Long-scale deformations of resonant three-dimensional patterns and the structure of confined dislocations

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It is shown that three-dimensional patterns dominated by triplet interactions in the vicinity of a symmetry-breaking bifurcation point acquire rigidity owing to resonant interactions between constituent modes. Phase equations determining the response of octahedral, tetrahedral, and icosahedral structures to long-scale perturbations are derived and analyzed. The nonisotropic long-scale response spectrum is universal, being dependent on the crystalline structure only. It is shown that the resonance condition causes confinement of dislocations in a number of constituent modes to a common dislocation line. The phase equations are applied to compute the far field structure of the dislocations.

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I. INTRODUCTION

Symmetry-breaking transitions from a homogeneous to a patterned or crystalline state in both equilibrium and nonequilibrium systems can be described within a common framework as long as possible nonpotential effects far from equilibrium are suppressed (as they usually are in the vicinity of a primary symmetry-breaking bifurcation point). This common ground is provided by the Landau mean field theory [1] describing the state of broken symmetry as a superposition of a number of excited modes that appear formally as degenerate neutrally stable eigenmodes of linearized macroscopic equations, and may admit, in various contexts, different physical interpretations (e.g., density waves in the equilibrium theory).

Since a symmetry-breaking transition in an isotropic system implies a preferred wavelength but no preferred direction, an indefinite number of modes may be excited, with the wave vectors $\mathbf{k}_j$ having the same absolute value (that can be taken as unity) but arbitrary direction. The emerging pattern or crystalline structure is selected by nonlinear interactions. Landau [1] was the first to notice a special role of lowest-order (triplet) interactions among modes forming an equilateral triangle. These interactions are *resonant*, i.e., phase dependent, and cause the phases of the interacting modes to be locked in such a way that the interaction is destabilizing. Unless forbidden by symmetry, resonant interactions are prevalent sufficiently close to a symmetry-breaking transition, and are responsible for its subcritical (first-order) character.

In a two-dimensional setting (most common in nonequilibrium systems), triplet interactions favor a hexagonal pattern, which is selected, in a generic case, sufficiently close to the bifurcation point. In three dimensions, triplet interactions favor a combination of modes forming one of the regular polyhedra with triangular faces—tetrahedron, octahedron, or icosahedron [2]. The former two correspond to the face-centered and body-centered cubic crystal lattices, and the latter to a quasicrystalline structure.

The firm ground of the theory sketched above ends when we approach the question of distortions of regular structures formed due to resonant interactions. Distorted patterns can be described (following the approach common in the theory of nonequilibrium symmetry-breaking transitions [3]) by adding to the Landau energy "kinetic" terms involving spatial derivatives of the amplitudes, and accounting for the dependence of energy on the wave number. An immediate problem is that such terms are strongly anisotropic for each particular mode, since the wave number is influenced, in the leading order, only by perturbations directed along the corresponding wave vector. The resulting lack of "rigidity" in the transverse direction is usually remedied [3] by introducing terms containing higher-order derivatives.

The aim of this communication is to describe long-scale deformations and dislocations in three-dimensional structures, or *resonant crystals* dominated by triplet interactions. We shall see that an additional rigidity contributed by resonant interactions makes terms containing higher-order transverse derivatives superfluous, and compute explicitly eigenvalues characterizing an anisotropic response of a resonant crystal to long-scale perturbations. Another resonant effect, which has been described before for two-dimensional patterns [4,5], but leads to a far larger variety of structures in three dimensions, is the *confinement* of dislocations in interacting modes.

II. AMPLITUDE EQUATIONS

A standard analysis in the vicinity of a symmetry-breaking transition is based on the Landau expansion of the interaction potential

$$V = -\mu \sum_{j=1}^{N} a_j a_j^* - \sum a_i a_j a_k + \sum \nu_{ijkl} a_i a_j a_k a_l + \cdots . \tag{1}$$

where the summation is carried out over all closed polygons formed by the wave vectors of extant modes. Presenting the complex amplitudes in the polar form $a_j = \rho_j \exp(i\theta_j)$ reduces Eq. (1) to

$$V = -\mu \sum_{j=1}^{N} \rho_j^2 - \sum \rho_i \rho_j \rho_k \cos \Theta_{ijk} + \sum \nu_{ijkl} \rho_i \rho_j \rho_k \rho_l \cos \Theta_{ijkl} + \cdots, \quad (2)$$

where $\Theta_{ijk} = \theta_i + \theta_j + \theta_k$ and Θ_{ijkl} are sums of phases around the respective triangles or rhombi. The homogeneous state is linearly unstable when the coefficient at the quadratic term μ is positive. The cubic interaction is universal, and therefore all cubic terms come with the same coefficient that can be taken as unity. The cubic terms are phase dependent, and the potential is at minimum when the sum of phases of modes forming an equilateral triangle is zero (modulo 2π). When phases adjust in this way, the triplet interactions are destabilizing; this is the reason why symmetry-breaking transitions leading to the formation of stationary patterns are always subcritical and of the first order. The emerging pattern is stabilized by quatric terms, which depend on angles between interacting modes and are specific for particular systems. The quatric terms are phase dependent only when the rhombus formed by the respective wave vectors is nonplanar.

Structures distorted on a scale far exceeding the basic wavelength can be described by allowing for amplitude modulation on an extended scale. The appropriate form of the energy functional (retaining the leading terms due to extensional modulations only) is

$$\mathcal{E} = \int \left[\sum_{j=1}^{N} (\mathbf{k}_j \cdot \nabla) a_j^* (\mathbf{k}_j \cdot \nabla) a_j + V \right] d\mathbf{x}, \quad (3)$$

where the potential V is defined by Eq. (1). The evolution equations can be derived from Eq. (3) assuming the gradient dynamics

$$\frac{\partial a_j}{\partial t} = -\frac{\delta \mathcal{E}}{\delta a_j^*} = (\mathbf{k}_j \cdot \nabla)^2 a_j - \frac{\partial V}{\partial a_j^*}. \quad (4)$$

The evolution equations can be formally derived through the usual scheme of small-amplitude expansion near a symmetry-breaking bifurcation point if the coefficient defining the strength of triplet interactions is of $O(\epsilon)$, so that destabilizing resonant triplet interactions are balanced by quadruplet interactions when $a_j = O(\epsilon)$. Both the energy functional (3) and the evolution equation (4) are valid at distances large compared to unity (i.e., to the basic characteristic scale of the pattern); therefore $\mathbf{x}$ has to be understood here as an *extended* coordinate, so that the spatial derivatives are scaled as $\nabla = O(\epsilon)$.

The amplitude equations in this form were originally used for the analysis of modulated hexagonal patterns [6]. In some later works, the simple differential term was replaced by Newell-Whitehead-Segel (NWS) operators [3] of the type $\left(\frac{\partial}{\partial x_j} - \frac{i}{2} \frac{\partial^2}{\partial y_j^2} \right)^2$, where x_j and y_j are extended coordinates directed, respectively, along and across the wave vector $\mathbf{k}_j$, and scaled as $\partial/\partial x_j = O(\epsilon)$, $\partial/\partial y_j = O(\sqrt{\epsilon})$. When resonant interactions are absent, a shorter characteristic scale for the transverse coordinate is necessary to prevent discontinuities in the transverse directions. Resonant interactions make, however, terms with higher-order transverse derivatives superfluous, since discontinuities in any constitutive mode would violate the constraint on the sum of phases Θ_{ijk}, and therefore must be suppressed. This has been demonstrated recently in a study of defects in hexagonal patterns [5] where smooth solutions were obtained using the amplitude equation in the form (4). In the following, I shall apply this equation to distorted three-dimensional resonant crystals.

III. RIGIDITY OF A RESONANT STRUCTURE

Any single mode obeying Eq. (4) would suffer a zigzag instability if resonant interaction terms were absent. Due to the triplet interactions, different modes are, however, tied up by resonant conditions that force the sum of phases to be zero (modulo 2π). If each interacting mode is identified with an edge (pq) directed from the pth to the qth vertex of a polyhedron, the resonance condition is satisfied automatically by presenting the respective amplitude as $a_{pq} = b_p^* b_q$. The corresponding phase is expressed as $\theta_{pq} = \theta_q - \theta_p$. Clearly, the phases sum up to zero around any closed polygon.

The phases θ_p associated with vertices correspond to translational degrees of freedom of the crystal. The octahedral and tetrahedral structures in the $\mathbf{k}$ space both contain six modes and their complex conjugates; in each case (constraining the field to be real), there are exactly three translational modes corresponding to the phases associated with three nonantipodal vertices of the octahedron, or with four vertices of the tetrahedron, minus an irrelevant constant shift of all four phases. The 15-mode structure forming an icosahedron in the $\mathbf{k}$ space has six arbitrary phases, and their shift causes a restructuring of a quasicrystalline structure that cannot be reduced to a mere translation.

In the following, I shall consider perturbations of the translational modes that occur on a scale large compared to the characteristic scale of Eq. (4). The phase equations valid on the extended scale are derived by presenting the complex amplitudes in the polar form, separating the real and imaginary parts, and rescaling the coordinates by another small factor, say, δ. For a pattern with a prevailing "optimal" wavelength ($|\mathbf{k}| = 1$) and equal amplitudes of all modes in the undistorted state, the long-scale phase equations derived in this way are

$$\frac{\partial \theta_{pq}}{\partial t} = \delta^2 (\mathbf{k}_{pq} \cdot \nabla)^2 \theta_{pq} - \sum_r \sin \Theta_{pqr} + \sum_{r,s} \nu_{pqrs} \sin \Theta_{pqrs}. \quad (5)$$

Here the modes are marked by the respective vertices as

stated above, and $\Theta_{pqr} = \theta_{pq} + \theta_{qr} + \theta_{rp}$, Θ_{pqrs} are sums of phases around the appropriate triangles or rhombi; the constant undistorted amplitude is rescaled to unity.

Under conditions when the triplet interactions are prevailing, the total phases Θ_{pqr} relax to zero on the $O(1)$ time scale; at longer times, these phase combinations remain of $O(\delta^2)$. The residual phases are not necessarily reducible to differences between the vertex phases. They can be, however, eliminated by summing up Eq. (5) around each vertex. In this sum, each polygon appears twice, being traversed in opposite directions, and therefore the corresponding phases cancel. In this way, one obtains the evolution equations for the translational phases θ_p valid on an extended time scale $T = \delta^{-2}t$:

$$\frac{\partial}{\partial T}\left(n\theta_p - \sum_{(pq)}\theta_q\right) = \sum_{(pq)}\left[(\boldsymbol{l}_p - \boldsymbol{l}_q)\cdot\boldsymbol{\nabla}\right]^2(\theta_p - \theta_q), \tag{6}$$

where the summation is carried over all n edges converging at a particular vertex, and $\boldsymbol{l}_p$ are vertex positions defining the wave vectors $\mathbf{k}_{pq} = \boldsymbol{l}_q - \boldsymbol{l}_p$. The response to long-scale perturbations is determined by the eigenvalues of Eq. (6). Generally, the response is nonisotropic, and the eigenfunctions mix stretching, bending, and twisting modes. The sum of the eigenvalues is, however, isotropic for all regular structures.

For the octahedron, Eq. (6) can be rewritten in an explicit form after taking into account that, due to the reality condition, the phases associated with opposite vertices are antisymmetric. This cancels the off-diagonal terms in the left-hand side of Eq. (6). Using the Cartesian frame spanned by the vectors $\boldsymbol{l}_p$ yields

$$\frac{\partial\theta_p}{\partial T} = \frac{1}{4}\sum_{k\neq p}\frac{\partial^2\theta_p}{\partial x_k^2} + \frac{1}{2}\frac{\partial}{\partial x_p}\sum_k\frac{\partial\theta_k}{\partial x_k}, \quad p = 1,2,3. \tag{7}$$

If the phase perturbations are presented as $\theta_p \propto \exp(i\mathbf{q}\cdot\mathbf{x})$, the eigenvalues of Eq. (7), or, generally, of Eq. (6), can be interpreted as phase diffusivities measuring the rate of relaxation to the regular structure. The number of eigenvalues—3—corresponds to the number of independent elastic moduli of a structure with cubic symmetry. The dependence of the "leading" eigenvalue λ (that with the smallest absolute value) on the direction of the perturbation wave vector is shown in Fig. 1(a). The structure is most rigid in the direction of vertices, where the leading eigenvalue is twice degenerate, and has the maximal absolute value $|\lambda| = \frac{1}{4}$. A lower local maximum with $|\lambda| = \frac{1}{6}$, again doubly degenerate, is observed in the directions pointing to the centers of faces. The softest response ($|\lambda| = \frac{1}{8}$) is in the midedge direction. At all extremal points (but not in other directions) the eigenfunctions corresponding to the leading eigenvalue are divergenceless, i.e., do not contain a stretching component.

For the tetrahedron, the number of variables in Eq. (6) can be reduced by 1 by introducing the Cartesian phase vector $\boldsymbol{\Theta} = \sum_p \boldsymbol{l}_p\theta_p$. The dependence of the leading eigenvalue on the direction of the perturbation wave vector is plotted in Fig. 1(b) using the polar coordinate sys-

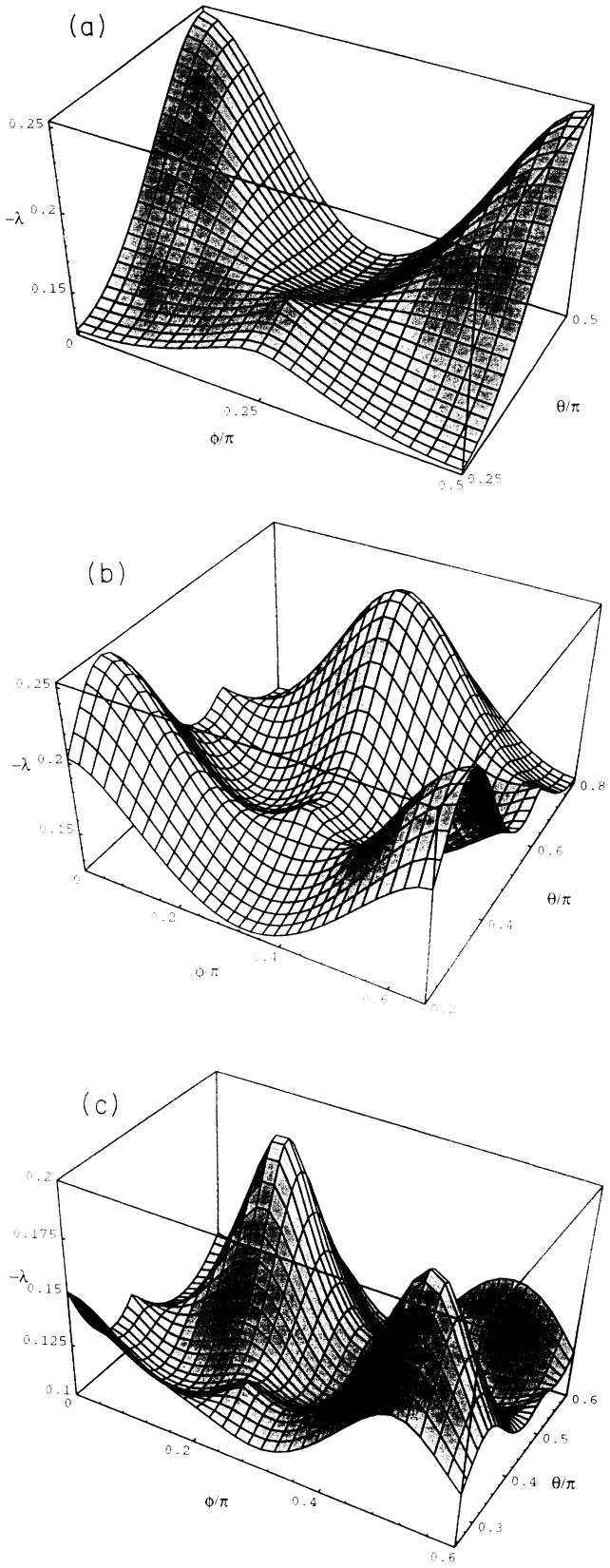

FIG. 1. The dependence of the leading eigenvalue λ on the direction of the perturbation wave vector (parametrized by spherical angles θ, ϕ) for the octahedral (a), tetrahedral (b), and icosahedral (c) structures.

tem with one of the vertices at the north pole. The profile is remarkably similar to the octahedral case, even though the identification of soft and rigid directions with the **k**-space structure is different. The structure is most rigid ($|\lambda| = \frac{1}{4}$) in six midedge directions, and most soft ($|\lambda| = \frac{1}{8}$) in twelve directions lying on the equatorial circle associated with each vertex at meridional positions $\phi = (2k+1)\pi/3$, $k = 0,\ldots,5$. Local maxima with $|\lambda| = \frac{1}{6}$ are observed at eight directions pointing to the vertices and their antipodal points (the eigenvalues are degenerate there). The number of extrema, the degeneracy, and even the values of λ are the same as for the octahedral structure, and the eigenfunctions corresponding to the leading eigenvalue are again divergenceless at the extremal points.

The Cartesian form of the phase equation (6) for the tetrahedral structure is, of course, not symmetric. A symmetric equation can be obtained by retaining all four phases and introducing four differential operators $\partial_p = \boldsymbol{l}_p \cdot \nabla$ associated with the four vertices. Since one of the phases is superfluous, one can impose the gauge condition $\sum_p \theta_p = 0$; with this choice, the left-hand side of Eq. (6) becomes diagonal. Using the identity $\sum_p \partial_p \equiv 0$, Eq. (6) is presented in the form

$$\frac{\partial \theta_p}{\partial T} = \frac{1}{4}\left(\partial_p^2 + \frac{1}{4}\sum_k \partial_k^2\right)\theta_p - \frac{1}{4}\sum_k (\partial_p - \partial_k)^2\,\theta_k, \qquad p = 1,\ldots,4. \quad (8)$$

Alternative forms of this equation can be obtained by applying again the above gauge condition.

For the icosahedral structure, the explicit form of Eq. (6) is too long to be reproduced here. Computation shows that this structure, similar to the octahedral one, is most rigid ($|\lambda| = 0.105\,146$) in the direction of vertices, and most soft ($|\lambda| = 0.067\,281$) in the midedge directions. A representative part of the relief of the leading eigenvalue is shown in Fig. 1(c).

In two dimensions, applying the same procedure to the hexagonal structure formed by a single equilateral triangle in **k** space shows that the response is isotropic, with the leading eigenvalue $|\lambda| = \frac{1}{4}$. The corresponding three-dimensional prismatic structure lacks, however, rigidity altogether in the third dimension.

IV. CONFINEMENT OF DISLOCATIONS

Equation (6) can be applied to the study of the far field structure of dislocations in resonant crystals. It is known that in two dimensions dislocations of opposite signs in any two modes of the resonant triplet tend to bind together, thereby insuring that the resonance condition be satisfied [4]. This localized structure corresponds to a penta-hepta defect in the hexagonal pattern. The far field structure of this defect is not quite simple, and lacks the circular symmetry of the phase field of a free defect [5].

A similar confinement of dislocations in constituent modes occurs in three dimensions. It is easy to see that circulation of a vertex phase around any axis does not violate the resonance conditions. At the same time, this circulation generates a dislocation in all modes connected to this vertex. We conclude that dislocations in three, four, or five modes, respectively, in the tetrahedral, octahedral, and icosahedral structures, should be confined to a common dislocation line.

The algebra of dislocations follows immediately from the structure of the **k**-space polyhedra. Geometrically, combinations of different dislocations can be understood by associating with each edge an arrow pointing, say, from the vertex when the respective vertex phase has a positive circulation, and towards the vertex when the circulation is negative. The mode corresponding to a directed edge acquires a topological charge. If there is circulation of more than one vertex phase, oppositely directed arrows cancel, while those directed in the same way add up to double the charge.

For the octahedral structure, one can see following the above rules and taking into account the reality condition that a collision of dislocations associated with any two different vertices should, irrespective of their signs, double the topological charge of some modes. In the tetrahedral case, dislocations associated with two vertices, being combined, give a structure identical to that formed by combining the dislocations associated with the other two vertices.

The far field deformation induced by a dislocation line is computed most easily for the octahedral structure where Eq. (7) applies. A simple static solution of this equation, corresponding to a screw dislocation line along the z axis, is

$$\theta_x = \theta_y = 0,\ \theta_z = \phi, \quad (9)$$

where the vertex phases θ_p are marked by the respective coordinate axes, and ϕ is the polar angle around the z axis. The phase gradients are singular on the dislocation line where the amplitudes of modes carrying phase circulation must vanish.

Another static solution, corresponding to an edge dislocation line along the z axis with the Burgers vector directed along the x axis, lacks circular symmetry. Though no closed solution is found in this case, the phase field can be expressed in the form of a Fourier series

$$\theta_x = \phi + \tfrac{1}{6}\sin 2\phi - \tfrac{1}{24}\sin 4\phi - \tfrac{1}{216}\sin 6\phi + \cdots,$$
$$\theta_y = -\tfrac{1}{3}\cos 2\phi + \tfrac{1}{108}\cos 6\phi + \cdots, \quad \theta_z = 0. \quad (10)$$

The series is converging rapidly, and, beyond the quadrupole term, higher harmonics are negligible. For the tetrahedral structure, the far field of both edge and screw dislocation lines is nonisotropic.

V. CONCLUSIONS

Equations (7) and (8) can be viewed as the simplest macroscopic models of deformations and dislocations in a crystalline solid. One can also consider conservative ver-

sions of these equations with the second rather than first time derivative in the left-hand side. The latter model replaces the gradient relaxation by an elastic response, and the eigenvalues computed above are reinterpreted as elastic constants.

Although the phase models are macroscopic and continuous, they reflect the anisotropy of the underlying crystalline structure. It is notable that the rigidity of the structure in both gradient and conservative models is a consequence of resonant interactions only, as any mode by itself is not resistant to bending.

The long-scale response is universal, being dependent on the crystalline structure only, and insensitive to specific values of mode interaction coefficients ν_{ijkl}. The latter, of course, determine the choice of the structure in the first place; they should also influence the spectrum on shorter wavelengths when changes of real amplitudes become significant.

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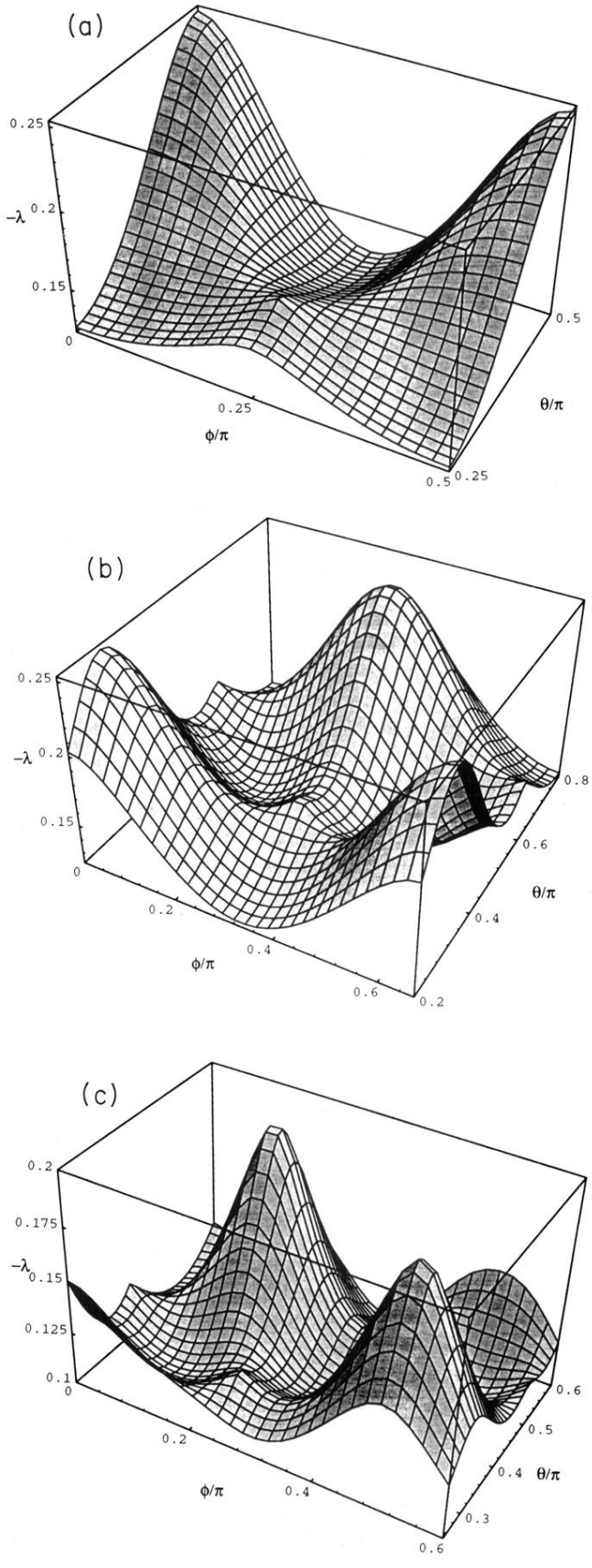

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