is of interest methodologically; instead of postulating a form for the strain energy based on symmetry and phenomenology [12,13], we derive it from a more microscopic model—in this case, the PFC model. Our work provides a theoretical connection to microscopic structures and opens up the possibility of deriving other forms of strain energy by similar approaches. Finally, in varying our level of

description from the PFC model to the complex amplitude representation to the strain energy formulation, we obtain the basis for a multiple scale approach to materials properties, accessing the realm of continuum mechanics from a density functional theory at atomic scales. This approach could be applicable to interpreting recent experiments on atomic force microscope nanoindentation of graphene [14,15], complementing theoretical approaches using tight-binding methods [16].

Model. The PFC model is defined by the free-energy density [5,6],

$$f = \frac{\rho}{2}(1 + \nabla^2)^2 \rho + \frac{r}{2}\rho^2 + \rho^4, \quad (1)$$

where $\rho(\vec{x}, t)$ is the phase field, or the order parameter. The dynamics is conservative and dissipative, given by

$$\frac{\partial \rho}{\partial t} = \nabla^2 \left(\frac{\delta F}{\delta \rho} \right), \quad (2)$$

where $F = \int d^2x f(x)$ is the total free energy of the system, taken here to be two dimensional. Thermal noise is ignored in our discussion here, but could be included if desired. Generally, it will not be important at system scales. There are three phases in this model, namely, uniform, stripe, and triangular. The ground state in the triangular phase can be written in the single-mode approximation by

$$\rho_{tri}(\vec{x}) = A \sum_{j=1}^3 (e^{i\vec{k}_j \cdot \vec{x}} + e^{-i\vec{k}_j \cdot \vec{x}}) + \rho_0, \quad (3)$$

where ρ_0 is the average density, A is the constant amplitude and

$$\vec{k}_1 = \hat{y}, \quad \vec{k}_2 = \frac{\sqrt{3}}{2}\hat{x} - \frac{1}{2}\hat{y}, \quad \vec{k}_3 = \frac{-\sqrt{3}}{2}\hat{x} - \frac{1}{2}\hat{y}, \quad (4)$$

are the three lattice vectors. Goldenfeld *et al.* showed [2,3] that instead of using $\rho(\vec{x}, t)$ as the dynamical variable, it is more efficient to generalize Eq. (3) and promote the constant amplitudes A to 3 slowly varying complex amplitudes $A_j(\vec{x}, t)$ and treat the $A_j(\vec{x}, t)$ as dynamical variables. Using renormalization-group techniques, they showed that the rotationally covariant equation of motion for $A_1(x, t)$ is given by [2,3]

$$\begin{aligned} \frac{\partial A_1}{\partial t} = & (1 - L_1)(\Gamma - L_1^2)A_1 - 6\rho_0 A_2^* A_3^* \\ & - 3A_1(|A_1|^2 + 2|A_2|^2 + 2|A_3|^2) + \dots, \end{aligned} \quad (5)$$

where $\Gamma \equiv -r - 3\rho_0^2$ and $L_1 \equiv \nabla^2 + 2i\vec{k}_1 \cdot \nabla$ is a rotationally covariant operator, and small nonlinear gradient terms [3] are not written out explicitly. The corresponding equations for $A_{2,3}(x, t)$ can be written down from the appropriate permutations.

Derivation of the amplitude equations without using renormalization group. The first attempt to derive the free energy in this representation is to note that Eq. (5), ignoring the nonlinear gradient terms, can be written as

$$\frac{\partial A_j}{\partial t} = - \frac{\delta F_{dyn}[A_j(x, t)]}{\delta A_j^*}, \quad (6)$$

where the free-energy F_{dyn} is given by the free-energy density,

$$\begin{aligned} f_{dyn} = & - \sum_{j=1}^3 A_j^*(1 - L_j)(\Gamma - L_j^2)A_j + 3 \sum_{j,k=1}^3 |A_j|^2 |A_k|^2 - \frac{3}{2} \sum_{j=1}^3 |A_j|^4 \\ & + 6\rho_0(A_1 A_2 A_3 + A_1^* A_2^* A_3^*). \end{aligned} \quad (7)$$

The dynamics given by Eq. (6) is purely dissipative, as opposed to the density-conserving dynamics in the original PFC equation, Eq. (2).

To resolve this conundrum, we note that mass conservation should only be exhibited in the dynamical equation of motion and not be represented in the equilibrium free energy. Thus, the correct way to derive the free energy in the complex amplitude representation is to derive it from the original PFC free energy, Eq. (1). The easiest way to do that is to substitute the ansatz, Eq. (3), into the free energy, Eq. (1). The first term of Eq. (1) can then be computed by using the identity

$$(1 + \nabla^2)\rho = \sum_{j=1}^3 (e^{i\vec{k}_j \cdot \vec{x}} L_j A_j + \text{c.c.}) + \rho_0, \quad (8)$$

where c.c. stands for complex conjugate. By performing an integration by parts, we find

$$\frac{\rho}{2}(1 + \nabla^2)^2 \rho = \frac{1}{2}[(1 + \nabla^2)\rho]^2, \quad (9)$$

$$= \sum_{j=1}^3 A_j^* L_j^2 A_j, \quad (10)$$

where in the last line constants and terms with the rapidly oscillating factor $\exp(i\vec{k}_j \cdot \vec{x})$ are neglected. We can neglect the oscillating terms because the complex amplitudes, A_j , are slowly varying on that scale, so the terms cancel themselves upon integration over space. Other terms in the free energy can be transformed in a similar fashion. The resulting free energy is given by

$$\begin{aligned} f_{amp} = & - \sum_{j=1}^3 A_j^*(\Gamma - L_j^2)A_j + 3 \sum_{j,l=1}^3 |A_j|^2 |A_l|^2 - \frac{3}{2} \sum_{j=1}^3 |A_j|^4 \\ & + 6\rho_0(A_1 A_2 A_3 + A_1^* A_2^* A_3^*). \end{aligned} \quad (11)$$

Note that this free energy is different from Eq. (7). The $1 - L_j$ operator in the first term in Eq. (7) is absent here. This is to be expected because this operator arises from the conservative Laplacian in the dynamical equation, and according to our discussion above, it should not appear in the free energy. The transformation of the dynamical equation, Eq. (2), into the complex amplitude representation can be performed by observing that for any function, $f(x)$, the identity,

$$\int_{-\infty}^{\infty} d^2x [(L_j - 1)f(x)] e^{i\vec{k}_j \cdot \vec{x}} \equiv 0, \quad (12)$$

holds, a counterpart to the identity,

$$\int_{-\infty}^{\infty} d^2x \nabla^2 g(x) = 0, \quad (13)$$

for any function $g(x)$. In fact, if we define $g(x) = f(x) e^{i\vec{k}_j \cdot \vec{x}}$, Eq. (13) implies Eq. (12). This shows that when we make the change of variables from the density, ρ , to the complex amplitudes, A_j , the Laplacian in the conservative dynamical equation has also to be transformed to $L_j - 1$. We, thus, arrive at the equation of motion

$$\frac{dA_j}{dt} = (L_j - 1) \frac{\delta F_{amp}}{\delta A_j^*}, \quad (14)$$

which, when written out explicitly, is

$$\begin{aligned} \frac{dA_1}{dt} = & (1 - L_1)[(\Gamma - L_1^2)A_1 - 6\rho_0 A_2^* A_3^* - 3A_1(|A_1|^2 + 2|A_2|^2 \\ & + 2|A_3|^2)] \end{aligned} \quad (15)$$

with appropriate permutations for $A_{2,3}(x, t)$. By construction, these equations conserve the density of the system.

Equation (15) is exactly the same as Eq. (5) with all the nonlinear gradient terms included. This derivation shows that the inclusion of the nonlinear gradient terms is crucial for density conservation, and that all those terms can actually be written in the condensed form of Eq. (15). Note, however, that our derivation does not explicitly use a renormalization-group argument, but follows from the integral identity, Eq. (12). The connection with the renormalization-group approach arises from the coarse-graining assumption after Eq. (10), where we asserted that the rapid oscillation averages to zero.

Nonlinear elasticity. The goal of this section is to derive the free energy as a function of the strain tensor, when the PFC crystal is deformed under a general deformation,

$$x'_m = F_{mn} x_n, \quad (16)$$

where F_{mn} is the deformation gradient. Einstein's summation convention is used throughout, except for the index j in \vec{k}_j , A_j , and L_j . In general, the deformation gradient can be written as [13]

$$F_{mn} = R_{mp} U_{pn}, \quad (17)$$

where R_{mp} is a pure rotation matrix and U_{pn} is a positive-definite, pure deformation matrix. Since our system is rotationally covariant, we expect that the free energy should only depend on the function $U^T U$, where U^T is the transpose of the matrix U .

Under the deformation, Eq. (16), the complex amplitudes transform as

$$A_j \rightarrow A'_j = A e^{ik_{jm} D_{mn} x_n}, \quad (18)$$

where we defined $D_{mn} \equiv R_{mk} U_{kn} - \delta_{mn}$ and assumed that $|A_j| = A$ for all j , where A is a constant. k_{jm} is the m -th component of the vector \vec{k}_j . Because the deformation gradient only enter the complex amplitude through its phase, describing local density deformations, the only relevant terms in the free energy are the gradient terms given by

$$E \equiv \sum_{j=1}^3 A_j^* L_j^2 A_j. \quad (19)$$

Other terms in the free energy, Eq. (11), only contribute when we minimize the free energy with respect to A at the end of the calculation. By using Eq. (18) and differentiating, we obtain

$$L_j A_j = (-k_{jm} k_{jn} R_{mp} R_{na} U_{pq} U_{aq} + 1) A_j. \quad (20)$$

Apply L_j again and substitute the result into Eq. (19) to obtain

$$E = A^2 (E_1 - 2E_2 + 3), \quad (21)$$

where

$$E_1 = \left(\sum_{j=1}^3 k_{jm} k_{jn} k_{ju} k_{jv} \right) F_{mq} F_{nq} F_{uw} F_{vw}, \quad (22)$$

and,

$$E_2 = \left(\sum_{j=1}^3 k_{jm} k_{jn} \right) F_{mq} F_{nq}. \quad (23)$$

The rest of the derivation concerns the evaluation of E_1 and E_2 . We first evaluate E_2 . By using the definition of \vec{k}_j from Eq. (4), we obtain

$$k_{1m} k_{1n} = \delta_{my} \delta_{ny} = \delta_{mn} \delta_{my}, \quad (24)$$

$$k_{2m} k_{2n} = \frac{3}{4} \delta_{mx} \delta_{nx} + \frac{1}{4} \delta_{my} - \frac{\sqrt{3}}{4} (\delta_{mx} \delta_{ny} + \delta_{nx} + \delta_{my}), \quad (25)$$

and,

$$k_{3m} k_{3n} = \frac{3}{4} \delta_{mx} \delta_{nx} + \frac{1}{4} \delta_{my} + \frac{\sqrt{3}}{4} (\delta_{mx} \delta_{ny} + \delta_{nx} + \delta_{my}). \quad (26)$$

Combining these three equations we have

$$\sum_{j=1}^3 k_{jm} k_{jn} = \frac{3}{2} \delta_{mn} (\delta_{mx} + \delta_{my}) = \frac{3}{2} \delta_{mn}. \quad (27)$$

Thus, E_2 is given by

$$E_2 = \frac{3}{2} F_{pq} F_{pq} = \frac{3}{2} U_{pq} U_{pq} = \frac{3}{2} \text{Tr}[U^T U], \quad (28)$$

where we used the property of the rotation matrix, that $R_{im} R_{jm} = R_{mi} R_{mj} = \delta_{ij}$, $\text{Tr}[A]$ and A^T are the trace and transpose of the matrix A , respectively. The evaluation of E_1 is more involved. We note that

$$k_{1m} k_{1n} k_{1u} k_{1v} = \delta_{mn} \delta_{nu} \delta_{uv} \delta_{my}, \quad (29)$$

and observe that the term

$$k_{2m} k_{2n} k_{2u} k_{2v} + k_{3m} k_{3n} k_{3u} k_{3v} \quad (30)$$

is equal to the term

$$2 \times [k_{2m} k_{2n} k_{2u} k_{2v} + \text{terms with positive coefficients}]. \quad (31)$$

By exploring this relation and using the definition of \vec{k}_j , Eq. (4), we obtain,

$$\begin{aligned} \sum_{j=1}^3 k_{jm} k_{jn} k_{ju} k_{jv} &= \frac{3}{8} (\Delta_{xxyy} + \Delta_{yyxx} + \Delta_{xyxy}) \\ &+ \frac{3}{8} (\Delta_{xyyx} + \Delta_{yxxy} + \Delta_{xyyx}) \\ &+ \frac{9}{8} (\Delta_{xxxx} + \Delta_{yyyy}), \end{aligned} \quad (32)$$

where we defined $\Delta_{abcd} \equiv \delta_{ma} \delta_{nb} \delta_{uc} \delta_{vd}$ for convenience. By this, we have

$$\begin{aligned} &\left(\sum_{j=1}^3 k_{jm} k_{jn} k_{ju} k_{jv} \right) R_{mp} R_{na} R_{us} R_{vt} \\ &= \frac{3}{8} [R_{xp} R_{xa} R_{ys} R_{yt} + R_{yp} R_{ya} R_{xs} R_{xt}] \\ &+ \frac{3}{8} [R_{xp} R_{ya} R_{xs} R_{yt} + R_{yp} R_{xa} R_{ys} R_{xt}] \\ &+ \frac{3}{8} [R_{xp} R_{ya} R_{ys} R_{xt} + R_{yp} R_{xa} R_{xs} R_{yt}] \\ &+ \frac{9}{8} [R_{xp} R_{xa} R_{xs} R_{xt} + R_{yp} R_{ya} R_{ys} R_{yt}]. \end{aligned} \quad (33)$$

To evaluate this expression, we note that we can combine terms judiciously. For example, by using the property of the rotation matrix, we obtain

$$R_{xp} R_{xa} R_{ys} R_{yt} + R_{xp} R_{xa} R_{xs} R_{xt} = R_{xp} R_{xa} \delta_{st}, \quad (34)$$

and,

$$R_{yp} R_{ya} R_{xs} R_{xt} + R_{yp} R_{ya} R_{ys} R_{yt} = R_{yp} R_{ya} \delta_{st}. \quad (35)$$

The sum of Eqs. (34) and (35) then give

$$R_{xp}R_{xa}R_{ys}R_{yt} + R_{xp}R_{xa}R_{xs}R_{xt} + R_{yp}R_{ya}R_{xs}R_{xt} + R_{yp}R_{ya}R_{ys}R_{yt} = \delta_{st}\delta_{ap}. \quad (36)$$

Repeating this for all the terms in Eq. (33), we obtain

$$\left(\sum_{j=1}^3 k_{jm}k_{jn}k_{ju}k_{jv} \right) R_{mp}R_{na}R_{us}R_{vt} = \frac{3}{8} [\delta_{ap}\delta_{st} + \delta_{sp}\delta_{at} + \delta_{as}\delta_{pt}]. \quad (37)$$

Substituting into Eq. (22), and then into Eq. (21), we obtain $E=3A^2\Delta$, where

$$\Delta = \frac{1}{8} \{ [\text{Tr}(U^T U)]^2 + 2 \text{Tr}(U^T U U^T U) \} - \text{Tr}(U^T U) + 1. \quad (38)$$

By using the relation $U_{mp}^T U_{pn} = \delta_{mn} + 2u_{mn}$, where $u_{ij} \equiv [\partial_i u_j + \partial_j u_i + (\partial_k u_i)(\partial_k u_j)]/2$ is the nonlinear strain tensor, we obtain

$$\Delta = \left(\frac{3}{2} u_{xx}^2 + \frac{3}{2} u_{yy}^2 + u_{xy}^2 + u_{yx}^2 + u_{xx}u_{yy} \right). \quad (39)$$

Finally, we substitute back into the free energy, Eq. (11), and minimize the whole expression with respect to A to obtain

$$A(\Delta) = \frac{1}{5} \left(-\rho_0 \pm \frac{1}{3} \sqrt{9\rho_0^2 + 15(\Gamma - \Delta)} \right), \quad (40)$$

which gives the free-energy density as

$$f(\Delta) = \frac{45}{2} A^4(\Delta) + 12\rho_0 A^3(\Delta) - 3(\Gamma - \Delta) A^2(\Delta). \quad (41)$$

This formula completely defines the elastic properties of the PFC model, and provides a starting point for conventional continuum mechanical applications of nonlinear elasticity theory.

Nonlinear elastic theories are coarse-grained models up to the level of the continuum, and so do not explicitly include defect structures; our results are most useful for understanding large deformation behavior, twinning, and phase transitions in ordered materials. However, for plastic deformations, dislocations need to be included, usually by postulating a free energy with contributions from nonlinear elasticity [such as Eq. (41)], vacancies and dislocations [17,18]. While this can yield useful insights, the most suitable level of description for probing the multiscale phenomena accompanying plastic flow remains the PFC equations taking into account vacancies [19] or their rotationally covariant renormalized counterparts (if only dislocations are present). We will report on this approach in a future publication [20].

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