Nonconvex Interactions and the Occurrence of Modulated Phases

Mario Marchand, Kevin Hood, and Alain Caillé

Département de Physique et Centre de Recherche en Physique du Solide, Université de Sherbrooke, Sherbrooke, Québec, Canada, J1K 2R1

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Nonconvex interparticle interactions are proposed as a mechanism for the occurrence of modulated order in condensed matter. This is examined by application of a recent numerical algorithm to two microscopic models in which frustration is present only if the interparticle interactions are nonconvex. A devil's staircase is found when the particles, in the ground state, make use only of the convex part of the interaction potential, whereas both first- and second-order transitions are found when the region of nonconvexity is felt by some of the particles.

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The nature of the ground states of frustrated microscopic models is relevant to the understanding of many different physical systems exhibiting periodically modulated structures.[1] The vast majority of theoretical studies have, so far, been limited to convex (positive curvature) interparticle interactions. However, it is now recognized that certain classes of condensed-matter systems do experience nonconvex interparticle interactions. A well-known classical example is the oscillating (Ruderman-Kittel-Kasuya-Yosida) exchange interaction between localized spins in a metal. Recently,[2] it has been shown that magnetoelastic coupling leads to an effective double-well interparticle interaction. More generally, and relevant to ferroelectricity, Villain and Gordon[3] have shown that oscillating interactions can be mediated through elastic strains and other harmonic fields.

The effects of nonconvexity are presently far from well understood.[4] This is not surprising since theoretical approaches to problems of nonconvex interparticle interactions have so far been limited to models where either competing periodicities or competing interactions (that lead to modulated ground states) are present even when the interparticle interactions are convex. For example, if we replace the interaction terms of the models studied by Aubry, Fesser, and Bishop,[5] Banerjea and Taylor,[6] and Yokai, Tang, and Chou[7] by convex harmonic interactions, we recover the well-known Frenkel-Kontorova model.[1,8] In this Letter, in order to focus only on nonconvex effects, we report our findings on two microscopic models having nonconvex interactions and for which the ground state is always uniform (unmodulated) when these interactions are convex. Furthermore, these models are representative of certain kinds of interactions that can occur in real solids. In fact, as shown in Ref. 2, they are related to certain magnetoelastic problems (more details will be given in a later publication). Our major conclusion is that nonconvex interparticle interactions alone can be responsible for the occurrence of periodically modulated structures when the substrate potential is convex and the kind of phase transitions present strongly depends on whether or not the particles experience the nonconvex part of the interaction.

We consider the one-dimensional (1D) Hamiltonian

$$H=\sum_n[V(u_n)+W(u_{n+1}-u_n)], \qquad (1)$$

where u_n is the displacement of nth particle with respect to some reference position, here assumed to be a regular 1D lattice of equally spaced points. The external one-particle potential has the form

$$V(x)=\tfrac{1}{2}Kx^2 \quad (K>0). \qquad (2)$$

Physically, $V(u_n)$ is the local potential experienced by a particle in the nth cell as a result of the interaction with a background of rigid atoms. If we restrict ourselves to small deviations from symmetric equilibrium positions, $V(u_n)$ is written as in (2). Note that although unbounded potentials such as (2) are useful for describing structural phase transitions,[9] they are not appropriate for materials where particles can jump from one unit cell to another. It is straightforward to show that for convex $V(x)$ and for $W(x)$ having a finite lower bound, the average lattice distortion $\langle u_{n+1}-u_n\rangle$ is zero in the ground state and that the only ground state is the uniform state when $W(x)$ is also convex. *Hence, for convex $V(x)$, the existence of any modulated ground state is solely due to the presence of a nonconvex $W(x)$.* The two models used for the nonconvex interparticle interaction are

$$W(x)=(x-\gamma)^2-|x-\gamma| \quad \text{(model 1)}, \qquad (3)$$

$$W(x)=-\tfrac{1}{2}(x-\gamma)^2+\tfrac{1}{4}(x-\gamma)^4 \quad \text{(model 2)}. \qquad (4)$$

Model 1 is typical of the $T=0$ double-quadratic-well effective interparticle interaction, which arises in the 1D magnetoelastic problem[2] involving n-component classical spins, $\mathbf{S}_n$, coupled to nearest neighbors through an exchange integral which varies linearly with interparticle spacing. The exchange energy of the bonds is proportional to $-|u_{n+1}-u_n-\gamma|$ since, at $T=0$, the classical

n vectors, $\mathbf{S}_n$, align themselves either ferromagnetically or antiferromagnetically with their first nearest neighbors, depending on the sign of the exchange integral between them. $-\gamma$ is the ratio of the exchange integral J_0 to its gradient $-J_1$ evaluated at $(u_{n+1}-u_n)=0$. The first term, $(x-\gamma)^2$, comes from the first-neighbor elastic interaction. At finite temperatures, it has been found[2] that the effective interaction between the u_n has the form of an analytic double-well potential if the spin-exchange interactions are 1D. In this case model 2 serves, under certain conditions, as an approximation to this effective, temperature-dependent, interparticle interaction. More generally, (4) represents the first terms of a Taylor expansion of a more general nonconvex interparticle interaction as has been shown for $TlHF_2$.[10] Notice that the particular choice of the coefficients in (2), (3), and (4) can always be achieved by an appropriate scaling of lengths and energies.

The vast majority of methods used to find the ground states are based upon searches among all extremal solutions satisfying $\partial H/\partial u_n=0$. It becomes extremely difficult to proceed by this method when nonconvex interactions are present since then the resulting two-dimensional map is not single valued. Recently, major progress was made in the theory of modulated structures. An algorithm valid for nonconvex interparticle interactions, that focuses directly on the ground state, was proposed by Griffiths and Chou.[11,12] The ground state of (1) is obtained, in the thermodynamic limit $N\rightarrow\infty$, by solution of the nonlinear eigenvalue equation

$$\lambda+R(u')=V(u')=\min_u[W(u'-u)+R(u)]. \quad (5)$$

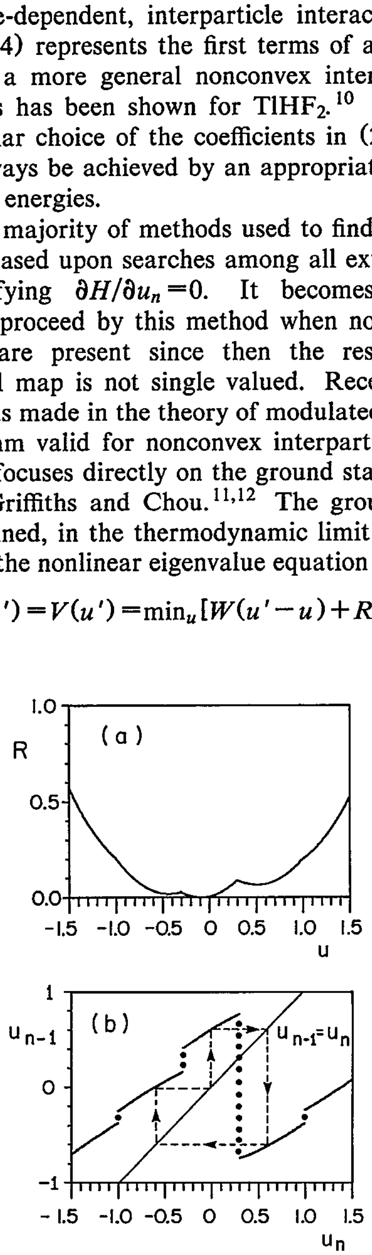

FIG. 1. (a) The effective potential $R(u)$ and (b) the associated map (full line) $M(u_n)=u_{n-1}$ of model 2 for $K=0.5$ and $\gamma=0.33$. Also shown in (b) are the discontinuities (dotted lines), the line $u_{n-1}=u_n$, and the limit cycle of period 3.

The solution $R(u)$ is called an effective potential and λ is the average energy per particle in the ground state. It has been shown[12] that a continuous solution, $R(u)$, always exists and that the corresponding λ is unique. The mapping

$$u=M(u'), \quad (6)$$

obtained by a search for the u which, for a given u', minimizes the right-hand side of (5), is used to generate the ground-state configuration. To solve (5), we use a grid of two hundred (or more) equally spaced points in an interval around $u=0$ and apply the right-hand side of (5) to functions defined on these points.[11,12] The sequence of iterations $R^{(n)}$ is stopped when $R^{(n+1)}$ and $R^{(n)}$ differ only by the constant λ within a chosen precision.[12]

Figure 1(a) is an example of the effective potential, $R(u)$, obtained for model 2. The first derivative of R is discontinuous at the same points where the mapping M, shown in Fig. 1(b), is discontinuous. Also shown in Fig. 1(b) is the limit cycle (of period 3) of the map M. We identify the phases by their m-point limit cycle defining the "winding number," ω, as the number of points of the cycle having $u_{n-1}-u_n\geq 0$ divided by the period m of the cycle

$$\omega=\frac{1}{m}\sum_{n=1}^{m}\theta(u_{n-1}-u_n), \quad (7)$$

where $u_0=u_m$ and $\theta(x)=+1$ for $x\geq 0$ and 0 for $x<0$.

The phase diagrams obtained with this numerical algorithm for models 1 and 2 are shown respectively in Figs. 2 and 3. Since $V(-x)=V(x)$ the phase $\omega=p/r$, for $\gamma>0$, becomes the phase $\omega=(r-p)/r$ when $\gamma\rightarrow-\gamma$. At $K\rightarrow 0+$ the phase $\omega=p/r$ is located at $\gamma=(2p-r)/(2r)$ and $\gamma=(2p-r)/r$ for models 1 and 2, respectively. These phase diagrams are qualitatively different. In particular, the ground state is always uniform for $K>4$ in model 2, whereas no such critical value

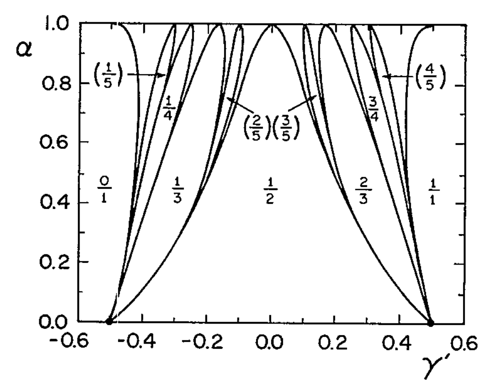

FIG. 2. The phase diagram for model 1. The numbers are values of the winding number ω. The unlabeled regions contain additional commensurate phases. $\alpha=(1+K/4)^{-1}$ and $\gamma'=\gamma/\alpha$.

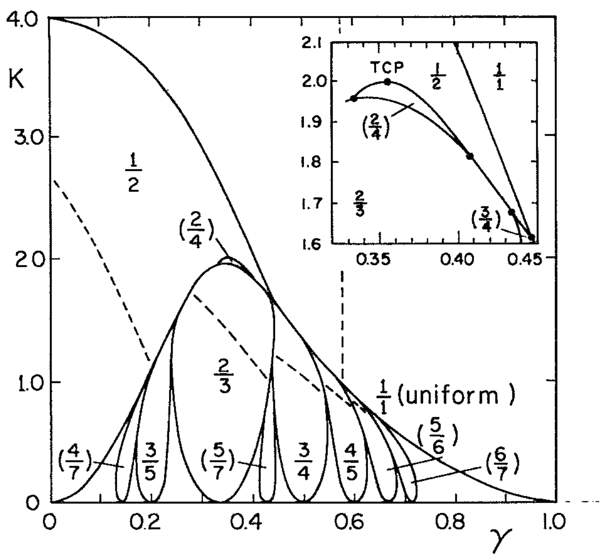

FIG. 3. The phase diagram for model 2. The numbers are values of the winding number ω. The unlabeled regions contain additional commensurate phases. The dashed lines are separation lines defined in the text. Inset: TCP indicates a tricritical point and the other points are triple points.

of K exists in model 1.

Following the notation of Ref. 7, a phase is called "nonconvex" if at least one pair of atoms uses the nonconvex part of $W(x)$; otherwise it is called "convex." A *separation line,* indicated by a dashed line in Fig. 3, separates the nonconvex region from the convex region of the same phase. It is crucial to note that only convex phases are present in model 1 since $W(x)$ is then nonconvex only at the nonanalytic point $x=0$, whereas in model 2 large portions of the phase diagram are filled with nonconvex phases which are located above (to the left of, for the 1/1 phase) the separation lines. Although there is no phase transition when these separation lines are crossed, the type of transition between any two given phases strongly depends on whether or not these phases are convex.

We have found clear indications which strongly suggest that model 1 exhibits a complete devil's staircase[13] even though a rigorous proof is presently not possible with this numerical algorithm. First, it is simple to show that all the extremal configurations are structurally stable. This means that each individual bond can only distort itself discontinuously and therefore hysteresis effects should be present. Second, a phase with winding number $\omega=(p+q)/(r+s)$ is always found [if we use a sufficiently fine grid of points to solve (5)] between phases p/r and q/s. Hence, if there is an infinite number of phases between any two given phases, the sequence of transitions should be continuous although irreversible. This is consistent with Aubry's picture[14] of a complete devil's staircase.

The discontinuous bond distortions are accompanied, in the corresponding magnetoelastic problem, by an inversion of the exchange integral. Indeed, the fraction of antiferromagnetic bonds ω_γ (for positive J_1) in a cycle of period m,

$$\omega_\gamma=\frac{1}{m}\sum_{n=1}^{m}\theta(u_{n-1}-u_n+\gamma), \tag{8}$$

is equal to ω everywhere in the phase diagram (except in the uniform phase where $\omega=1/1$, whereas $\omega_\gamma=1/1$ for $\gamma>0$ and $\omega_\gamma=0/1$ for $\gamma<0$ as shown in Fig. 2). As indicated above, model 1 is representative of the magnetoelastic problem *for any n-component classical spin model.* With this in mind, it is instructive to recall the recent results of Ishimura[15] for an effective *Ising* spin Hamiltonian where the elastic variables have been integrated out. Using the method of Bak and Bruinsma,[16] Ishimura has obtained a phase diagram, strikingly similar to our own, and has predicted a complete devil's-staircase behavior for any finite value of K. On the basis of the above observations, this result should be valid for all n-vector models. As $K\to\infty$, the first-neighbor elastic interactions becomes negligible and we recover the magnetoelastic model of De Simone, Stratt, and Tobochnik[17] which is identical to the axial next-nearest-neighbor Ising (ANNNI) model. Hence, the two multiphase points of Fig. 2 are those of the ANNNI model at $T=0$.

The same numerical evidence that strongly supports the existence of a devil's staircase in model 1 is also present in model 2 for transitions between convex phases. For example, between the 3/5 phase (which is convex everywhere) and the convex 2/3 phase, we were always able to find (with a sufficiently fine grid of points) an intervening phase between any two given phases, all of which are convex. Furthermore, in this model, convex phases are structurally stable and therefore hysteresis effects should be present. Hence, in this region of the phase diagram, our results are consistent with a complete devil's-staircase behavior. Although it is impossible to distinguish high-order commensurate states from truly incommensurate states when one uses this numerical algorithm, we do not believe that there exist incommensurate ground states in this model because, in order that hysteresis effects be absent, it is necessary (since V is convex) that some of the particles experience the nonconvex part of W. On the other hand, between nonconvex phases we have observed two different kinds of behavior. First, we have found second-order phase transitions between the nonconvex 1/1 and 1/2 phases where the uniform state is unstable against dimerization along the parabola $K+4(3\gamma^2-1)=0$ and between the phase 1/2 and the (everywhere nonconvex) 2/4 phase[18] at the left of the tricritical point (TCP) (the transition is first order at the right of TCP). Second, first-order transitions and triple points have been found in several places in the phase diagram. Examples of triple points are shown in the inset in Fig. 3 and it is straightforward to show analytically that the transitions between the phases 1/2 and 2/3, 1/2 and 3/4, 2/4 and 2/3, and 2/3 and 3/4 are all first order.

It is obvious that the phase diagram of model 2 is much more complex than that of model 1. In a later publication we shall show, by different methods, that several features similar to the ones found by Yokoi, Tang, and Chou[7] for the chiral *XY* model are present in this model. These are superdegenerate points[19] (where the entropy is infinite) and possibly infinite sequences of first-order transitions between convex and nonconvex phases.

It is clear that nonconvex interparticle potentials alone can lead to frustration and be responsible for the presence of modulated phases. The nature of the transition between any two given phases strongly depends on whether or not these phases are convex. It is also clear that the phase diagrams obtained for a convex $V(x)$ and nonconvex $W(x)$ are qualitatively different from those obtained, for example, by Axel and Aubry[20] for nonconvex $V(x)$ and convex interparticle interactions.

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