Quasicontinuum modeling of short-wave instabilities in crystal lattices

Lev Truskinovsky * Anna Vainchtein †

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Abstract

We propose a new hybrid quasicontinuum model which captures both long and short-wave instabilities of crystal lattices and combines the advantages of weakly nonlocal (gradient) and strongly nonlocal continuum models. To illustrate the idea we consider a simple one-dimensional lattice exhibiting commensurate and incommensurate short-wave instabilities and compute stability limits for the homogeneous phase using both discrete and quasicontinuum models. As we show, the quasicontinuum approximation is capable of reproducing a detailed structure of the discrete stability diagram.

1 Introduction

An important class of lattice instabilities gives rise to displacive phase transitions. Macro or long-wave instabilities, typical for martensites, result in a formation of finite-size domains of a new phase [21, 24]. Microinstabilities, associated with wave lengths comparable with lattice spacing, are responsible for the formation of multi-lattices and modulated "tweed" patterns preceding first-order transitions; in addition to martensites, they have been detected in quartz, high-temperature perovskites, ferroelectrics, ceramics and composite crystals [3, 15, 24].

Both micro and macroinstabilities are visible on the phonon dispersion curves [11, 27]. The high-symmetry phase loses stability when the minimum of the dispersion relation representing frequencies of the normal modes, becomes equal to zero. Such minima often

*Laboratoire de Mechanique des Solides, CNRS-UMR 7649, Ecole Polytechnique, 91128, Palaiseau, France

†Department of Mathematics, University of Pittsburgh, Pittsburgh, PA 15260

occur at special points in the Brillouin zone where the first unstable wave number has rational components producing modulations commensurate with the lattice. The instability with a generic value of the unstable wave number leads to formation of an incommensurate phase [13].

A macroscopic description of lattice instabilities is usually presented in the framework of Landau theory. When the unstable wave number is equal to zero, the order parameter coincides with a component of the macroscopic strain and the continuum description is adequate. If the unstable wave number is different from zero, the macroscopic order parameter is identified with the amplitude of an unstable normal mode; a macroscopic model containing such order parameters (shuffles) effectively averages out modulations at small scales. This may become a problem when a higher resolution is required, as in thin films or near defects.

To preserve the fine structure of the modulations in the continuum setting we propose in this paper a quasicontinuum model which captures both long and short-wave instabilities. The model is obtained by a high-derivative long-wave expansion of the discrete energy and it shares with the discrete model the property that strain is the only order parameter. To make the truncated long-wave expansion well-posed, we extend the polynomial dispersion relation by zero outside the Brillouin zone. The model therefore restricts modulations in the physical space to length scales on the order of or larger than the lattice size. These restriction allows one to avoid the unphysical small-scale oscillations that are an inevitable feature of the straightforward gradient models. As we show, the cut-off makes such gradient model equivalent to a fully nonlocal model with oscillatory kernel.

To illustrate the idea, we consider a prototypical one-dimensional lattice with interactions of up to third nearest neighbors, which is known to be the simplest model exhibiting both commensurate and incommensurate short-wave instabilities (e.g. [10, 14]). The analytical simplicity of the problem allows us to explicitly compute the stability boundaries for a homogeneous phase in both discrete and quasicontinuum models. We show that to capture commensurate microinstabilities in the discrete model, the quasicontinuum approximation must contain at least two competing gradient terms favoring either coarsening or refinement of the microstructure; to predict incommensurate mode of instability, the continuum model must include at least three competing higher gradient terms.

Lattice instabilities have been previously modeled in the framework of discrete [1, 4, 9, 10, 14], higher-gradient [2, 5, 7, 12, 18, 19, 20] and nonlocal continuum [8, 22] models. A general relation between discrete theories and various continuum approximations was studied in [16, 25], to cite just two representative approaches. The new quasicontinuum model presented here combines the convenience of the gradient models at long waves with the physically correct description provided by strongly nonlocal models at small scales. A potential usefulness of this model is suggested by the fact that it captures all relevant instability modes exhibited by a generic discrete model.

2 Discrete model

Consider an infinite chain of interacting particles with the total energy

$$W = \varepsilon \sum_{n=-\infty}^{\infty} \sum_{p=1}^{q} p\phi_p\left(\frac{u_{n+p} - u_n}{p\varepsilon}\right). \tag{1}$$

Here $\phi_p(w)$ is the energy density of an effective spring with reference length $p\varepsilon$ representing interaction of pth nearest neighbors ($1 \le p \le q$) and $u_n(t)$ is the displacement of nth particle. The equilibria in this system can be found from the following infinite system of finite difference equations:

$$\sum_{p=1}^{q}\left[\phi_p'\left(\frac{u_{n+p} - u_n}{p\varepsilon}\right) - \phi_p'\left(\frac{u_n - u_{n-p}}{p\varepsilon}\right)\right] = 0. \tag{2}$$

To access stability of a homogeneous equilibrium state $u_n^0 = n\varepsilon w$, where w is the average strain, we need to introduce perturbations $v_n = u_n - u_n^0$ and study the positive definiteness of the quadratic part of the energy. This leads to the following eigenvalue problem:

$$\omega^2 v_n = \sum_{p=1}^{q} K_p(v_{n+p} - 2v_n + v_{n-p}), \tag{3}$$

where $K_p = \phi_p''(w)/p$ and ω^2 is the square of the characteristic frequency. By representing the normal modes in the form $v_n = \exp(ink)$, where k is a real wave number, we obtain the dispersion relation

$$\omega^2(k) = 4\sum_{p=1}^{q} K_p \sin^2 \frac{pk}{2}. \tag{4}$$

Notice that the modes with $|k| > \pi$ correspond to perturbations whose length scale is smaller than the lattice size which makes them physically irrelevant. Therefore, if the symmetry is taken into account, it suffices to consider the wave numbers in the interval $0 \le k \le \pi$ (Brillouin zone). A uniform deformation is then stable if and only if $\omega^2(k) > 0$ for all $k \in (0, \pi]$.

We obtain the necessary conditions for stability if we require that

$$\frac{d^2\omega^2(0)}{dk^2} > 0 \quad \text{and} \quad \omega^2(\pi) > 0.$$

The first condition,

$$E = \sum_{p=1}^{q} p^2 K_p > 0, \tag{5}$$

means physically that the effective elastic modulus along the homogeneous branch of equilibria is positive; the corresponding eigenmode is infinitely long: $v_n = 1$. The second condition,

$$\sum_{k=1,\, k \text{ odd}}^{q} K_p > 0, \tag{6}$$

is less transparent; the corresponding eigenmode is commensurate and has the smallest possible wave length $v_n = (-1)^n$.

To find stability conditions that are both necessary and sufficient, we need to specify the number of interactions, and the first generic case is $q = 3$ (e.g. [10, 17]). The dispersion relation reads

$$\omega^2(k) = 4\sin^2\frac{k}{2}\left(16K_3\sin^4\frac{k}{2} - 4(K_2 + 6K_3)\sin^2\frac{k}{2} + K_1 + 4K_2 + 9K_3\right) \tag{7}$$

A straightforward analysis of this expression produces the desired necessary and sufficient conditions of stability:

$$\begin{aligned} &K_1 + 4K_2 + 9K_3 > 0 \\ &K_1 + K_3 > 0 \\ &K_2^2 < 4K_3(K_1 + K_2) \quad \text{if} \quad -6 < \frac{K_2}{K_3} < 2 \end{aligned} \tag{8}$$

The first two of these conditions have been already obtained as necessary and can be identified with macroinstability ($k = 0$) and commensurate microinstability ($k = \pi$), respectively. The third mode of instability corresponding to the condition $(8)_3$ is incommensurate with $0 < k = 2\arcsin\sqrt{(K_2 + 6K_3)/(8K_3)} < \pi$. The full stability diagram in the plane of nondimensional parameters

$$\alpha = \frac{K_2}{K_1}, \qquad \beta = \frac{K_3}{K_1} \tag{9}$$

is shown in Figure 1. Notice that at $K_3 \leq 0$ the first two conditions in (8) are both necessary and sufficient.

3 Quasicontinuum approximation

To obtain a higher derivative quasicontinuum approximation of our lattice model we replace the discrete dispersion relation (4) by the first few terms of the Taylor expansion around $k = 0$ (e.g. [1, 17]). In order to capture all three types of instabilities (long-wave,

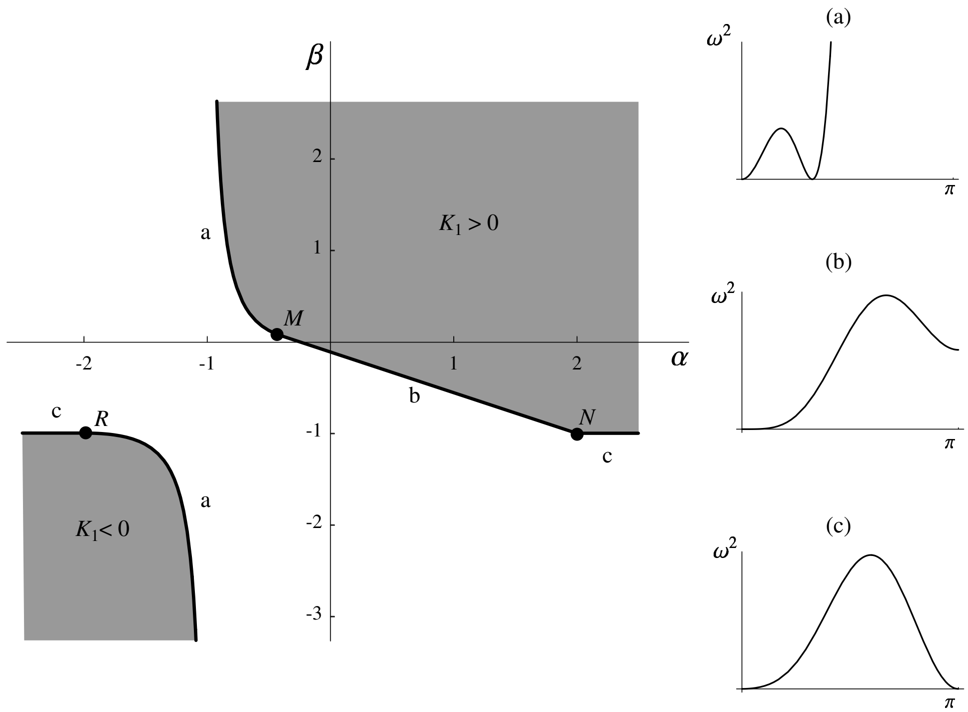

Figure 1: Stability diagram for the discrete model with $q = 3$ and the corresponding dispersion relation: (a) incommensurate microinstability (above $M = (-2/5, 1/15)$ and below $R = (-2, -1)$), $\alpha^2 = 4\beta(1 + \alpha)$; (b) macroinstability ($MN$), $1 + 4\alpha + 9\beta = 0$; (c) commensurate microinstability (to the right of $N = (2, -1)$ and to the left of R), $\beta = -1$. The regions of stability of the trivial solution are shown in gray.

commensurate and incommensurate) exhibited by the discrete model with $q \geq 3$, it is necessary to consider a polynomial expansion of the 6th order:

$$\omega^2(k) \approx k^2(E + A_1 k^2 + A_2 k^4). \tag{10}$$

We select the coefficients by approximating discrete dispersion relation near $k = 0$ to obtain

$$\begin{aligned} E &= K_1 + 4K_2 + 9K_3 \\ A_1 &= -\frac{K_1 + 16K_2 + 81K_3}{12} \\ A_2 &= \frac{K_1 + 64K_2 + 729K_3}{360}. \end{aligned} \tag{11}$$

The quadratic part of the energy corresponding to (10) can be written as

$$W = \frac{1}{2}\int_{-\infty}^{\infty} [Eu_x^2 + A_1\varepsilon^2 u_{xx}^2 + A_2\varepsilon^4 u_{xxx}^2]dx. \tag{12}$$

To access stability of the homogeneous state we need to solve the eigenvalue problem for the sixth-order ODE:

$$\omega^2 v = Ev_{xx} + A_1\varepsilon^2 v_{xxxx} + A_2\varepsilon^4 v_{xxxxxx}. \tag{13}$$

Even without solving this equation one can see that if $A_2 < 0$ the energy is unbounded from below; the trivial solution is then unstable since $\omega^2(k) < 0$ for sufficiently large $|k|$. This short-wave instability, however, is unphysical if the unstable wave length is shorter than the lattice spacing. To remove this possibility we can constrain the class of perturbations by requiring that $|k| \leq \pi$. This is equivalent to replacing (10) with $\omega^2(k) = 0$ outside the Brillouin zone (for $|k| \geq \pi$). The resulting dispersion relation with a short wave cut-off constitutes the main assumption of our quasicontinuum model; in Figure 2 it is compared with the dispersion relation for the discrete model.

The new model can be used to investigate stability of a homogeneous state. Following the same procedure as in the discrete case, we obtain

$$\begin{aligned} &E > 0 \\ &E + A_1\pi^2 + A_2\pi^4 > 0 \\ &4A_2E - A_1^2 > 0 \quad \text{if} \;\; 0 < -\frac{A_1}{2A_2} < \pi^2. \end{aligned} \tag{14}$$

The first condition indicating macroinstability coincides with $(8)_1$. The second condition is the analog of $(8)_2$ and the unstable mode is again $k = \pi$. The last inequality in (14) is

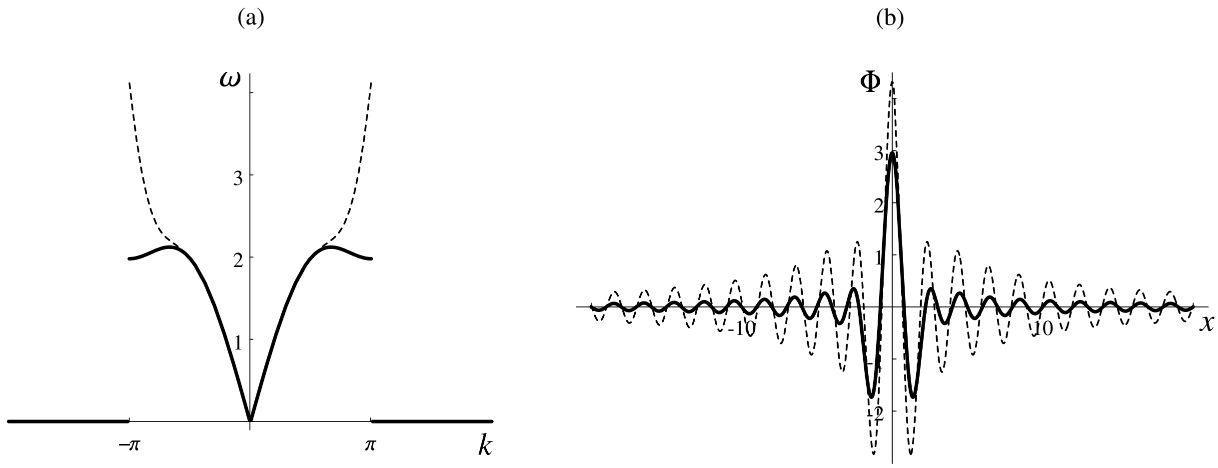

Figure 2: (a) The cut-off dispersion relations $\omega(k)$ for exact quasicontinuum model (solid curve) and the polynomial approximation (dashed). (b) The kernels $\Phi(x)$ of the corresponding nonlocal models in physical space. Parameters: $K_1 = 1$, $K_2 = 0.5$, $K_3 = -0.02$.

analogous to $(8)_3$, and its failure corresponds to incommensurate microinstability with

$$k = k^* = \sqrt{-\frac{A_1}{2A_2}}. \tag{15}$$

A stability diagram illustrating the above conditions is presented in Figure 3 where it is compared to the stability diagram for the discrete model. Observe that exactly as in the discrete case, at $K_1 > 0$ the incommensurate instability is located above point M, which coincides with the point M in the discrete case. Moreover, the stability boundaries in the quasicontinuum model and in the discrete case are expectedly tangent at the point M where the unstable mode has infinitely long wave ($k = 0$). Above the point M the model with a cut-off reproduces the main qualitative features of the discrete stability diagram while overestimating stability of the homogeneous configuration.

Between points M and $Q = (-4(\pi^2-3)/(13\pi^2-30), (\pi^2-6)/(3(13\pi^2-30))$ the instability mode is macroscopic in both models; note however that this interval is much shorter in the quasicontinuum model than its analog in the discrete case. Between points Q and $P = (-(120-80\pi^2+3\pi^4)/(10(30-26\pi^2+3\pi^4)), -(30\pi^2-\pi^4-90)/(45(30-26\pi^2+3\pi^4))$ the stability is lost via microscopic mode with $k = \pi$, and below P the instability mode is again incommensurate. This last transition from microscopic to incommensurate instability mode is not observed in the discrete case at $K_1 > 0$. Overall, the quasicontinuum approximation underestimates the stability of the trivial state in the region $\{K_1 > 0, K_2 > 0, K_3 < 0\}$. This is due to the larger contribution of the oscillation-producing K_3 terms in the polynomial model compared to the discrete case. At $K_1 < 0$

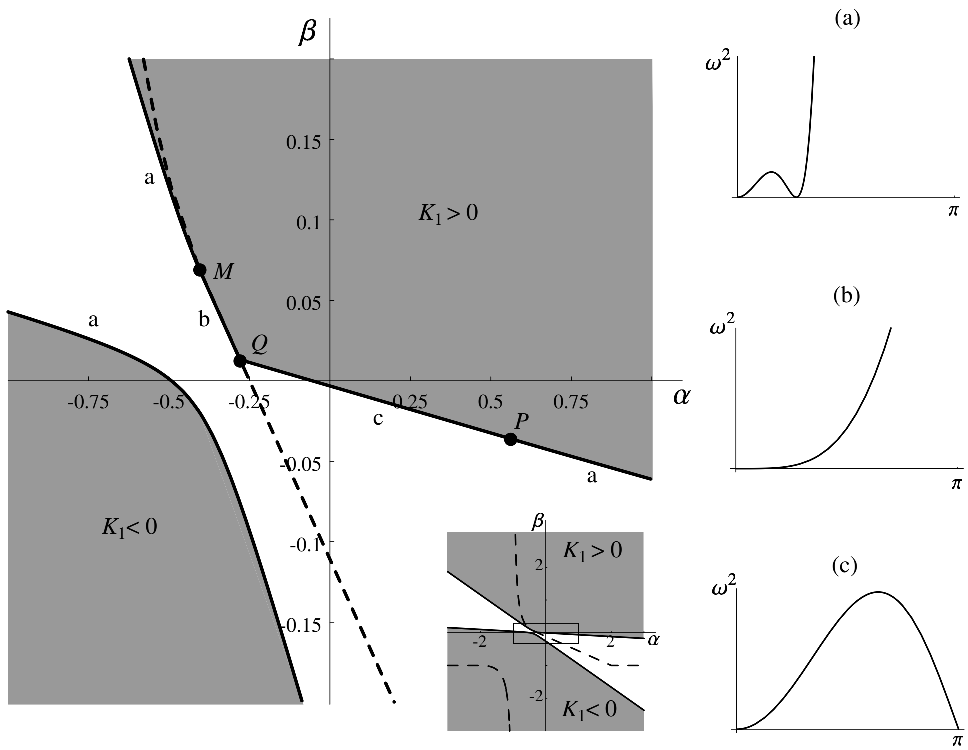

Figure 3: Stability diagram for the cut-off polynomial model and the corresponding graphs of $\omega^2(k)$: (a) incommensurate microinstability (above M, below P and entire boundary of $K_1 < 0$ stability region), $4A_2E - A_1^2 = 0$; (b) macroinstability (MQ), $E = 0$; (c) commensurate microinstability (QP), $E + A_1\pi^2 + A_2\pi^4 = 0$. Insert: stability diagram at a larger scale; the main picture is the magnification of the area inside the rectangle.

the approximation has the opposite effect: it overestimates the stability of the uniform state (compare with Figure 1). This is again caused by the very nature of the polynomial approximation: for positive K_2 and K_3 (which at $K_1 < 0$ result in negative α and β) the homogenizing quadratic and quartic terms easily dominate the unstable contribution due to K_1. Notice also that while in the discrete case the stability boundary contains a segment corresponding to microscopic instability which at point R becomes incommensurate, in the polynomial model the whole boundary corresponds to the incommensurate mode.

Despite its standard appearance, the proposed gradient approximation with a cut-off is essentially a model with long-range interactions and can be shown to be formally equivalent to a spatially nonlocal continuum model with the energy

$$W = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \Phi(x-\xi)u(x,t)u(\xi,t)dxd\xi \tag{16}$$

The kernel $\Phi(x)$ can be computed in the closed form as the inverse Fourier transform of the cut-off dispersion relation (10), yielding

$$\begin{aligned}\Phi(x) =& \frac{1}{\pi x^7}[2\pi x((E + 2A_1\pi^2 + 3A_2\pi^4)x^4 - 12(A_1 + 5A_2\pi^2)x^2 + 360A_2)\cos\pi x \\ &+ (\pi^2 x^6(E + A_1\pi^2 + A_2\pi^4) + 2(E + 6A_1\pi^2 + 15A_2\pi^4)x^4 \\ &+ 24(A_1 + 15A_2\pi^2)x^2 - 720A_2)\sin\pi x].\end{aligned} \tag{17}$$

Meanwhile, the exact nonlocal counterpart of the discrete model has the kernel (e.g. [16])

$$\Phi_D(x) = \frac{4\sin\pi x}{\pi x}\left(K_1\frac{x^2 - 1/2}{x^2 - 1} - \frac{2K_2}{x^2 - 4} + K_3\frac{x^2 - 9/2}{x^2 - 9}\right) \tag{18}$$

The two kernels are compared in Figure 2b. One can see that characteristically for the models with long-range interactions, the average amplitude decays in both cases as a power of distance.

4 Conclusions

It has been long recognized that the gradient approximations of the lattice models generate in the continuum limit either unbounded or nonpositive definite operators leading to ill-posed mathematical problems (e.g. [6, 23]). To overcome this difficulty we suggested to regularize such operators by restricting them to a finite sphere in the Fourier space. This restriction is somewhat similar to the finite element approximation in the sense that it smears out sufficiently small scales. The difference is that in our case the filtered scales are unphysical. As we showed, the resulting model effectively replaces at short waves partial differential equations by integral equations. The dual nature of such cut-off polynomial model may be used to design hybrid computational schemes filtering parasitic small-scale oscillations while taking full advantage of the availability of simple partial differential equations for slowly varying fields. As the first step in the direction of legitimization of such hybrid schemes we demonstrated in this paper that they generate an adequate description of the relevant bifurcations at least for the homogeneous configurations. An important open problem is the generalization of the present idea of a local-nonlocal description to the nonlinear case. Another challenging issue is dynamics where the wave numbers located outside the Brillouin zone are known to be important (e.g. [26]).

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