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Stability of crystalline solids—I: Continuum and atomic lattice considerations

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Abstract

Many crystalline materials exhibit solid-to-solid martensitic phase transformations in response to certain changes in temperature or applied load. These martensitic transformations result from a change in the *stability* of the material's crystal structure. It is, therefore, desirable to have a detailed understanding of the possible modes through which a crystal structure may become unstable. The current work establishes the connections between three crystalline stability criteria: phonon-stability, homogenized-continuum-stability, and the presently introduced Cauchy-Born-stability criterion. Stability with respect to phonon perturbations, which probe all bounded perturbations of a uniformly deformed specimen under "hard-device" loading (i.e., all around displacement type boundary conditions) is hereby called "constrained material stability". A more general "material stability" criterion, motivated by considering "soft" loading devices, is also introduced. This criterion considers, in addition to all bounded perturbations, all "quasi-uniform" perturbations (i.e., uniform deformations and internal atomic shifts) of a uniformly deformed specimen, and it is recommend as the relevant crystal stability criterion.

Keywords: Phase transformation; Vibrations; Finite strain; Stability and bifurcation; Asymptotic analysis

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1. Introduction

Many crystalline materials exhibit solid-to-solid phase transformations (PTs) in response to certain changes in temperature or applied load. These phase transformations can be categorized as "diffusional" or "diffusionless". In diffusional PTs the atoms in the crystal migrate over large distances with respect to the lattice spacing, eventually reaching a new equilibrium state (or phase). The diffusionless PTs, also called "martensitic" or "displacive" transformations, involve the coordinated motion of atoms in the crystal in response to an instability which results in the material's transformation from one lattice to another. In such a transformation neighboring atoms move small relative distances compared to the lattice spacing by a process that involves latent heat (a "first order" PT). Martensitic phase transformations (MTs) produce material property changes that have often been utilized for technological purposes. For example, the martensitic transformations in *steel* lead to its notable strength and hardness, *ferroelectric* materials exhibit MTs which result in the generation of a piezoelectric effect (Dove, 1993), *magnetostrictive* materials undergo MTs which lead to magnetic–mechanical coupling (Kittel, 1956), and MTs are responsible for the remarkable properties exhibited by *shape memory alloys* (Otsuka and Wayman, 1998).

For all of these materials a transformation occurs when the underlying crystal structure becomes unstable at a particular thermomechanical load, and the material reconfigures into a new stable structure. It is, therefore, desirable to have a detailed theory of the possible modes by which a crystal structure may become unstable. This should ultimately lead to a better understanding of why certain crystalline solids exhibit MTs. This knowledge can subsequently be applied toward the development of improved modeling techniques for existing materials and could potentially help guide the search for undiscovered materials that possess unique properties.

The solid-state physics community has partially addressed this need by considering a crystal as an assembly of independent atoms and calculating the phonon-spectra (normal mode frequencies) corresponding to bounded perturbations.[2] Thus, an infinite crystal is considered stable if all its phonon frequencies are real. First developed by Born and Huang (1962), some examples of use of the "phonon-stability" criterion include the quantum mechanics calculations of Huang et al. (2002) and the molecular dynamics calculations of Yu and Clapp (1989) and Kastner (2003). In contrast to these explicit atomic calculations, many investigators have adopted a continuum level energy minimization definition of stability. In this case, a homogenized material energy density is employed and energy minimizing equilibrium configurations are considered to be stable. This "homogenized-continuum" (HC-stability) criterion ensures stability of the crystal with respect to both rank-one and non-rank-one uniform deformation perturbations. The HC-stability convention is prevalent in the continuum physics and mechanics communities and is embodied in the classic Landau theory of phase transition (see, for example, Dove, 1993) as well as more recent investigations by James (1987), Pitteri and Zanzotto (2002), and Bhattacharya (2003).

The relationships between these two differing views of crystalline stability have received little attention in the literature. The notable exception is Born's *method of long waves* (Born

[2]In the mechanics community a similar method, known as the "Bloch-wave method", has been developed for continuous periodic solids of infinite extent (see Geymonat et al., 1993).

and Huang, 1962), which investigates the behavior of long wavelength "acoustic phonon" modes and proves that these modes are equivalent to the rank-one "uniform" perturbations considered by the HC-stability criterion.

To provide a better understanding of crystalline stability and the different criteria used to measure it, the current work introduces a new, generalized, continuum stability criterion, named the "Cauchy–Born (CB) stability" criterion. CB-stability is based on CB kinematics for multilattices and includes uniform deformation and "internal atomic shift" perturbations of the crystal structure. The three stability criteria (phonon-, CB-, and HC-) are, for the first time, concurrently presented in a *unified context* to illuminate the relationships between them. Stability with respect to phonon perturbations, which probe all bounded perturbations of a uniformly deformed specimen under "hard-device" loading (i.e., all around displacement type boundary conditions) is, hereby, called "constrained material stability". A more general "material stability" criterion, motivated by considering "soft" loading devices, is also introduced. This criterion considers, in addition to all bounded perturbations, all "quasi-uniform" perturbations (i.e., uniform deformations and internal atomic shifts) of a uniformly deformed specimen and it is recommend as the relevant crystal stability criterion.

Although several parts of the lattice-based atomistic stability calculations exist in one form or another in the physics literature, the current general, full Lagrangian description as well as the connections made between the existing (phonon- and HC-) stability criteria and the presently introduced (CB) stability criterion are novel and merit, in the authors' opinion, an independent presentation.

The stability of a three-dimensional (3-D) multi-atomic perfect crystalline solid is investigated in Section 2. First, the crystal's atomic equilibrium equations and stability conditions for bounded perturbations of all wavelengths (phonon-stability) are presented. The crystal's atomic energy density is then homogenized, resulting in the CB model that allows uniform deformations of the crystal as well as "internal shifts", collectively called "quasi-uniform" deformations. Additionally, it is shown that CB equilibrium ensures that every atom in the crystal is in force equilibrium. Finally, these internal shifts are condensed out of the energy density to obtain the HC model. Section 3 compares the three stability criteria—phonon-, CB-, and HC-stability—with respect to the types of perturbations that they consider, and the "material stability" criterion is recommended. In the companion paper (Part II of this work, Elliott et al., 2004) the recommended stability criteria are applied to a specific atomic-scale model to study the temperature-dependent stability of bi-atomic shape memory alloys.

2. Three-dimensional crystals

In this section an arbitrary finite crystal with long range (non-local) interactions between its atoms is considered. At the expense of some rather heavy notation, the equations are kept as explicit as possible.[3]

[3]In Elliott (2004) attention is also given to the analogous 1-D formulation in an effort to more clearly display the general mathematical structure.

2.1. Problem description

The potential energy of the crystal is derived as a function of the atomic displacements from the reference configuration. A two-dimensional 4-lattice finite crystal of atoms, Ω, consisting of the four sub-lattices $\mathscr{L}_0, \mathscr{L}_1, \mathscr{L}_2, \mathscr{L}_3$ is shown in Fig. 1. The atoms interact through a set of long range multi-atomic potentials (not illustrated) and are subjected to a set of externally applied forces $\mathbf{f}\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]$ (also not illustrated) on the "boundary atoms" (defined below). The "extra" atoms comprising the positive faces of the crystal $\mathscr{F}^+$ are included to facilitate the subsequent application of periodic boundary conditions.

2.1.1. Multilattices

A three-dimensional simple lattice (1-lattice) is an infinite collection of discrete "lattice points" in $\mathbb{R}^3$ generated by three non-coplanar lattice basis vectors $\mathbf{G}_i$, such that each

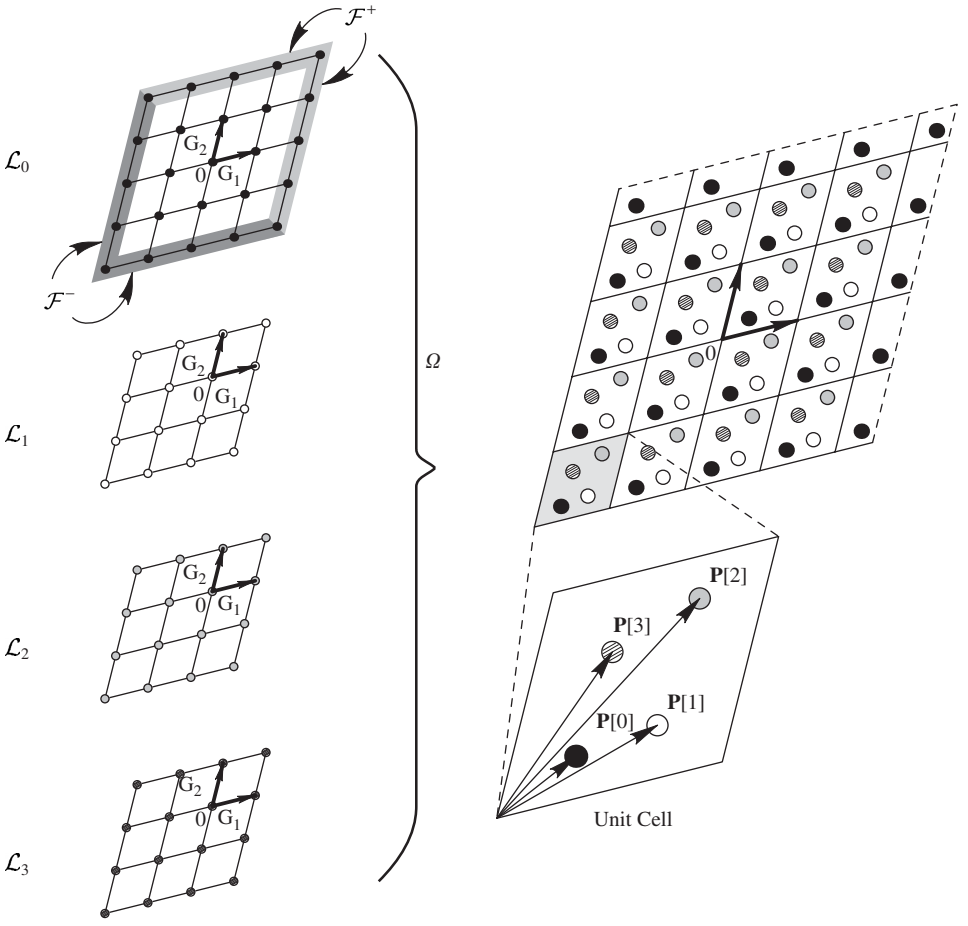

Fig. 1. Multilattice ($M = 4$) description of a 2-D finite crystal showing the individual sub-lattices $\mathscr{L}_\alpha$, the composite crystal Ω, and the fractional position vectors $\mathbf{P}[\alpha]$ within a unit cell.

lattice point has the position vector[4]

$$\mathbf{X}[\ell] = \ell^i \mathbf{G}_i, \quad \ell^i \in \mathbb{Z}, \tag{2.1}$$

where the label ℓ for the lattice point is a short-hand notation referring to the triplet of integers ℓ^i. The $\mathbf{G}_i$'s for a lattice are not unique, but belong to a class of linearly independent lattice vectors that generate all lattice points when the linear combinations (Eq. (2.1)) are formed (see Miller, 1972). Associated with each set of lattice basis vectors $\mathbf{G}_i$ is a set of "reciprocal lattice basis vectors" $\mathbf{G}^i$ defined by

$$\mathbf{G}^i \bullet \mathbf{G}_j = \delta^i_j \quad \text{(Kroneker delta)}, \tag{2.2}$$

while the metric tensor components in these bases are $G_{ij} \equiv \mathbf{G}_i \bullet \mathbf{G}_j$, and $G^{ij} \equiv \mathbf{G}^i \bullet \mathbf{G}^j$. The lattice vectors transform to and from the reciprocal lattice vectors through the above defined metric tensor components

$$\mathbf{G}^i = G^{ij}\mathbf{G}_j, \quad \mathbf{G}_i = G_{ij}\mathbf{G}^j, \tag{2.3}$$

as do the components of any tensor.

An M-lattice is a collection of M distinct mono-atomic sub-lattices, with the same set of basis lattice vectors $\mathbf{G}_i$, which are located relative to each other by simple translations. The M-lattice is described by giving the M relative position vectors $\mathbf{P}[\alpha]$ of each sub-lattice with respect to some imaginary "skeletal lattice". These relative position vectors are called the "fractional position vectors" (Sands, 1993), since their components range between zero and one

$$\mathbf{P}[\alpha] = P^i[\alpha]\mathbf{G}_i, \quad 0 \leqslant P^i[\alpha] < 1, \quad \alpha = 0, 1, \ldots, M-1. \tag{2.4}$$

This description allows the unique identification of each atom in the crystal with a single sub-lattice index α and a single skeletal lattice point ℓ. The reference position vector of atom $\begin{bmatrix} \ell \\ \alpha \end{bmatrix}$ is

$$\mathbf{X}\begin{bmatrix} \ell \\ \alpha \end{bmatrix} = \mathbf{X}[\ell] + \mathbf{P}[\alpha], \quad \mathbf{X}[\ell] \equiv \ell^i \mathbf{G}_i, \quad \mathbf{P}[\alpha] \equiv P^i[\alpha]\mathbf{G}_i, \tag{2.5}$$

where $\mathbf{X}[\ell]$ is the position vector of the skeletal lattice point ℓ. Eq. (2.5) (arbitrarily) chooses the skeletal lattice to be centered on the coordinate origin, implying $\mathbf{X}[0] = \mathbf{0}$. Each periodic volume containing M atoms associated with the lattice point ℓ is called a "unit cell" (see the shaded region in Fig. 1).

The 3-D finite crystal Ω, consisting of $n \equiv (2N)^3$ unit cells, is chosen so that the domain Ω is much larger than the effective range of atomic interactions (large N). This guarantees that boundary effects on the crystal's bulk properties can be neglected. Atoms belonging to the zeroth sub-lattice are placed on the set of lattice points (which includes the positive faces of the crystal $\mathscr{F}^+$, see Fig. 1)

$$\mathscr{N}^+ \equiv \{\ell \in \mathbb{Z}^3 | -N \leqslant \ell^i \leqslant N\}, \tag{2.6}$$

[4]Unless otherwise specified, Latin indices represent spatial tensor components and Einstein's summation convention is employed for repeated indices (over $i = 1, 2, 3$).

resulting in the finite sub-lattice of atoms

$$\mathscr{L}_0 \equiv \left\{ \begin{bmatrix} \ell \\ 0 \end{bmatrix} | \ell \in \mathscr{N}^+ \right\}. \tag{2.7}$$

The remaining sub-lattices have atoms on the set of lattice points (without the positive faces)

$$\mathscr{N} \equiv \{\ell \in \mathbb{Z}^3 | -N \leqslant \ell^i < N\}, \tag{2.8}$$

resulting in the $M-1$ finite sub-lattices of atoms

$$\mathscr{L}_\alpha \equiv \left\{ \begin{bmatrix} \ell \\ \alpha \end{bmatrix} | \ell \in \mathscr{N} \right\}, \quad \alpha = 1, 2, \ldots, M-1. \tag{2.9}$$

The sub-lattices $\mathscr{L}_\alpha$ for $\alpha = 0, 1, \ldots, M-1$ are shown in Fig. 1 (for the case $M = 4$). The finite crystal Ω is the union of these sub-lattices

$$\Omega \equiv \bigcup_{\alpha=0}^{M-1} (\mathscr{L}_\alpha). \tag{2.10}$$

Additionally, the subsets of lattice points $\mathscr{F}^+, \mathscr{F}^- \subset \mathscr{L}_0$ are defined as the three positive faces, $\ell^1 = N, \ell^2 = N, \ell^3 = N$, and three negative faces, $\ell^1 = -N, \ell^2 = -N, \ell^3 = -N$, of the crystal, respectively. These sets are illustrated for the 2-D crystal in Fig. 1 where it can be seen that these faces only contain atoms from the finite sub-lattice $\mathscr{L}_0$.

If one imagines Ω as a subset of an infinite M-lattice the total force $\mathbf{f}_\mathrm{T}$ on atom $\begin{bmatrix} \ell \\ \alpha \end{bmatrix}$ will have a component $\mathbf{f}_\Omega$ due to atoms in Ω and a component $\mathbf{f}_{\Omega^\mathrm{c}}$ due to atoms outside Ω, i.e.,

$$\mathbf{f}_\mathrm{T} \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = \mathbf{f}_\Omega \begin{bmatrix} \ell \\ \alpha \end{bmatrix} + \mathbf{f}_{\Omega^\mathrm{c}} \begin{bmatrix} \ell \\ \alpha \end{bmatrix}, \tag{2.11}$$

where Ω^c is the (complementary) set of atoms in the infinite crystal outside the parallelepiped Ω. Assuming the finite crystal Ω is large, only those atoms near the boundary of Ω will have non-zero forces $\mathbf{f}_{\Omega^\mathrm{c}} \begin{bmatrix} \ell \\ \alpha \end{bmatrix}$, due to the rapid decay of atomic forces with distance. This set of atoms (which have "broken bonds" in the finite crystal) is called the *boundary* of Ω,

$$\partial\Omega \equiv \left\{ \begin{bmatrix} \ell \\ \alpha \end{bmatrix} \in \Omega \,\middle|\, \left\| \mathbf{f}_{\Omega^\mathrm{c}} \begin{bmatrix} \ell \\ \alpha \end{bmatrix} \right\| > \varepsilon \right\}, \tag{2.12}$$

where ε is a small number, and the remaining atoms in Ω, called *interior* atoms, are

$$\Omega_0 \equiv \Omega \backslash \partial\Omega = \left\{ \begin{bmatrix} \ell \\ \alpha \end{bmatrix} \in \Omega \,\middle|\, \left\| \mathbf{f}_{\Omega^\mathrm{c}} \begin{bmatrix} \ell \\ \alpha \end{bmatrix} \right\| < \varepsilon \right\}. \tag{2.13}$$

Note that $\mathscr{F}^+ \cup \mathscr{F}^- \subset \partial\Omega$ (strictly), since in general the crystal faces $\mathscr{F}^+$ and $\mathscr{F}^-$ do not constitute the entire set of boundary atoms $\partial\Omega$.

2.1.2. Energy density and atomic potentials

Atoms in the crystal are assumed to interact through a set of atomic potentials given by the individual atomic energy contributions $\psi \begin{bmatrix} \ell \\ \alpha \end{bmatrix}$ resulting in the internal potential energy density W of the finite crystal as a function of the atomic displacements $\mathbf{u} \begin{bmatrix} \ell \\ \alpha \end{bmatrix}$ from their

reference positions $\mathbf{X}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}$

$$W(\mathbf{u}) = \frac{1}{nV}\sum_{\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\in\Omega} \psi\begin{bmatrix}\ell\\ \alpha\end{bmatrix}, \quad \mathbf{u} \equiv \left\{\mathbf{u}\begin{bmatrix}\ell\\ \alpha\end{bmatrix} \,\middle|\, \begin{bmatrix}\ell\\ \alpha\end{bmatrix} \in \Omega\right\}, \tag{2.14}$$

where $\mathbf{u}$ is the vector of all atomic displacements and $V = \mathbf{G}_1 \bullet (\mathbf{G}_2 \times \mathbf{G}_3)$ is the reference unit cell volume.

Applied external forces on the boundary atoms are denoted by

$$\mathbf{f}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}, \quad \begin{bmatrix}\ell\\ \alpha\end{bmatrix} \in \partial\Omega. \tag{2.15}$$

The total potential energy density for the finite crystal is

$$\mathscr{E}(\mathbf{u}) = W(\mathbf{u}) - \frac{1}{nV}\sum_{\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\in\partial\Omega} \mathbf{f}\begin{bmatrix}\ell\\ \alpha\end{bmatrix} \bullet \mathbf{u}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}. \tag{2.16}$$

The derivatives of the internal potential energy (nVW), Eq. (2.14), are often evaluated at some specified configuration of the crystal (usually an equilibrium configuration), denoted by $\overset{o}{\mathbf{u}}$, so it is convenient to introduce the following notation[5]:

$$\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix} \equiv \left.\frac{\partial(nVW)}{\partial u^i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}}\right|_{\overset{o}{\mathbf{u}}}, \quad \overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix} \equiv \left.\frac{\partial^2(nVW)}{\partial u^i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\partial u^j\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}}\right|_{\overset{o}{\mathbf{u}}}. \tag{2.17}$$

Eq. $(2.17)_1$ represents the total sum of forces in the $\mathbf{G}_i$ direction on each atom due to all other atoms in Ω, and Eq. $(2.17)_2$ represents the stiffness between the atoms $\begin{bmatrix}\ell\\ \alpha\end{bmatrix}$ and $\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}$ in Ω. With these definitions the energy density is expanded in powers of the perturbation $\delta\mathbf{u}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}$ about the configuration $\overset{o}{\mathbf{u}}$ as

$$\begin{aligned}\mathscr{E}(\overset{o}{\mathbf{u}}+\delta\mathbf{u}) = \mathscr{E}(\overset{o}{\mathbf{u}}) + \frac{1}{nV}\Bigg[&\sum_{\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\in\Omega} \overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\delta u^i\begin{bmatrix}\ell\\ \alpha\end{bmatrix} + \frac{1}{2}\sum_{\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\in\Omega}\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}\delta u^i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\delta u^j\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} + \cdots\Bigg]\\ &- \frac{1}{nV}\sum_{\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\in\partial\Omega} f_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\delta u^i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}.\end{aligned} \tag{2.18}$$

For an arbitrary non-linear crystal the equilibrium and stability conditions must be calculated numerically, in general, but can be analyzed by considering perturbed equations in terms of the quantities $\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}$ and $\overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}$.

2.1.3. Translational invariance and periodicity relations

The internal energy density must be invariant with respect to arbitrary rigid-body translations ($\mathbf{u}\begin{bmatrix}\ell\\ \alpha\end{bmatrix} - \overset{o}{\mathbf{u}}\begin{bmatrix}\ell\\ \alpha\end{bmatrix} = \mathbf{d}$ constant vector) of the configuration $\overset{o}{\mathbf{u}}$. Consequently, its

[5]In the remainder of this work, an 'o' above a variable indicates evaluation of the quantity at configuration $\overset{o}{\mathbf{u}}$.

derivative with respect to $\mathbf{d}$ must be zero[6]

$$\left.\frac{\partial W}{\partial d^i}\right|_{\mathbf{d}=\mathbf{0}} = \sum_{\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\in\Omega} \left.\frac{\partial W}{\partial u^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}}\right|_{\overset{o}{\mathbf{u}}} \frac{\partial u^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}}{\partial d^i} = \frac{1}{nV}\sum_{\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\in\Omega} \overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix} = 0. \tag{2.19}$$

Furthermore, higher-order derivatives of W must also be invariant with respect to rigid-body translations, giving

$$\left.\frac{\partial^2 W}{\partial u^i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\partial d^j}\right|_{\mathbf{d}=\mathbf{0}} = \frac{1}{nV}\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix} = \frac{1}{nV}\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \overset{o}{\Phi}_{ji}\begin{bmatrix}\ell' & \ell\\ \alpha' & \alpha\end{bmatrix} = 0, \tag{2.20}$$

where the symmetry of the coefficients $\overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}$ with respect to its indices (due to their definition as a second-order derivative of the internal energy),

$$\overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix} = \overset{o}{\Phi}_{ji}\begin{bmatrix}\ell' & \ell\\ \alpha' & \alpha\end{bmatrix}, \tag{2.21}$$

has been used in the second equality of Eq. (2.20). There is also a similar set of conditions expressing the invariance of the energy density with respect to rigid-body rotations, although these do not contribute anything in the present theory since rigid-body rotation modes are eliminated by the application of periodic boundary conditions. The reader is referred to Wallace (1998) for invariance relations corresponding to infinitesimal rigid-body rotations.

The stability criteria discussed later pertain to 3-D crystals with deformed equilibrium configurations that retain the periodicity of the referential 3-D multilattice. Consequently, the deformed configuration of the M-lattice remains describable as an M-lattice, and thus, the current atomic positions can be written in the form

$$\overset{o}{\mathbf{x}}\begin{bmatrix}\ell\\ \alpha\end{bmatrix} = \overset{o}{\mathbf{F}}\bullet\mathbf{X}\begin{bmatrix}\ell\\ \alpha\end{bmatrix} + \overset{o}{\mathbf{s}}[\alpha] = \overset{o}{\mathbf{u}}\begin{bmatrix}\ell\\ \alpha\end{bmatrix} + \mathbf{X}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}, \tag{2.22}$$

where $\overset{o}{\mathbf{F}}$ is the uniform deformation gradient tensor and $\overset{o}{\mathbf{s}}[\alpha]$ are the sub-lattice's current "internal shift" vectors away from their affine locations. The space of equilibrium configurations that are consistent with a user selected M-lattice description will be called "CB" configurations. For these equilibrium configurations the force and stiffness coefficients, Eq. (2.17), must satisfy periodicity conditions for atoms in the interior of the crystal

$$\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix} = \overset{o}{\Phi}_i\begin{bmatrix}\ell+l\\ \alpha\end{bmatrix}, \quad \begin{bmatrix}\ell\\ \alpha\end{bmatrix}, \begin{bmatrix}\ell+l\\ \alpha\end{bmatrix} \in \Omega_0, \tag{2.23}$$

and

$$\overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix} = \overset{o}{\Phi}_{ij}\begin{bmatrix}\ell+l & \ell'+l\\ \alpha & \alpha'\end{bmatrix}, \quad \begin{bmatrix}\ell\\ \alpha\end{bmatrix}, \begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}, \begin{bmatrix}\ell+l\\ \alpha\end{bmatrix}, \begin{bmatrix}\ell'+l\\ \alpha'\end{bmatrix} \in \Omega_0. \tag{2.24}$$

[6]In fact, Eq. (2.19) is valid for any $\mathbf{d}$. Evaluation at $\mathbf{d}=\mathbf{0}$ results in the corresponding expression in terms of the coefficients $\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}$.

2.2. Phonon model

All three of the stability criteria (phonon, CB, and HC) will address stability of a CB configuration of the form of Eq. (2.22), but the perturbations that they allow are different. In this section, the phonon-stability criterion for 3-D crystals is derived using the atomistic model presented in the previous section. This criterion addresses stability of CB equilibrium configurations of the form of Eq. (2.22).

2.2.1. Microscopic equilibrium and phonon-stability conditions

The conditions for $\overset{o}{\mathbf{u}}$ to be an equilibrium configuration are given by

$$\left.\frac{\partial \mathscr{E}}{\partial u^i \begin{bmatrix} \ell \\ \alpha \end{bmatrix}}\right|_{\overset{o}{\mathbf{u}}} = 0 \begin{cases} \ell \in \Omega, \\ \alpha = 0, 1, \ldots, M-1, \\ i = 1, 2, 3. \end{cases} \tag{2.25}$$

Using Eq. (2.18) equilibrium becomes

$$\overset{o}{\Phi}_i \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = \begin{cases} 0, & \begin{bmatrix} \ell \\ \alpha \end{bmatrix} \in \Omega_0, \\ f_i \begin{bmatrix} \ell \\ \alpha \end{bmatrix}, & \begin{bmatrix} \ell \\ \alpha \end{bmatrix} \in \partial\Omega. \end{cases} \tag{2.26}$$

This expresses "microscopic" equilibrium of each atom. Application of the translational invariance relation, Eq. (2.19), results in an expression of global force equilibrium, or

$$\sum_{\begin{bmatrix} \ell \\ \alpha \end{bmatrix} \in \partial\Omega} f_i \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = 0. \tag{2.27}$$

Stability of the crystal is evaluated by considering the linearized dynamical behavior of the system about its equilibrium configuration with periodic boundary conditions, given by

$$m_\alpha \delta\ddot{u}^j \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = -\sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} G^{jk} \overset{o}{\Phi}_{kp} \begin{bmatrix} \ell & \ell' \\ \alpha & \alpha' \end{bmatrix} \delta u^p \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix},$$

$$\delta\mathbf{u} \begin{bmatrix} \ell \\ 0 \end{bmatrix} = \delta\mathbf{u} \begin{bmatrix} \ell' \\ 0 \end{bmatrix}, \quad \begin{bmatrix} \ell \\ 0 \end{bmatrix} \in \mathscr{F}^+, \quad \begin{bmatrix} \ell' \\ 0 \end{bmatrix} \in \mathscr{F}^-, \tag{2.28}$$

where $\ddot{(\,)} \equiv \partial^2(\,)/\partial t^2$ (t is time), m_α is the mass of atom α, and $\begin{bmatrix} \ell \\ 0 \end{bmatrix}$ and $\begin{bmatrix} \ell' \\ 0 \end{bmatrix}$ in Eq. $(2.28)_2$ are located directly across from each other on their respective positive and negative crystal faces. Of interest are the normal modes of vibration (eigenmodes of Eq. (2.28)) that are characteristic of the bulk material, which means that the effect of free surfaces on the stability of the crystal will be neglected. The application of periodic boundary conditions, Eq. $(2.28)_2$, first introduced by Born, is now customary and can be interpreted as providing a procedure for obtaining a "representative sampling" of vibration modes in an infinitely extended crystal (see further, Born and Huang, 1962, p. 45). Additionally, the use of periodic boundary conditions restricts the stability criterion to consider perturbations with zero average deformation of the specimen and thus eliminates "structural" type modes

(such as bending) and uniform deformations (such as multi-axial strain and uniform dilatation) from consideration. Note that periodic boundary conditions are not applied to the entirety of $\partial\Omega$ but only to the faces $\mathscr{F}^+$ and $\mathscr{F}^- \subset \mathscr{L}_0$. Furthermore, the rigid-body translation modes are still included in Eq. (2.28) and must be eliminated before any stability criterion is applied.

The 3-D crystal is considered stable if there exists a $\delta(\varepsilon)>0$ such that for any given $\varepsilon>0$ solutions to the linearized equations of motion Eq. (2.28) satisfy

$$\left\|\delta\mathbf{u}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}(0)\right\|, \left\|\delta\dot{\mathbf{u}}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}(0)\right\| < \varepsilon, \quad \Rightarrow \left\|\delta\mathbf{u}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}(t)\right\|, \left\|\delta\dot{\mathbf{u}}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}(t)\right\| < \delta(\varepsilon) \quad \forall t>0, \tag{2.29}$$

where $\delta\mathbf{u}\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right](0)$ and $\delta\dot{\mathbf{u}}\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right](0)$ are the respective vector of initial displacements and velocities at time $t=0$, and $\|\cdot\|$ is any norm for $\mathbf{u}\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]$. Equivalently, for the conservative system under consideration, all solutions remain bounded if and only if all eigenvalues of the global stiffness matrix $\overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}$ (with the rigid-body modes eliminated) are positive.

2.2.2. *Block-diagonalization of the system's global stiffness matrix*

The equations of motion, Eq. (2.28), are a set of $3Mn \times 3Mn$ linear, constant coefficient, ordinary differential equations with exponential solutions of the form

$$\delta u^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}^{(r)} = \Delta u^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}^{(r)} \exp\{i\omega^{(r)}t\}, \quad r=1,2,\ldots,3Mn; \tag{2.30}$$

where $i \equiv \sqrt{-1}$, $\Delta u^j\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]^{(r)}$ are the eigenvectors, and $\omega^{(r)}$ are the corresponding cyclic frequencies. The normal modes of vibration in the crystal are referred to as *phonons*, taken from the condensed matter physics literature, and every solution of Eq. (2.28) can be expressed as a linear combination of the $3Mn$ independent phonon modes (Eq. (2.30)). Stability requires that the phonon frequencies be real quantities, or equivalently all

$$(\omega^{(r)})^2>0, \quad r=4,5,\ldots,3Mn; \tag{2.31}$$

where the rigid-body translation modes are associated with $r=1,2,3$ (rigid-body rotations having been eliminated by the periodic boundary conditions).

Generally, diagonalization of the stiffness matrix $G^{jk}\overset{o}{\Phi}_{kp}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}$ of Eq. (2.28) to determine the phonon frequencies is a time consuming computation due to the large dimension of the system ($3Mn$, with the number of cells $n \equiv (2N)^3$ large). Fortunately, the periodicity conditions, Eq. (2.24), ensure that a block-diagonalization (to $3M \times 3M$ blocks) exists, which greatly reduces the computational effort required to obtain the phonon frequencies.[7]

The *block-Fourier transform* for the 3-D crystal's displacement perturbation is

$$\delta\widehat{u}^j\begin{bmatrix}h\\ \alpha\end{bmatrix} = \frac{1}{\sqrt{n}}\sum_{\ell\in\mathscr{N}} \delta u^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix} \exp\{i(\mathbf{k}[h]\bullet\mathbf{X}[\ell])\}, \tag{2.32}$$

[7]The periodicity conditions (Eq. (2.24)) imply that the stiffness matrix is "block-circulant" (Davis, 1994), and therefore, the existence of the block-diagonalization is ensured.

and the *inverse block-Fourier transform* is

$$\delta u^j \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = \frac{1}{\sqrt{n}} \sum_{h \in \mathscr{N}} \delta \hat{u}^j \begin{bmatrix} h \\ \alpha \end{bmatrix} \exp\{-i(\mathbf{k}[h] \bullet \mathbf{X}[\ell])\}. \tag{2.33}$$

Here, $\mathbf{k}[h]$ is the wave vector with respect to the reference configuration

$$\mathbf{k}[h] = 2\pi \frac{h_j}{2N} \mathbf{G}^j, \quad h_j \in \mathscr{N}, \tag{2.34}$$

and $h \equiv h_j$ (a triplet of indices) are the coordinates of reciprocal lattice points. Defining the Fourier transform on the set of lattice points $\mathscr{N}$ results in the automatic application of periodic boundary conditions when the crystal's equations of motion, Eq. (2.28), are transformed to Fourier-space (see Elliott, 2004 for further details).

Taking the block-Fourier transform of Eq. (2.28), substituting the inverse transform, Eq. (2.33), for $\delta \mathbf{u} \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix}$ on the right hand side, and combining the exponential terms results in the Fourier-space equations of motion[8]

$$\begin{aligned} m_\alpha \delta \ddot{\hat{u}}^j \begin{bmatrix} h \\ \alpha \end{bmatrix} = & -\frac{1}{n} \sum_{\alpha'=0}^{M-1} \sum_{h' \in \mathscr{N}} \left[\sum_{\ell \in \mathscr{N}} \sum_{\ell' \in \mathscr{N}^+} G^{jk} \overset{o}{\Phi}_{kp} \begin{bmatrix} \ell & \ell' \\ \alpha & \alpha' \end{bmatrix} \right. \\ & \left. \times \exp\{i(\mathbf{k}[h] \bullet \mathbf{X}[\ell] - \mathbf{k}[h'] \bullet \mathbf{X}[\ell'])\} \right] \delta \hat{u}^p \begin{bmatrix} h' \\ \alpha' \end{bmatrix}, \end{aligned} \tag{2.35}$$

which after multiplying and dividing by $\exp\{i\mathbf{k}[h'] \bullet \mathbf{X}[\ell]\}$ can be written as

$$\begin{aligned} m_\alpha \delta \ddot{\hat{u}}^j \begin{bmatrix} h \\ \alpha \end{bmatrix} = & -\frac{1}{n} \sum_{\alpha'=0}^{M-1} \sum_{h' \in \mathscr{N}} \left[\sum_{\ell \in \mathscr{N}} \exp\{i(\{\mathbf{k}[h] - \mathbf{k}[h']\}) \bullet \mathbf{X}[\ell]\} \right. \\ & \left. \times \sum_{\ell' \in \mathscr{N}^+} G^{jk} \overset{o}{\Phi}_{kp} \begin{bmatrix} \ell & \ell' \\ \alpha & \alpha' \end{bmatrix} \exp\{-i(\mathbf{k}[h'] \bullet \{\mathbf{X}[\ell'] - \mathbf{X}[\ell]\})\} \right] \delta \hat{u}^p \begin{bmatrix} h' \\ \alpha' \end{bmatrix}. \end{aligned} \tag{2.36}$$

In the interior of the crystal the stiffness coefficients $\overset{o}{\Phi}_{ij} \begin{bmatrix} \ell & \ell' \\ \alpha & \alpha' \end{bmatrix}$ correspond to a block-circulant matrix. The non-circulant boundary effects are small since a "large" crystal, where the number of boundary atoms is much smaller than the number of interior atoms, is considered. Neglecting these boundary effects and replacing $\overset{o}{\Phi}$ with a purely block-circulant approximation is called "evaluating in the interior" and involves replacing $\overset{o}{\Phi}_{ij} \begin{bmatrix} \ell & \ell' \\ \alpha & \alpha' \end{bmatrix}$ by $\overset{o}{\Phi}_{ij} \begin{bmatrix} 0 & \ell' - \ell \\ \alpha & \alpha' \end{bmatrix}$, relabeling $(\ell' - \ell)$ as ℓ'' and noting that $\mathbf{X}[\ell''] = \mathbf{X}[\ell'] - \mathbf{X}[\ell]$, which results in

$$\begin{aligned} m_\alpha \delta \ddot{\hat{u}}^j \begin{bmatrix} h \\ \alpha \end{bmatrix} = & -\frac{1}{n} \sum_{\alpha'=0}^{M-1} \sum_{h' \in \mathscr{N}} \left[\sum_{\ell \in \mathscr{N}} \exp\{i(\{\mathbf{k}[h] - \mathbf{k}[h']\}) \bullet \mathbf{X}[\ell]\} \right. \\ & \left. \times \sum_{\ell'' \in \mathscr{N}^+} G^{jk} \overset{o}{\Phi}_{kp} \begin{bmatrix} 0 & \ell'' \\ \alpha & \alpha' \end{bmatrix} \exp\{-i(\mathbf{k}[h'] \bullet \mathbf{X}[\ell''])\} \right] \delta \hat{u}^p \begin{bmatrix} h' \\ \alpha' \end{bmatrix}, \end{aligned} \tag{2.37}$$

[8]For the summations $\sum_{\alpha'=0}^{M-1} \sum_{\ell' \in \mathscr{N}^+} (\cdot)$, the terms $\sum_{\alpha'=1}^{M-1} \sum_{\ell' \in \mathscr{F}^+ \cup \mathscr{F}^-} (\cdot)$ are implicitly set to zero since they involve atoms $\begin{bmatrix} \ell \\ \alpha \end{bmatrix} \notin \Omega$ which are outside the finite crystal.

where the sum on ℓ is now independent of the sum on ℓ''. It is well known that the finite Fourier modes (or basis vectors) are orthonormal giving, in particular,

$$\sum_{\ell\in\mathcal{N}} \exp\{i(\{\mathbf{k}[h]-\mathbf{k}[h']\})\bullet\mathbf{X}[\ell]\} = n\delta_{h'h}, \quad \delta_{h'h} = \begin{cases} 1, & h'_j = h_j,\ j=1,2,3, \\ 0, & \text{otherwise.} \end{cases} \tag{2.38}$$

Thus, the equations of motion are expressed more simply as (with the sum on ℓ canceling the factor of n)

$$m_\alpha \delta\ddot{\hat{u}}^j \begin{bmatrix} h \\ \alpha \end{bmatrix} = -\sum_{\alpha'=0}^{M-1} \left[\sum_{\ell'\in\mathcal{N}^+} G^{jk} \overset{o}{\Phi}_{kp} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \exp\{-i(\mathbf{k}[h]\bullet\mathbf{X}[\ell'])\} \right] \delta\hat{u}^p \begin{bmatrix} h \\ \alpha' \end{bmatrix} \tag{2.39}$$

for all $h\in\mathcal{N}$.

2.2.3. *Phonon frequencies—normal modes of vibration*

It is convenient to change variables, normalize by the atomic masses, and introduce a complex phase factor to recast the eigenvalue problem in terms of perturbations $\delta\hat{\mathbf{v}}\begin{bmatrix} h \\ \alpha \end{bmatrix}$ defined by

$$\delta\hat{u}^j \begin{bmatrix} h \\ \alpha \end{bmatrix} = \frac{\exp\{-i(\mathbf{k}[h]\bullet\mathbf{P}[\alpha])\}}{\sqrt{m_\alpha}} \delta\hat{v}^j \begin{bmatrix} h \\ \alpha \end{bmatrix}. \tag{2.40}$$

Substituting into the transformed equations of motion (Eq. (2.39)) gives

$$\delta\ddot{\hat{v}}^j \begin{bmatrix} h \\ \alpha \end{bmatrix} = -\sum_{\alpha'=0}^{M-1} \mathbb{K}^j{}_p \begin{bmatrix} h \\ \alpha\ \alpha' \end{bmatrix} \delta\hat{v}^p \begin{bmatrix} h \\ \alpha' \end{bmatrix}, \quad \forall h\in\mathcal{N}, \tag{2.41}$$

with the "dynamical matrix" $\mathbb{K}^j{}_p \begin{bmatrix} h \\ \alpha\ \alpha' \end{bmatrix}$ defined by

$$\mathbb{K}^j{}_p \begin{bmatrix} h \\ \alpha\ \alpha' \end{bmatrix} \equiv (m_\alpha m_{\alpha'})^{-1/2} \sum_{\ell'\in\mathcal{N}^+} G^{jk} \overset{o}{\Phi}_{kp} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \exp\left\{ -i\mathbf{k}[h]\bullet\left(\mathbf{X}\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - \mathbf{X}\begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \right\}. \tag{2.42}$$

The dynamical matrix, Eq. (2.42), is a $3M\times 3M$ matrix, which has real eigenvalues.[9] It therefore has real eigenvalues denoted by $(\omega^{(q)}[h])^2$, $q=1,2,\ldots,3M$. Assuming exponential solutions, $\delta\hat{v}^j\begin{bmatrix} h \\ \alpha \end{bmatrix} = \Delta\hat{v}^j\begin{bmatrix} h \\ \alpha \end{bmatrix}\exp\{i\omega^{(q)}[h]t\}$, the final eigenvalue problem is

$$(\omega^{(q)}[h])^2 \Delta\hat{v}^j \begin{bmatrix} h \\ \alpha \end{bmatrix}^{(q)} = \sum_{\alpha'=0}^{M-1} \mathbb{K}^j{}_p \begin{bmatrix} h \\ \alpha\ \alpha' \end{bmatrix} \Delta\hat{v}^p \begin{bmatrix} h \\ \alpha' \end{bmatrix}^{(q)}, \quad \forall h\in\mathcal{N}, \quad q=1,2,\ldots,3M. \tag{2.43}$$

The perturbations remain bounded for all time if the square of each phonon frequency $\omega^{(q)}[h]$ is positive. The crystal is phonon-stable if

$$(\omega^{(q)}[h])^2 > 0 \begin{cases} \text{for } h\neq 0, & q=1,2,\ldots,3M, \\ \text{for } h=0, & q=4,5,\ldots,3M, \end{cases} \tag{2.44}$$

[9] Wallace (1998) shows that $\mathbb{K}$ is Hermitian with respect to an orthonormal coordinate system. See Elliott (2004) for a demonstration of its Hermitian nature with respect to the non-orthogonal set of lattice basis vectors used here.

where the second line omits the rigid-body translation modes (labeled $q = 1, 2, 3$ and corresponding to $\mathbf{k}[0] = \mathbf{0}$, since for this special case $\Delta u^j \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = d$) from the stability criterion. All solutions of Eq. (2.28) are given by combining Eqs. (2.33) and (2.40)

$$\delta u^j \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = \frac{1}{\sqrt{m_\alpha n}} \sum_{h \in \mathcal{N}} \sum_{q=1}^{3M} \Delta \widehat{v}^j \begin{bmatrix} h \\ \alpha \end{bmatrix}^{(q)} \exp\left\{ -i \left(\mathbf{k}[h] \bullet \mathbf{X} \begin{bmatrix} \ell \\ \alpha \end{bmatrix} - \omega^{(q)}[h] t \right) \right\}, \tag{2.45}$$

where $\Delta \widehat{v}^j \begin{bmatrix} h \\ \alpha \end{bmatrix}^{(q)}$ is the eigenvector of Eq. (2.43) associated with the eigenvalue $(\omega^{(q)}[h])^2$.

2.2.4. *Zero wave vector limit—acoustic and optic phonons*

As the wave vector $\mathbf{k}$ approaches zero, phonon-stability pertains to long wavelength perturbations of the crystal structure, and a distinction between "acoustic" and "optic" phonon-modes develops. As discussed by Dove (1993), a general 3-D crystal with M atoms per unit cell will have $3M$ different dispersion branches for each wave vector direction. There are three acoustic branches that correspond to rank-one "quasi-uniform" deformations[10] of the crystal in the long wavelength limit. The remaining $3M - 3$ branches are optic branches that represent perturbations with period close to the unit cell spacing in the neighborhood of $\mathbf{k} = \mathbf{0}$. At exactly $\mathbf{k} = \mathbf{0}$ the optic modes exist with non-zero cyclic frequency and pertain to a deformation with unit cell period where the sub-lattices move rigidly with respect to one another. The acoustic modes in the neighborhood of $\mathbf{k} = \mathbf{0}$ represent internally equilibrated long wavelength modes. The acoustic branches at $\mathbf{k} = \mathbf{0}$ correspond to the rigid-body translations of the crystal (with zero cyclic frequency) and do not coincide with their limiting behavior (tending toward a rank-one quasi-uniform perturbation).

In the limit $N \to \infty$, where all unit cells in the infinite crystal are considered (i.e., $\ell \in \mathbb{Z}^3$), the matrix $\mathbb{K}^i{}_j \begin{bmatrix} \mathbf{k} & \\ \alpha & \alpha' \end{bmatrix}$ becomes a continuous, differentiable function of $\mathbf{k}$ except at $\mathbf{k} = \mathbf{0}$. In fact, the $\lim_{\mathbf{k} \to \mathbf{0}} \mathbb{K}^i{}_j \begin{bmatrix} \mathbf{k} & \\ \alpha & \alpha' \end{bmatrix}$ is well defined for $\mathbf{k} \to \mathbf{0}$ only along a fixed direction $\mathbf{n}$ (Wallace, 1998). Thus, any examination of the long wavelength behavior of the crystal must specify a fixed $\mathbf{n}$ direction and expand about the magnitude of $\mathbf{k}$.

Taking $\mathbf{k} = k\mathbf{n}$, $\|\mathbf{n}\| = 1$ the eigenvalue problem (Eq. (2.43)) is

$$(\omega^{(q)}(k\mathbf{n}))^2 \Delta \widehat{v}^j \begin{bmatrix} k\mathbf{n} \\ \alpha \end{bmatrix}^{(q)} = \sum_{\alpha'=0}^{M-1} \mathbb{K}^j{}_r \begin{bmatrix} k\mathbf{n} & \\ \alpha & \alpha' \end{bmatrix} \Delta \widehat{v}^r \begin{bmatrix} k\mathbf{n} \\ \alpha' \end{bmatrix}^{(q)}. \tag{2.46}$$

Expanding the quantities ω and $\Delta \widehat{\mathbf{v}}$ in k (as in Born and Huang (1962) and Wallace (1998)) and suppressing the index q (for notational simplicity) gives

$$\omega(k\mathbf{n}) = \overset{[0]}{\omega}(\mathbf{n}) + \overset{[1]}{\omega}(\mathbf{n}) k + \overset{[2]}{\omega}(\mathbf{n}) \frac{k^2}{2} + \cdots, \tag{2.47}$$

[10] *Note*: the term "homogeneous" is often used (see Born and Huang, 1962; Wallace, 1998), but here the term "quasi-uniform" is introduced in an effort to be more precise. Quasi-uniform deformation should not be confused with a *uniform* or *affine* deformation. A quasi-uniform deformation can be viewed as having sub-lattices that deform uniformly, but shifts between them are still allowed. A uniform or affine deformation, by contrast, has no such internal shifts and all sub-lattices move in "lock-step" with the unit cell deformation.

$$\Delta\widehat{v}^{j}\begin{bmatrix} k\mathbf{n} \\ \alpha \end{bmatrix} = \Delta\overset{[0]}{v}{}^{j}\begin{bmatrix} \mathbf{n} \\ \alpha \end{bmatrix} + \Delta\overset{[1]}{v}{}^{j}\begin{bmatrix} \mathbf{n} \\ \alpha \end{bmatrix} ik + \Delta\overset{[2]}{v}{}^{j}\begin{bmatrix} \mathbf{n} \\ \alpha \end{bmatrix}\frac{k^2}{2} + \cdots. \tag{2.48}$$

The corresponding expansion of $\mathbb{K}$ is

$$\mathbb{K}^{j}{}_{k}\begin{bmatrix} k\mathbf{n} & \\ \alpha & \alpha' \end{bmatrix} = \overset{[0]}{\mathbb{K}}{}^{j}{}_{k}[\alpha\ \alpha'] + \overset{[1]}{\mathbb{K}}{}^{j,p}_{k}[\alpha\ \alpha']n_p ik + \overset{[2]}{\mathbb{K}}{}^{j,pq}_{k}[\alpha\ \alpha']n_p n_q\frac{k^2}{2} + \cdots, \tag{2.49}$$

with the coefficients defined by (now summing over integers in $\mathbb{Z}^3$)

$$\overset{[0]}{\mathbb{K}}{}^{j}{}_{k}[\alpha\ \alpha'] = \frac{1}{\sqrt{m_\alpha m_{\alpha'}}}\sum_{\ell'\in\mathbb{Z}^3} G^{jr}\overset{o}{\Phi}_{rk}\begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix},$$

$$\overset{[1]}{\mathbb{K}}{}^{j,p}_{k}[\alpha\ \alpha'] = \frac{-1}{\sqrt{m_\alpha m_{\alpha'}}}\sum_{\ell'\in\mathbb{Z}^3} G^{jr}\overset{o}{\Phi}_{rk}\begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix}\left(X^p\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p\begin{bmatrix} 0 \\ \alpha \end{bmatrix}\right),$$

$$\begin{aligned}\overset{[2]}{\mathbb{K}}{}^{j,pq}_{k}[\alpha\ \alpha'] = {} & \frac{-1}{\sqrt{m_\alpha m_{\alpha'}}}\sum_{\ell'\in\mathbb{Z}^3} G^{jr}\overset{o}{\Phi}_{rk}\begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix}\left(X^p\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p\begin{bmatrix} 0 \\ \alpha \end{bmatrix}\right) \\ & \times\left(X^q\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^q\begin{bmatrix} 0 \\ \alpha \end{bmatrix}\right).\end{aligned} \tag{2.50}$$

Acoustic phonons: The long wavelength acoustic phonon frequencies are determined up to first order in k, and the resulting expressions are used later to establish the equivalence of long wavelength phonons and elastic wave solutions for the HC model (Section 2.4). The acoustic phonon modes are defined by the property $\omega\to 0$ as $k\to 0$, implying $\overset{[0]}{\omega}(\mathbf{n}) = 0$. In Appendix A, the expansions Eqs. (2.47)–(2.49) are substituted into the eigenvalue problem Eq. (2.46) and a lengthy calculation (following Wallace, 1998) results in an eigenvalue problem for the first order frequencies $\overset{[1]}{\omega}(\mathbf{n})$ and the zeroth-order eigenvectors $\Delta\overset{[0]}{\mathbf{v}}\begin{bmatrix}\mathbf{n}\\ \alpha\end{bmatrix} = \sqrt{m_c m_\alpha}\tilde{\mathbf{v}}(\mathbf{n})$ given by

$$(\overset{[1]}{\omega}(\mathbf{n}))^2\tilde{v}^{j}(\mathbf{n}) = \left[\frac{G^{jk}Z_{k\ l}^{\ p\ q}}{m_c/V}\right]n_p n_q\tilde{v}^{l}(\mathbf{n}),\quad j=1,2,3, \tag{2.51}$$

where m_c is the total mass of one unit cell. The "*dynamical coefficients*" $Z_{k\ l}^{\ p\ q}$ governing the behavior of the long wavelength acoustic phonons are given by

$$\begin{aligned} Z_{k\ l}^{\ p\ q} \equiv \frac{-1}{2V}\sum_{\alpha=0}^{M-1}\Bigg[& \sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}}\overset{o}{\Phi}_{nk}\begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix}\Bigg\{\left(X^p\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p\begin{bmatrix} 0 \\ \alpha \end{bmatrix}\right)\Lambda_l^{n,q}[\alpha] \\ & + \left(X^q\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^q\begin{bmatrix} 0 \\ \alpha \end{bmatrix}\right)\Lambda_l^{n,p}[\alpha]\Bigg\} \\ & + \sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}}\overset{o}{\Phi}_{kl}\begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix}\left(X^p\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p\begin{bmatrix} 0 \\ \alpha \end{bmatrix}\right)\left(X^q\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^q\begin{bmatrix} 0 \\ \alpha \end{bmatrix}\right)\Bigg], \end{aligned} \tag{2.52}$$

where ℓ' is summed over $\mathbb{Z}^3$, α' is summed over $0,\ldots,M-1$; and the quantities $\Lambda_j^{n,p}[\alpha]$ are defined by Eq. (A.16) of Appendix A.

These coefficients describe the crystal's response to rank-one long wavelength traveling waves and are therefore related to the HC behavior of the material.

Optic phonons: The long wavelength optic phonons are distinguished by the property that $\omega \not\to 0$ as $k \to 0$. The zeroth-order (infinite wavelength) optic phonon frequencies are determined by substituting the expansions Eqs. (2.47)–(2.49) into the eigenvalue problem Eq. (2.46). This results in the eigenvalue problem, of dimension $3M-3$, which determines the $3M-3$ (infinite wavelength) optic mode frequencies

$$(\overset{[0]}{\omega}(\mathbf{n}))^2 \Delta \overset{[0]}{v}{}^j \begin{bmatrix} \mathbf{n} \\ \alpha \end{bmatrix} = \frac{1}{\sqrt{m_\alpha m_{\alpha'}}} \sum_{\alpha'=0}^{M-1} \sum_{\ell' \in \mathbb{Z}^3} G^{jp} \overset{o}{\Phi}_{pk} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \Delta \overset{[0]}{v}{}^k \begin{bmatrix} \mathbf{n} \\ \alpha' \end{bmatrix}, \tag{2.53}$$

where one of the $\Delta \overset{[0]}{\mathbf{v}} \begin{bmatrix} \mathbf{n} \\ \alpha \end{bmatrix}$ (say $\alpha = 0$) are set equal to zero so as to eliminate the translational modes. These phonon modes correspond to periodic modes over the unit cell, i.e., rigid translations of the crystal's sub-lattices with zero average strain for the unit cell. Hence, they are related to the CB behavior of the material, as shown in the next section.

2.3. CB continuum model

Often the engineering problem of interest is at a length-scale much larger than the atomic length-scale of Section 2.1. In these cases it is impractical to explicitly consider all atomic DOFs, and a continuum model based on a reduced set of DOFs is desired. Of course, such a model inherently contains less information than the corresponding full atomic model and cannot consider all possible modes of instability. In this subsection the atomic model is homogenized according to CB kinematics, and the new 3-D CB-stability criterion is defined.

2.3.1. CB kinematics

CB kinematics give the current position vectors in terms of the uniform deformation gradient tensor $\mathbf{F}$ and the internal shift vectors $\mathbf{s}[\alpha]$

$$\mathbf{x} \begin{bmatrix} \ell \\ \alpha \end{bmatrix} = \mathbf{F} \bullet \mathbf{X} \begin{bmatrix} \ell \\ \alpha \end{bmatrix} + \mathbf{s}[\alpha], \tag{2.54}$$

where now $\mathbf{F}$ and $\mathbf{s}[\alpha]$ do not necessarily correspond to an equilibrium configuration. The internal shift $\mathbf{s}[0] = \mathbf{0}$ is set to zero to eliminate the three rigid-body translation degrees of freedom. The essential characteristic of the CB kinematic assumption is that it takes a 3-D M-lattice into another 3-D M-lattice. Additionally, as shown below, the equilibrium equations written in terms of CB DOFs ensure that "microscopic"[11] equilibrium of each atom can be obtained.

2.3.2. Equilibrium and CB-stability conditions

Substituting CB kinematics into the total energy density expansion, Eq. (2.18), and introducing the notation

$$\mathbf{s} = \{\mathbf{s}[1], \mathbf{s}[2], \ldots, \mathbf{s}[M-1]\} \tag{2.55}$$

[11]Since the atomic scale is actually the nano-scale, "micro" and "macro" are used in this context to distinguish between discrete and continuum viewpoints, not necessarily to indicate the actual length-scale.

for the internal shift vectors results in the CB energy density

$$\tilde{\mathscr{E}}(\mathbf{F},\mathbf{s}) \equiv \mathscr{E}\left(\mathbf{u}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}(\mathbf{F},\mathbf{s})\right), \tag{2.56}$$

which can be expanded about the configuration $(\overset{o}{\mathbf{F}},\overset{o}{\mathbf{s}})$ as

$$\begin{aligned}\tilde{\mathscr{E}}(\overset{o}{\mathbf{F}}+\delta\mathbf{F},\overset{o}{\mathbf{s}}+\delta\mathbf{s}) = \tilde{\mathscr{E}}(\overset{o}{\mathbf{F}},\overset{o}{\mathbf{s}})\\ &+\frac{1}{nV}\left\{\left(\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\Omega}\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right)\delta F^i{}_j+\sum_{\alpha=1}^{M-1}\left(\sum_{\ell\in\mathscr{N}^+}\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right)\delta s^i[\alpha]\right.\\ &+\frac{1}{2}\left[\left(\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\Omega}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega}\overset{o}{\Phi}_{ij}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}X^k\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^l\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\right)\delta F^i{}_k\delta F^j{}_l\right.\\ &+2n\sum_{\alpha=1}^{M-1}\left(\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega}\overset{o}{\Phi}_{ij}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}X^k\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\right)\delta F^j{}_k\delta s^i[\alpha]\\ &\left.\left.+n\sum_{\alpha=1}^{M-1}\sum_{\alpha'=1}^{M-1}\left(\sum_{\ell'\in\mathscr{N}^+}\overset{o}{\Phi}_{ij}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\right)\delta s^i[\alpha]\delta s^j[\alpha']\right]+\cdots\right\}\\ &-\frac{1}{nV}\left[\left(\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\partial\Omega}f_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right)\delta F^i{}_j+\sum_{\alpha=1}^{M-1}\left(\sum_{\ell\in\partial\Omega}f_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right)\delta s^i[\alpha]\right],\end{aligned} \tag{2.57}$$

where the shorthand notation, $\ell\in\partial\Omega$, indicates the summation over all skeletal lattice points in $\partial\Omega$, and the sums on α range over $1,\ldots,M-1$ (since $\delta\mathbf{s}[0]=\mathbf{0}$). The conditions for configuration $\overset{o}{\mathbf{u}}$ to satisfy equilibrium are given by the first-order derivatives of the CB energy density

$$\left.\frac{\partial\tilde{\mathscr{E}}}{\partial F^i{}_j}\right|_{\overset{o}{\mathbf{F}},\overset{o}{\mathbf{s}}}=\frac{1}{nV}\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\Omega}\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}-\frac{1}{nV}\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\partial\Omega}f_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}=0, \tag{2.58}$$

and

$$\left.\frac{\partial\tilde{\mathscr{E}}}{\partial s^i[\alpha]}\right|_{\overset{o}{\mathbf{F}},\overset{o}{\mathbf{s}}}=\frac{1}{nV}\sum_{\ell\in\mathscr{N}^+}\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}-\frac{1}{nV}\sum_{\ell\in\partial\Omega}f_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}=0. \tag{2.59}$$

Together Eqs. (2.58) and (2.59) imply that "microscopic" equilibrium, Eq. (2.25), is satisfied. To prove this, it is first assumed that the boundary forces $\mathbf{f}\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]$ are applied at the atomic scale to all atoms in the "boundary layer" so that Eq. (2.26) is satisfied for atoms in $\partial\Omega$. If this is not the case, then the boundary layer will exhibit non-uniform deformation which cannot be accommodated by CB kinematics. It remains to be shown that with this assumption Eq. (2.26) is satisfied for the atoms in Ω_0 when Eqs. (2.58) and (2.59) are

satisfied. Eq. (2.59) is now given by

$$\left.\frac{\partial\tilde{\mathscr{E}}}{\partial s^i[\alpha]}\right|_{\overset{o}{\mathbf{F}},\overset{o}{\mathbf{s}}} = \frac{1}{nV}\sum_{\ell\in\Omega_0}\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix} = 0, \tag{2.60}$$

for $\alpha\neq 0$. Using the periodicity condition, Eq. (2.23), $\overset{o}{\Phi}_i\left[\begin{smallmatrix}\ell+l\\ \alpha\end{smallmatrix}\right] = \overset{o}{\Phi}_i\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]$, Eq. (2.60) becomes

$$\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix} = 0, \quad \alpha\neq 0. \tag{2.61}$$

Recalling that $\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}$ represents the sum of forces on an atom, this shows that each atom of type $\alpha\neq 0$ is in force equilibrium. Due to the assumed values for $\mathbf{f}\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]$ Eq. (2.58) reduces to

$$\left.\frac{\partial\tilde{\mathscr{E}}}{\partial F^i{}_j}\right|_{\overset{o}{\mathbf{F}},\overset{o}{\mathbf{s}}} = \frac{1}{nV}\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\Omega_0}\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix} = 0. \tag{2.62}$$

Using the periodicity condition (Eqs. (2.23) and (2.61)), Eq. (2.62) can be written as

$$\overset{o}{\Phi}_i\begin{bmatrix}\ell\\ 0\end{bmatrix}\left(\frac{1}{nV}\sum_{\ell'\in\Omega_0}X^j\begin{bmatrix}\ell'\\ 0\end{bmatrix}\right) = 0. \tag{2.63}$$

It is easily seen that the forces on each atom of type 0 are balanced, i.e., $\overset{o}{\Phi}_i\left[\begin{smallmatrix}\ell\\ 0\end{smallmatrix}\right] = 0$. The reader may notice that the summation of the term in parentheses in Eq. (2.63) is actually zero due to the choice of coordinate system, but for a different choice of coordinate system this summation is not zero. This proves that Eqs. (2.58) and (2.59) assure that every atom in the 3-D crystal is in force equilibrium.

An equilibrium configuration $\overset{o}{\mathbf{u}}$ is CB-stable if it is a local minimum of the energy density $\tilde{\mathscr{E}}(\mathbf{F},\mathbf{s})$. Equivalently, the second derivative of $\tilde{\mathscr{E}}$ must be positive definite with respect to all non-zero *symmetric*[12] right stretch tensor perturbations $\delta\mathbf{U}$ and all non-zero internal shift perturbations $\delta\mathbf{s}$

$$[\delta\mathbf{U},\delta\mathbf{s}]\begin{bmatrix}\dfrac{\partial^2\tilde{\mathscr{E}}}{\partial\mathbf{F}\partial\mathbf{F}} & \dfrac{\partial^2\tilde{\mathscr{E}}}{\partial\mathbf{F}\partial\mathbf{s}}\\ \dfrac{\partial^2\tilde{\mathscr{E}}}{\partial\mathbf{s}\partial\mathbf{F}} & \dfrac{\partial^2\tilde{\mathscr{E}}}{\partial\mathbf{s}\partial\mathbf{s}}\end{bmatrix}_{\overset{o}{\mathbf{F}},\overset{o}{\mathbf{s}}}\begin{bmatrix}\delta\mathbf{U}\\ \delta\mathbf{s}\end{bmatrix}$$
$$= \frac{1}{V}\left\{\left(\frac{1}{n}\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right],\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega}\overset{o}{\Phi}_{im}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^n\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\right)\delta U^i{}_j\delta U^m{}_n\right.$$

[12]The restriction to symmetric deformation tensor perturbations eliminates rigid-body rotations from the stability criterion.

$$+\sum_{\alpha'=1}^{M-1}\left(\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\Omega}\overset{o}{\Phi}_{ir}\begin{bmatrix}\ell & 0\\ \alpha & \alpha'\end{bmatrix}X^{j}\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right)\delta U^{i}{}_{j}\delta s^{r}[\alpha']$$

$$+\sum_{\alpha=1}^{M-1}\left(\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega}\overset{o}{\Phi}_{km}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}X^{n}\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\right)\delta s^{k}[\alpha]\delta U^{m}_{n}$$

$$+\sum_{\alpha,\alpha'=1}^{M-1}\left(\sum_{\ell'\in\mathcal{N}^{+}}\overset{o}{\Phi}_{kr}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\right)\delta s^{k}[\alpha]\delta s^{r}[\alpha']\Bigg\}>0. \tag{2.64}$$

The stability operator has dimension $(3M+3)\times(3M+3)$.

There is overlap between the phonon-stability and CB-stability criteria. If one considers perturbations with no uniform deformation component, $\delta\mathbf{U}=\mathbf{0}$, then the components $\partial^2\tilde{\mathscr{E}}/\partial\mathbf{s}^2$ of the CB-stability matrix, Eq. (2.64) (in the limit as $n\to\infty$, for the infinite crystal), are positive definite only if the optic phonon equations (2.53) have positive eigenvalues, $(\overset{[0]}{\omega}(\mathbf{n}))^2>0$. However, there is significant non-overlap between the perturbations they address. Thus, the phonon- and CB-stability criteria are, in a sense, complementary. The phonon-stability criterion indicates stability with respect to bounded perturbations of all wavelengths. These perturbations approach the rank-one uniform perturbations, in the limit as $\mathbf{k}\to\mathbf{0}$, but do not contain all possible quasi-uniform perturbations. On the other hand, CB-stability indicates stability with respect to all quasi-uniform perturbations but not for all finite wavelength modes.

2.4. Homogenized continuum (HC) model

Eliminating the internal shifts from the CB model to obtain an HC model is a prevalent approach encountered in the literature (Bhattacharya, 2003; Pitteri and Zanzotto, 2002). Physically this may be reasonable, since one can imagine the characteristic time scale for the internal variable's dynamic evolution to be much shorter than the rate of evolution for the uniform deformation $\mathbf{F}$. In this subsection the CB model is reduced to the HC model, the HC-stability criterion is presented, and it is demonstrated that the elastic wave solutions to the equations of motion for the HC model are equivalent to the long wavelength acoustic phonon modes.

The HC energy density is defined by

$$\overset{\approx}{\mathscr{E}}(\mathbf{F})\equiv\tilde{\mathscr{E}}(\mathbf{F},\mathbf{s}(\mathbf{F})), \tag{2.65}$$

where $\mathbf{s}(\mathbf{F})$ is obtained implicitly by Eq. (2.59). Note that this definition does not guarantee that the shifts correspond to local energy minimum. In particular, if a $\mathbf{k}=\mathbf{0}$ optic phonon-mode is unstable, then the shifts will correspond to a local energy maximum with respect to the shift DOFs. Often an energy minimization definition of the HC energy density is advocated (Pitteri and Zanzotto, 2002), e.g. $\overset{\approx}{\mathscr{E}}(\mathbf{F})\equiv\inf_{\mathbf{s}}(\tilde{\mathscr{E}}(\mathbf{F},\mathbf{s}))$. We prefer to adopt the homogenized energy density defined in Eq. (2.65), which is consistent with the long wavelength asymptotic phonon calculation.

2.4.1. Elimination of internal shift DOFs

The non-linear system of equilibrium equations (2.59) is analyzed through the perturbed internal shift equilibrium equations

$$\frac{1}{V}\left[\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{ij}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}X^k\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\delta F^j{}_k + \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{ij}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\delta s^j[\alpha'] + \cdots\right] = 0. \tag{2.66}$$

To first order in $\delta\mathbf{F}$ the internal shift perturbations from their equilibrium values are defined by

$$\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{ij}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\delta s^j[\alpha'] = -\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{ij}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}X^k\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\delta F^j{}_k. \tag{2.67}$$

This expression is closely related to the first order equations (A.7) obtained in the long wavelength acoustic phonon asymptotics of Appendix A. Adding zero in the form $0 = \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{ij}\begin{bmatrix}0 & \ell\\ \alpha & \alpha'\end{bmatrix}X^k\begin{bmatrix}0\\ \alpha\end{bmatrix}$ (due to the translational invariance relations Eq. (2.20)) to the right hand side of Eq. (2.67) and making the following associations

$$(-\delta s^j[\alpha'])\sim\left((m_\alpha m_{\alpha'})^{-1/2}\Delta^{[1]}v^j\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix}\right),\quad \delta F^j{}_k\sim n_k\tilde{v}^j(\mathbf{n}), \tag{2.68}$$

reveals that the form of Eq. (A.7) and Eq. (2.67) are identical. Therefore, following the arguments of Appendix A, the shift perturbations are

$$\delta s^j[\alpha] = -\Lambda^{i,k}_j[\alpha]\delta F^j{}_k, \tag{2.69}$$

where $\Lambda^{i,k}_j[\alpha]$ is defined by Eq. (A.16).

Next, the internal shifts are eliminated from the energy density by the corresponding equilibrium equations. Substitution of Eq. (2.67) in the $(\delta\mathbf{s}[\alpha]\delta\mathbf{s}[\alpha'])$ term of Eq. (2.57) results in a cross term $(\delta\mathbf{F}\delta\mathbf{s}[\alpha])$ which combines with the existing cross term. Then Eq. (2.69) is substituted for the shifts and simplification leads to the HC energy density

$$\begin{aligned}\overset{\approx}{\mathscr{E}}\overset{o}{(}\mathbf{F}+\delta\mathbf{F}) = \overset{\approx}{\mathscr{E}}\overset{o}{(}\mathbf{F}) + \frac{1}{nV}\Bigg\{&\left(\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right)\delta F^i{}_j\\ &+\frac{1}{2}\Bigg(\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\Omega}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{ik}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^l\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\\ &-n\sum_{\alpha=0}^{M-1}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{mi}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}X^j\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\Lambda^{m,l}_k[\alpha]\Bigg)\delta F^i{}_j\delta F^k{}_l + \cdots\Bigg\}\\ &-\frac{1}{nV}\left(\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]\in\partial\Omega} f_i\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^j\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right)\delta F^i{}_j.\end{aligned} \tag{2.70}$$

Because the shifts are implicit (hidden) in the HC description, only the components of $\delta\mathbf{F}$ remain as apparent perturbation DOFs. For brevity, we call these "uniform" perturbations. These are a subset of the (CB) "quasi-uniform" perturbations where the shifts remain as explicit DOFs.

2.4.2. Equilibrium and HC-stability conditions

In general an HC energy density $\overset{\approx}{\mathscr{E}}(\mathbf{F})$ can be expanded as

$$\overset{\approx}{\mathscr{E}}(\overset{o}{\mathbf{F}}+\delta\mathbf{F}) = \overset{\approx}{\mathscr{E}}(\overset{o}{\mathbf{F}}) + \left\{ \Pi_i{}^j \delta F^i{}_j + \frac{1}{2} L_i{}^j{}_k{}^l \delta F^i{}_j \delta F^k{}_l + \cdots \right\} - \Pi_i{}^j \delta F^i{}_j, \tag{2.71}$$

where $\Pi_i{}^j$ are the "first Piola–Kirchhoff" stress components and $L_i{}^j{}_k{}^l$ are the "elastic moduli" components.

The HC equilibrium conditions are

$$\left. \frac{\partial \overset{\approx}{\mathscr{E}}}{\partial F^i{}_j} \right|_{\overset{o}{\mathbf{F}}} = \frac{1}{nV} \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] \in \Omega} \overset{o}{\Phi}_i \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] X^j \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] - \frac{1}{nV} \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] \in \partial\Omega} f_i \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] X^j \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] = 0. \tag{2.72}$$

Comparing Eqs. (2.70) and (2.71) the expression for the applied first Piola–Kirchhoff stress is revealed to be

$$\Pi_i{}^j \equiv \frac{1}{nV} \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] \in \Omega} \overset{o}{\Phi}_i \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] X^j \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] = \frac{1}{nV} \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] \in \partial\Omega} f_i \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] X^j \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right], \tag{2.73}$$

where Eq. (2.72) has been used to obtain the second equality in Eq. (2.73). The elastic moduli for the HC model are similarly found to be

$$L_i{}^j{}_k{}^l \equiv \frac{1}{nV} \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] \in \Omega} \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right] \in \Omega} \overset{o}{\Phi}_{ik} \left[\begin{smallmatrix}\ell & \ell'\\ \alpha & \alpha'\end{smallmatrix}\right] X^j \left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right] X^l \left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right] - \frac{1}{V} \sum_{\alpha=0}^{M-1} \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right] \in \Omega} \overset{o}{\Phi}_{mi} \left[\begin{smallmatrix}0 & \ell'\\ \alpha & \alpha'\end{smallmatrix}\right] X^j \left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right] \Lambda_k^{m,l}[\alpha]. \tag{2.74}$$

The HC-stability criterion, which ensures the equilibrium configuration is an energy minimizer with respect to "uniform" perturbations (consistent with the long wavelength acoustic phonons), is that $L_i{}^j{}_k{}^l$ be positive definite with respect to all symmetric uniform deformation perturbations, or

$$\delta U^i{}_j (L_i{}^j{}_k{}^l) \delta U^k{}_l > 0, \quad \forall \delta\mathbf{U} = \delta\mathbf{U}^{\mathrm{T}} \neq \mathbf{0}. \tag{2.75}$$

The linearized equations of motion for a 3-D continuum are given by

$$\rho \delta \ddot{u}^j = (G^{jk} L_k{}^p{}_l{}^q) \frac{\partial^2 \delta u^l}{\partial X^q \partial X^p}, \tag{2.76}$$

where ρ is the reference mass density. Assuming rank-one traveling plane wave solutions of the form $\delta u^j = \Delta u^j \exp\{-i(\mathbf{k} \bullet \mathbf{X} - \omega(\mathbf{k})t)\}$, substituting L from Eq. (2.74) and considering

the limit as $n \to \infty$ results in

$$\rho(\omega(\mathbf{k}))^2 \Delta u^j = \frac{1}{nV}\left[\sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{kl}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix} X^p\begin{bmatrix}\ell\\ \alpha\end{bmatrix} X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - n\sum_{\alpha=0}^{M-1}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{nk}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix} X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} \Lambda_l^{n,q}[\alpha]\right] k_p k_q \Delta u^l, \tag{2.77}$$

where the sums on ℓ and ℓ' range over $\mathbb{Z}^3$. Noting that the sums over the spatial indices p and q ensure that only the symmetric part of $L_k{}^{p}{}_{l}{}^{q}$ contributes gives

$$\begin{aligned}\rho(\omega(\mathbf{k}))^2 \Delta u^j &= \frac{1}{2nV}\left[-n\sum_{\alpha=0}^{M-1}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{nk}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left\{X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\Lambda_l^{n,q}[\alpha] + X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\Lambda_l^{n,p}[\alpha]\right\}\right.\\ &\left. + \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{kl}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}\left\{X^p\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} + X^q\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\right\}\right] k_p k_q \Delta u^l.\end{aligned} \tag{2.78}$$

Adding the following terms, which are individually zero by the translational invariance relation, Eq. (2.20),

$$\begin{aligned}&\frac{1}{2nV}\left[n\sum_{\alpha=0}^{M-1}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{nk}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left\{X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\Lambda_l^{n,p}[\alpha] + X^q\begin{bmatrix}0\\ \alpha\end{bmatrix}\Lambda_l^{n,p}[\alpha]\right\}\right.\\ &\left. - \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right]}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{kl}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}\left\{X^p\begin{bmatrix}\ell\\ \alpha\end{bmatrix}X^q\begin{bmatrix}\ell\\ \alpha\end{bmatrix} + X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\right\}\right] k_p k_q \Delta u^l = 0\end{aligned} \tag{2.79}$$

to the right hand side of Eq. (2.78), rearranging and evaluating in the interior gives

$$\begin{aligned}\rho(\omega(\mathbf{k}))^2 \Delta u^j = \frac{-1}{2V}&\left[\sum_{\alpha=0}^{M-1}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{nk}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left\{\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\Lambda_l^{n,q}[\alpha]\right.\right.\\ &\left.\left. + \left(X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^q\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\Lambda_l^{n,p}[\alpha]\right\} + \sum_{\alpha=0}^{M-1}\sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]} G^{jk}\overset{o}{\Phi}_{kl}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\right.\\ &\left.\times\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\left(X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^q\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\right] k_p k_q \Delta u^l.\end{aligned} \tag{2.80}$$

Table 1
Number of HC eigenvalues corresponding to rank-one and non-rank-one eigenmodes for the seven crystal symmetry classes and isotropic materials

Symmetry	Number of HC eigenvalues corresponding to	
	Rank-one modes	Non-rank-one modes
Isotropic	5	1
Cubic	5	1
Hexagonal	4	2
Trigonal	4	2
Tetragonal	4	2
Orthorhombic	3	3
Monoclinic	2	4
Triclinic	0	6

Recalling that the reference mass density of the crystal is $\rho = m_c/V$ allows Eq. (2.80) to be written as

$$(\omega(\mathbf{k}))^2 \Delta u^j = \left[\frac{G^{jk} Z_k{}^p{}_l{}^q}{m_c/V}\right] k_p k_q \Delta u^l, \tag{2.81}$$

where the dynamical coefficients $Z_k{}^p{}_l{}^q$ are defined in Eq. (2.52). The eigenvalue problem for the HC (rank-one) elastic waves is identical to Eq. (2.51) for the long wavelength acoustic phonons, thereby proving that they are equivalent. The dynamical coefficients $Z_k{}^q{}_l{}^p = [L_k{}^q{}_l{}^p + L_k{}^q{}_l{}^p]/2$ are the symmetric part (in p, q) of the elastic moduli, Eq. (2.74). Appendix B derives the inverse relationship which establishes the elastic moduli in terms of the dynamical coefficients.

Since the dynamical coefficients are defined in terms of the HC elastic moduli (see Eq. (2.71)), it follows that each of the long wavelength acoustic phonon frequencies can be expressed explicitly in terms of the six HC elastic moduli eigenvalues. Cowley (1976) has classified each HC eigenvalue, for each crystal symmetry class, based on the existence of a phonon frequency which goes to zero when the corresponding HC eigenvalue vanishes.[13] This classification identifies the number of eigenvalues corresponding to rank-one eigenmodes. Table 1 presents the number of HC eigenvalues corresponding to rank-one and non-rank-one eigenmodes for each of the seven crystal symmetry systems and for an isotropic material. As can be seen, HC rank-one convexity, in general, is not sufficient to guarantee HC-stability.

3. Discussion of phonon-, CB- and HC-stability criteria

Each of the three stability criteria considered interrogates a different set of perturbations and no single set encompasses the other two; however, some overlap exists. Therefore, this section discusses the relative strengths of the different stability criteria. A Venn-like diagram of the space of all perturbations of an infinite perfect M-lattice about an equilibrium state is shown in Fig. 2, and Table 2 presents a comparison of the five disjoint sets (a)–(e) that make up the space of perturbations collectively considered by the three

[13] See also Terhune et al. (1985), for corrections to the results in Cowley (1976).

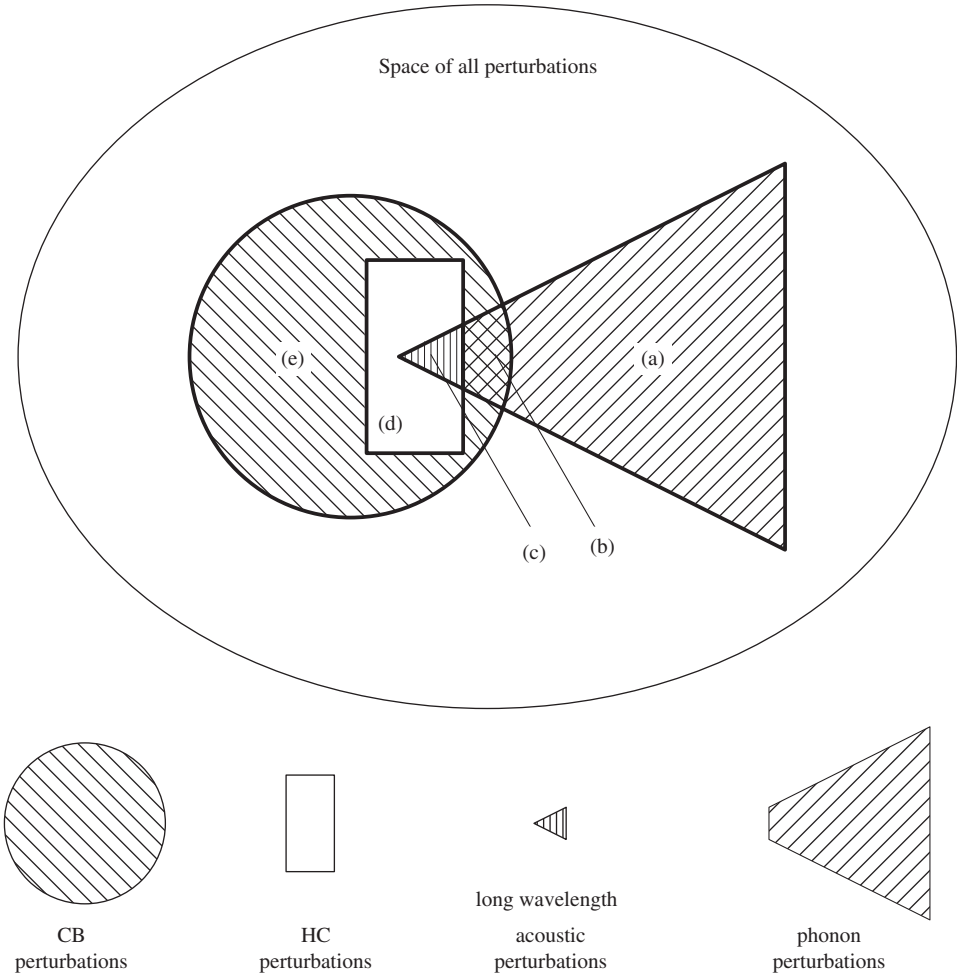

Fig. 2. Perturbation space for perfect M-lattices showing: (a) bounded perturbations of all wavelengths; (b) quasi-uniform perturbations corresponding to optic phonon modes; (c) rank-one "uniform" perturbations corresponding to acoustic phonon modes; (d) non-rank-one "uniform" perturbations; and (e) all other quasi-uniform perturbations.

stability criteria. Set (a) consists of bounded perturbations (of an infinite crystal) of all wavelengths where $\mathbf{k} \neq \mathbf{0}$ and no macroscopic deformation of the crystal exists, since $\delta\mathbf{U} = \mathbf{0}$. All atomic DOFs are explicitly considered in the perturbation and thus, both optic and acoustic phonon modes are included. The atomic DOFs for the remaining perturbation sets (b)–(e) conform to CB kinematics. Set (b) consists of the infinite wavelength ($\mathbf{k} = \mathbf{0}$) optic phonon modes where the sub-lattices translate rigidly with respect to one another, i.e., $\delta\mathbf{U} = \mathbf{0}$ but $\delta\mathbf{s}[\alpha] \neq \mathbf{0}$. Set (c) consists of the rank-one "uniform" perturbations where $\delta\mathbf{F} = \delta\mathbf{a} \otimes \mathbf{n}$ (or equivalently $\delta\mathbf{U} = (\delta\mathbf{a} \otimes \mathbf{n})_s$, where the subscript s indicates the symmetric part of the quantity in parenthesis) and the internal shift perturbations are constrained according to equilibrium. Set (d) consists of the non-rank-one "uniform" perturbations where again the internal shifts are constrained according to

Table 2
Comparison of the five perturbation sets of Fig. 2

Criterion	Set	$\mathbf{k}$	$\delta\mathbf{U}$	$\delta\mathbf{s}$	All perturbation atomic DOFs	Modes included	
						Optic	Acoustic
phonon	(a)	$\neq\mathbf{0}$	$\mathbf{0}$	N/A	unconstrained	yes	yes
	(b)	$\mathbf{0}$	$\mathbf{0}$	unconstrained	CB-constrained	yes	no
CB, HC	(c)	$\mathbf{0}$	$(\delta\mathbf{a}\otimes\mathbf{n})_s$	equil. constrained	HC-constrained	no	yes
CB, HC	(d)	$\mathbf{0}$	$\neq(\delta\mathbf{a}\otimes\mathbf{n})_s$	equil. constrained	HC-constrained	N/A	N/A
CB	(e)	$\mathbf{0}$	$\neq\mathbf{0}$	unconstrained	CB-constrained	N/A	N/A

equilibrium. Set (e) consists of all quasi-uniform perturbations that have non-zero $\delta\mathbf{U}$ and independent $\delta\mathbf{s}$ perturbations.

The leftmost column of Table 2 indicates the sets of perturbations considered by the three stability criteria. Thus, the phonon-stability criterion investigates the trapezoidal region of perturbations in Fig. 2 given by the union of sets (a) and (b). The CB-stability criterion investigates the circular region (Fig. 2) of perturbations given by the union of sets (b)–(e). The HC-stability criterion investigates the rectangular region of perturbations given by the union of sets (c) and (d). Stability information about the set of perturbations (c) can also be obtained with Born's method of long waves, consisting of an asymptotic analysis of the long wavelength acoustic phonon modes. It should be noted that the set of CB perturbations depends on the choice of M-lattice. However, the phonon perturbations of an equilibrium configuration are independent of this choice. This important point is discussed further in the context of the application presented in Part II.

Stability of a solid depends on the type of loading device used. For hard-device loading (which imposes displacement conditions on the entire boundary of the body), classical continuum mechanics indicates that HC rank-one-convexity (stability of region (c) in Fig. 2) is necessary and sufficient for stability. When information is available about the microstructure of a material, the HC rank-one-convexity criterion must be extended down to all appropriate length scales. The phonon-stability criterion does exactly this for the crystalline models considered here. Thus, for hard-device loading (which imposes periodic displacement conditions on the entire boundary of the body), necessary and sufficient conditions for crystal stability are phonon-stability and its limiting behavior (HC rank-one-convexity), i.e., stability with respect to all perturbation modes in regions (a)–(c) of Fig. 2. Stability of this type is called "constrained material stability".

Stability with respect to soft-device loading is more difficult to establish. Stability under soft-device loading conditions (i.e., where displacement on part of the boundary is not constrained) requires consideration of a boundary value problem, which includes "structural" modes such as bending (strain gradient) or surface modes. Thus, no constitutive-level criterion can provide necessary and sufficient conditions for stability. Therefore, our goal is to propose the strongest possible *necessary* criterion for stability under soft-device loading conditions. In this spirit, necessary conditions for stability require the equilibrium configuration to correspond to a local energy minimum, point-wise at the homogenized scale. This is ensured by requiring both CB- and phonon-stability criteria to be satisfied. Note that the CB criterion is chosen in preference to the HC

criterion because the CB criterion interrogates all quasi-uniform perturbations of the energy density, some of which are ignored by the HC criterion. It is, therefore, proposed that "material stability" be defined such that *both* phonon- and CB-stability are satisfied, i.e., stability with respect to *all* perturbations in regions (a)–(e) of Fig. 2.

4. Summary and conclusions

The stability of crystalline solids is a fundamental problem addressed by the physics, materials science, and mechanics communities. There is strong interest in this subject due to novel engineering applications for phase transformations. This work investigates equilibrium configurations and stability properties of multi-atomic perfect crystals from an atomic viewpoint. Stability criteria with respect to perturbations at the atomic scale (phonon-stability) and the continuum scale (homogenized-continuum-stability) are reviewed and a new stability criterion (CB-stability) is introduced that provides an intermediate criterion by considering perturbations at both the atomistic and continuum scales. The goal of this work is to provide a unified presentation of these stability criteria for crystalline solids in equilibrium configurations describable by CB kinematics (uniform deformation and internal shifts of sub-lattices) and to directly compare their relative strengths and weaknesses.

Phonon-stability is defined in terms of the normal modes of vibration (or phonons) for the crystal. It requires that the natural frequency for each phonon be a real quantity, or equivalently, that the eigenvalues of the crystal's stiffness matrix with respect to all atomic DOFs be positive definite. Phonon modes produce no average deformation in the crystal but instead result in periodic displacements of arbitrary wavelength. The set of all phonon modes for an infinite crystal constitute all possible bounded perturbations within a crystal. However, the phonon-stability criterion does not directly provide stability information for unbounded perturbations such as a uniform deformation gradient. The determination of the natural frequencies for such a crystal, and thus evaluation of the phonon-stability criterion, is simplified by application of a block-Fourier transformation to the crystal's equations of motion that results in a block-diagonalized stiffness matrix. In Fourier space the phonon modes, which are labeled by the Fourier "wave vector" that indicates their wavelength and direction of propagation, are classified as either "acoustic" phonons, which have a finite group velocity in the long wavelength limit, or "optic" phonons, which have zero group velocity in the long wavelength limit. Although asymptotic expressions for rank-one "uniform" perturbations of the crystal can be recovered from the phonon mode calculations (via Born's method of long waves), this information is more conveniently obtained with the HC- or CB-stability criteria.

CB stability is introduced to investigate stability of the crystal with respect to independent perturbations in the uniform deformation gradient and the internal shifts, which together comprise the so-called "quasi-uniform" deformations (i.e., those allowed under CB kinematics). The CB-stability criterion bridges the length scales between the phonon- and HC-stability criteria by accounting for the atomic scale internal shifts (optic phonon modes) and a superset of the continuum scale uniform perturbations considered by the HC-stability criterion.

HC-stability considers "uniform" perturbations of the crystal structure where the "internal shifts" are required to take on equilibrium values and are thus "condensed out" of the crystal's kinematics. This criterion demands that the crystal's homogenized elastic modulus tensor be positive definite. HC-stability is strictly weaker than CB-stability.

For hard-device loading conditions, phonon-stability and its limiting behavior (HC rank-one-convexity) are necessary and sufficient for stability which we call "constrained material stability". For soft-device loading conditions, it is clear that phonon-stability *and* CB-stability are necessary (but not sufficient due to possible "structural" modes), and we take this as our proposed definition of "material stability".

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Appendix A. Asymptotic calculations for acoustic phonon branches

Born's method of long waves was the first to make a direct connection between the stability of discrete and continuum views of crystals. The method considers the acoustic phonon modes and analyzes the behavior of phonon dispersion relations in the limit as $\mathbf{k} \to \mathbf{0}$ along a fixed direction $\mathbf{n}$. This analysis is required to perform a comparison of the phonon-stability and HC-stability criteria. The current presentation essentially follows that of Wallace (1998), but is formulated in a non-orthogonal coordinate system and with respect to the reference equilibrium configuration rather than the current equilibrium configuration.

The acoustic phonon modes are defined by the property $\omega \to 0$ as $k \to 0$, thereby implying $\overset{[0]}{\omega}(\mathbf{n}) = 0$. Substituting the expansions (Eqs. (2.47)–(2.49)) into the eigenvalue problem (Eq. (2.46)) and grouping terms of like order in k results in the following:

- zeroth-order terms

$$\sum_{\alpha'=0}^{M-1} \overset{[0]}{\mathbb{K}}{}^{i}{}_{j}[\alpha\ \alpha']\Delta \overset{[0]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix} = 0, \tag{A.1}$$

- first-order terms

$$\sum_{\alpha'=0}^{M-1}\left(\overset{[0]}{\mathbb{K}}{}^{i}{}_{j}[\alpha\ \alpha']\Delta \overset{[1]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix} + \overset{[1]}{\mathbb{K}}{}^{i,p}_{j}[\alpha\ \alpha']n_p\Delta \overset{[0]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix}\right) = 0, \tag{A.2}$$

- second-order terms

$$(\overset{[1]}{\omega}(\mathbf{n}))^2\Delta \overset{[0]}{v}{}^{i}\begin{bmatrix}\mathbf{n}\\ \alpha\end{bmatrix} = \frac{1}{2}\sum_{\alpha'=0}^{M-1}\left(\overset{[0]}{\mathbb{K}}{}^{i}{}_{j}[\alpha\ \alpha']\Delta \overset{[2]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix} - 2\,\overset{[1]}{\mathbb{K}}{}^{i,p}_{j}[\alpha\ \alpha']n_p\Delta \overset{[1]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix} + \overset{[2]}{\mathbb{K}}{}^{i,pq}_{j}[\alpha\ \alpha']n_pn_q\Delta \overset{[0]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix}\right). \tag{A.3}$$

The zeroth-order equations have solutions

$$\Delta \overset{[0]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha\end{bmatrix} = \sqrt{m_{\mathrm{c}} m_\alpha}\,\tilde{v}^j(\mathbf{n}), \tag{A.4}$$

with $m_{\mathrm{c}} = \sum_\alpha m_\alpha$ and $\tilde{\mathbf{v}}(\mathbf{n})$ a constant vector. Substituting into Eq. (A.1) gives

$$\sum_{\alpha'=0}^{M-1} \frac{G^{ir}}{\sqrt{m_\alpha m_{\alpha'}}} \sum_{\ell' \in \mathcal{N}^+} \overset{o}{\Phi}_{rj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(\sqrt{m_{\mathrm{c}} m_{\alpha'}}\,\tilde{v}^j(\mathbf{n})\right) = \sqrt{\frac{m_{\mathrm{c}}}{m_\alpha}} G^{ir}\left[\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \overset{o}{\Phi}_{rj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\right]\tilde{v}^j(\mathbf{n}) = 0, \tag{A.5}$$

where the last equality is due to translational invariance, Eq. (2.20). In $\mathbb{R}^3$ there are three linearly independent solutions $\tilde{\mathbf{v}}^{(q)}(\mathbf{n})$, corresponding to acoustic waves, labeled $q = 1, 2, 3$ for each wave vector direction $\mathbf{n}$. One of these acoustic waves corresponds to a longitudinal mode with atomic motion in the direction of $\mathbf{n}$ and the remaining two correspond to transverse modes with atomic motion perpendicular to $\mathbf{n}$. The remaining $3M - 3$ long wavelength modes (solutions to Eq. (2.46) with non-zero $\overset{[0]}{\omega}(\mathbf{n})$) correspond to optic phonon branches which are not of interest here.

Next, Eqs. $(2.50)_{1,2}$ and the zeroth-order solution Eq. (A.4) are substituted into the first-order equations, giving

$$\begin{aligned}&\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \frac{G^{ir}}{\sqrt{m_\alpha m_{\alpha'}}} \overset{o}{\Phi}_{rj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\Delta \overset{[1]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix}\\ &\quad = \sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \sqrt{\frac{m_{\mathrm{c}}}{m_\alpha}} G^{ir} \overset{o}{\Phi}_{rj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right) n_p \tilde{v}^j(\mathbf{n}).\end{aligned} \tag{A.6}$$

Multiplying by $(m_\alpha/m_{\mathrm{c}})^{1/2}$ and G_{ni} gives

$$\begin{aligned}&\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left[(m_{\mathrm{c}} m_{\alpha'})^{-1/2}\Delta \overset{[1]}{v}{}^{j}\begin{bmatrix}\mathbf{n}\\ \alpha'\end{bmatrix}\right]\\ &\quad = \sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right) n_p \tilde{v}^j(\mathbf{n}).\end{aligned} \tag{A.7}$$

The homogeneous part of this equation (left hand side) has an arbitrary constant vector $\mathbf{T}$ as its solution, since

$$\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix} T^j = 0, \tag{A.8}$$

by translational invariance. Thus, the inhomogeneous part (right hand side of Eq. (A.7)) must be orthogonal to an arbitrary constant vector for a unique solution

to exist, i.e.,

$$\sum_{\alpha=0}^{M-1} T^n \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right) n_p \tilde{v}^j(\mathbf{n}) = 0. \tag{A.9}$$

This is satisfied by neglecting surface effects, as can be shown by starting from the identity (due to translational invariance)

$$\begin{aligned} 0 &= \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right],\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix} X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right],\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix} X^p\begin{bmatrix}\ell\\ \alpha\end{bmatrix} \\ &= \sum_{\left[\begin{smallmatrix}\ell\\ \alpha\end{smallmatrix}\right],\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}\ell & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}\ell\\ \alpha\end{bmatrix}\right), \end{aligned} \tag{A.10}$$

and evaluating this in the interior of the crystal gives

$$0 = n\sum_{\alpha=0}^{M-1} \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right). \tag{A.11}$$

The solvability condition (A.9) is satisfied and the solution to the first-order equations is arbitrary up to a constant vector. This ambiguity is eliminated from the equations by setting one of the $\Delta^{[1]}\mathbf{v}\begin{bmatrix}\mathbf{n}\\ \alpha\end{bmatrix}$ to zero, say,

$$\Delta^{[1]}\mathbf{v}\begin{bmatrix}\mathbf{n}\\ 0\end{bmatrix} = \mathbf{0}. \tag{A.12}$$

Restricting Eq. (A.7) to $\alpha, \alpha' \neq 0$, the system in general becomes non-singular and can be solved by inverting the left hand side of (A.7). Defining $\tilde{\Gamma}^{ij}[\alpha\ \alpha']$ for $\alpha, \alpha' \neq 0$ as the inverse of $\overset{o}{\Phi}$, then $\tilde{\Gamma}^{ij}[\alpha\ \alpha']$ satisfies

$$\sum_{\alpha''=1}^{M-1} \tilde{\Gamma}^{ij}[\alpha\ \alpha''] \sum_{\ell'\in\mathcal{N}^+} \overset{o}{\Phi}_{jk}\begin{bmatrix}0 & \ell'\\ \alpha'' & \alpha'\end{bmatrix} = \delta^i_k \delta_{\alpha\alpha'}, \quad \alpha, \alpha', \alpha'' \neq 0. \tag{A.13}$$

Next, the matrix $\tilde{\Gamma}^{ij}[\alpha\ \alpha']$ is augmented to include rows and columns of zeros in the $\alpha, \alpha' = 0$ positions,

$$\Gamma^{ij}[\alpha\ \alpha'] \equiv \begin{cases} \tilde{\Gamma}^{ij}[\alpha\ \alpha'], & \alpha, \alpha' \neq 0, \\ 0, & \text{otherwise}, \end{cases} \tag{A.14}$$

and a general expression for the solution of Eq. (A.2) is obtained as

$$\Delta^{[1]}v^j\begin{bmatrix}\mathbf{n}\\ \alpha\end{bmatrix} = (m_c m_\alpha)^{1/2} \sum_{\alpha''=0}^{M-1} \Gamma^{in}[\alpha\ \alpha''] \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \overset{o}{\Phi}_{nj}\begin{bmatrix}0 & \ell'\\ \alpha'' & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha''\end{bmatrix}\right) n_p \tilde{v}^j(\mathbf{n}). \tag{A.15}$$

For convenience, the coefficients $\Lambda^{i,p}_j[\alpha]$ are defined as

$$\Lambda^{i,p}_j[\alpha] = \sum_{\alpha''=0}^{M-1} \sum_{\left[\begin{smallmatrix}\ell'\\ \alpha'\end{smallmatrix}\right]\in\Omega} \Gamma^{in}[\alpha\ \alpha'']\overset{o}{\Phi}_{nj}\begin{bmatrix}0 & \ell'\\ \alpha'' & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha''\end{bmatrix}\right), \tag{A.16}$$

giving

$$\Delta^{[1]}_i \begin{bmatrix} \mathbf{n} \\ \alpha \end{bmatrix} = (m_c m_\alpha)^{1/2} \Lambda^{i,p}_j[\alpha] n_p \tilde{v}^j(\mathbf{n}). \tag{A.17}$$

Next, Eqs. (A.4) and (A.17) are substituted into the second-order equations (A.3) and rearranged to obtain

$$\begin{aligned}
&\sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} (m_\alpha m_{\alpha'})^{-1/2} \overset{o}{\Phi}_{ij} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \Delta^{[2]}_j \begin{bmatrix} \mathbf{n} \\ \alpha' \end{bmatrix} \\
&\quad = \left(\frac{m_c}{m_\alpha}\right)^{1/2} \sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} \overset{o}{\Phi}_{ij} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \left(X^p \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \left(X^q \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^q \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) n_p n_q \tilde{v}^j(\mathbf{n}) \\
&\qquad - 2\left(\frac{m_c}{m_\alpha}\right)^{1/2} \sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} \overset{o}{\Phi}_{ij} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \left(X^p \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \Lambda^{j,r}_q[\alpha'] n_p n_r \tilde{v}^q(\mathbf{n}) \\
&\qquad + 2(m_c m_\alpha)^{1/2} (\overset{[1]}{\omega}(\mathbf{n}))^2 G_{ij} \tilde{v}^j(\mathbf{n}).
\end{aligned} \tag{A.18}$$

The homogeneous part has solutions $(m_{\alpha'})^{1/2} T^j$ ($\mathbf{T}$ an arbitrary vector) due to translational invariance. The solvability condition for (A.18) is similar to Eq. (A.9) and requires that the right hand side of Eq. (A.18) (multiplied by $(m_\alpha)^{1/2}$ and summed over α) be zero, which written explicitly is

$$\begin{aligned}
&\sum_{\alpha=0}^{M-1} \Bigg[(m_c)^{1/2} \sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} \overset{o}{\Phi}_{ij} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \left(X^p \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \left(X^q \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^q \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) n_p n_q \tilde{v}^j(\mathbf{n}) \\
&\qquad - 2(m_c)^{1/2} \sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} \overset{o}{\Phi}_{ij} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \left(X^p \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \Lambda^{j,r}_q[\alpha'] n_p n_r \tilde{v}^q(\mathbf{n}) \\
&\qquad + 2(m_c)^{3/2} (\overset{[1]}{\omega}(\mathbf{n}))^2 G_{ij} \tilde{v}^j \begin{bmatrix} \mathbf{n} \\ q \end{bmatrix} \Bigg] = 0.
\end{aligned} \tag{A.19}$$

Rearranging gives

$$\begin{aligned}
m_c \left(\overset{[1]}{\omega}(\mathbf{n}) \right)^2 \tilde{v}^i(\mathbf{n}) = \Bigg[&\sum_{\alpha=0}^{M-1} \sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} G^{ik} \overset{o}{\Phi}_{kn} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \left(X^p \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \Lambda^{n,q}_j[\alpha'] \\
&- \frac{1}{2} \sum_{\alpha=0}^{M-1} \sum_{\begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} \in \Omega} G^{ik} \overset{o}{\Phi}_{kj} \begin{bmatrix} 0 & \ell' \\ \alpha & \alpha' \end{bmatrix} \left(X^p \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^p \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \\
&\times \left(X^q \begin{bmatrix} \ell' \\ \alpha' \end{bmatrix} - X^q \begin{bmatrix} 0 \\ \alpha \end{bmatrix} \right) \Bigg] n_p n_q \tilde{v}^j(\mathbf{n}),
\end{aligned} \tag{A.20}$$

which is an eigenvalue problem for the first-order coefficient $\overset{[1]}{\omega}(\mathbf{n})$ of the acoustic phonon frequencies. The following series of operations reveal the coordinate system invariance of the first term on the right hand side. The quantities $\overset{o}{\Phi}_{kn}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}$ are replaced by $\overset{o}{\Phi}_{nk}\begin{bmatrix}\ell' & 0\\ \alpha' & \alpha\end{bmatrix}$, and the periodicity relation is used to obtain $\overset{o}{\Phi}_{nk}\begin{bmatrix}0-\ell' \\ \alpha' & \alpha\end{bmatrix}$. Next, $-\ell'$ is relabeled ℓ'' and it is noted that $(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix})$ can now be written as $(-X^p\begin{bmatrix}\ell''\\ \alpha\end{bmatrix} + X^p\begin{bmatrix}0\\ \alpha'\end{bmatrix})$.Finally, the dummy indices α and α' are switched to obtain

$$
\begin{aligned}
m_c\left(\overset{[1]}{\omega}(\mathbf{n})\right)^2\tilde{v}^i(\mathbf{n}) = -\Bigg[&\sum_{\alpha=0}^{M-1}\sum_{\begin{bmatrix}\ell''\\ \alpha'\end{bmatrix}\in\Omega} G^{ik}\overset{o}{\Phi}_{nk}\begin{bmatrix}0 & \ell''\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell''\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\Lambda_j^{n,q}[\alpha]\\
&+\frac{1}{2}\sum_{\alpha=0}^{M-1}\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega} G^{ik}\overset{o}{\Phi}_{kj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\\
&\times\left(X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^q\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\Bigg] n_p n_q\tilde{v}^j(\mathbf{n}).
\end{aligned}
\tag{A.21}
$$

Note that the Einstein sums over p and q ensure that only the symmetric part of the right hand side contributes, and normalizing by the unit cell volume, gives

$$
\left(\overset{[1]}{\omega}(\mathbf{n})\right)^2\tilde{v}^i(\mathbf{n}) = \left[\frac{G^{ik}Z_{k\ j}^{\ p\ q}}{m_c/V}\right]n_p n_q\tilde{v}^j(\mathbf{n}),
\tag{A.22}
$$

where $Z_{k\ j}^{\ p\ q}$ are the dynamical coefficients governing the behavior of the long wavelength acoustic phonons defined by

$$
\begin{aligned}
Z_{k\ j}^{\ p\ q} \equiv \frac{-1}{2V}\sum_{\alpha=0}^{M-1}\Bigg[&\sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega}\overset{o}{\Phi}_{nk}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\Bigg\{\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\Lambda_j^{n,q}[\alpha]\\
&+\left(X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^q\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\Lambda_j^{n,p}[\alpha]\Bigg\} + \sum_{\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix}\in\Omega}\overset{o}{\Phi}_{kj}\begin{bmatrix}0 & \ell'\\ \alpha & \alpha'\end{bmatrix}\left(X^p\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^p\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\\
&\times\left(X^q\begin{bmatrix}\ell'\\ \alpha'\end{bmatrix} - X^q\begin{bmatrix}0\\ \alpha\end{bmatrix}\right)\Bigg].
\end{aligned}
\tag{A.23}
$$

Eqs. (A.22) and (A.23) are the results that are repeated in Eqs. (2.51) and (2.52).

Appendix B. Homogenized continuum elastic moduli

In Section 2.4.2, a relationship between the HC elastic moduli, Eq. (2.74), and the dynamical coefficients, Eq. (2.52), is established. In this appendix the inverse relationship is

derived. Following Born and Huang (1962), an argument based on the symmetry of the HC elastic moduli C^{ijkl} (with respect to the Lagrangian strain) is used to establish the HC elastic moduli $L_i^{\ j}{}_k^{\ l}$ (with respect to the deformation gradient) in terms of the dynamical coefficients, $Z_i^{\ j}{}_k^{\ l}$, the first Piola–Kirchhoff stress, $\mathbf{\Pi}$, and the uniform deformation gradient, $\mathbf{F}$.

The dynamical coefficients, Eq. (2.52), for long wavelength acoustic phonons and HC elastic waves are the symmetric part of the full HC elastic moduli, Eq. (2.74),

$$Z_k^{\ p}{}_l^{\ q} = \tfrac{1}{2}[L_k^{\ p}{}_l^{\ q} + L_k^{\ q}{}_l^{\ p}]. \tag{B.1}$$

Here the inverse relation is derived in general by taking advantage of the full symmetry of the HC elastic moduli and applied stress with respect to the Lagrangian strain tensor, which is given by

$$E_{ij} \equiv \tfrac{1}{2}(F^s{}_i F_{sj} - G_{ij}), \tag{B.2}$$

where $F_{sj} = G_{sr}F^r{}_j$. Thus, expanding the HC internal energy density in powers of the Lagrangian strain gives

$$\overset{\approx}{W}(\mathbf{E}+\delta\mathbf{E}) = \overset{\approx}{W}(\mathbf{E}) + S^{ij}\delta E_{ij} + \tfrac{1}{2}C^{ijkl}\delta E_{ij}\delta E_{kl} + \cdots, \tag{B.3}$$

where S^{ij} are the components of the *second Piola–Kirchhoff* stress tensor and C^{ijkl} are the HC elastic moduli. These quantities have the following symmetries due to the symmetry of the Lagrangian strain tensor and their definition as derivatives of the scalar HC energy density

$$S^{ij} = S^{ji}, \tag{B.4}$$

$$C^{ijkl} = C^{jikl} = C^{ijlk} = C^{klij}. \tag{B.5}$$

Using the chain-rule leads to expressions for the *first Piola–Kirchhoff* stress $\mathbf{\Pi}$ (of Eq. (2.73)) and the elastic moduli $\mathbf{L}$ (of Eq. (2.74))

$$\Pi_i^{\ j} = S^{jp}F_{ip}, \tag{B.6}$$

$$L_i^{\ j}{}_k^{\ l} = F_{ip}C^{pjql}F_{kq} + S^{jl}G_{ik}. \tag{B.7}$$

Solving this for the elastic moduli C^{ijkl} (and defining $H^{ij} \equiv (F_{ij})^{-1}$) gives

$$C^{ijkl} = L_p^{\ i}{}_q^{\ l}H^{kq}H^{jp} - S^{il}H^{kp}G_{pq}H^{jq}. \tag{B.8}$$

Taking the symmetric part of C^{ijkl} (in i,l) results in

$$\tfrac{1}{2}[C^{ijkl} + C^{ljki}] = [Z_p^{\ i}{}_q^{\ l} - S^{il}G_{pq}]H^{kq}H^{jp}, \tag{B.9}$$

exchanging i and j in Eq. (B.9) gives

$$\tfrac{1}{2}[C^{jikl} + C^{likj}] = [Z_p^{\ j}{}_q^{\ l} - S^{jl}G_{pq}]H^{kq}H^{ip}, \tag{B.10}$$

and interchanging j and k in Eq. (B.10) leads to

$$\tfrac{1}{2}[C^{kijl} + C^{lijk}] = [Z_p^{\ k}{}_q^{\ l} - S^{kl}G_{pq}]H^{jq}H^{ip}. \tag{B.11}$$

Now, taking the sum of Eqs. (B.9) and (B.10) and subtracting Eq. (B.11) one finds that the left hand side reduces to C^{ijkl} giving

$$C^{ijkl} = [Z_p{}^{i}{}_{q}{}^{l} H^{kq} H^{jp} + Z_p{}^{j}{}_{q}{}^{l} H^{hq} H^{ip} - Z_p{}^{k}{}_{q}{}^{l} H^{jq} H^{ip}] - [S^{il} G_{pq} H^{kq} H^{jp} + S^{jl} G_{pq} H^{kq} H^{ip} - S^{kl} G_{pq} H^{jq} H^{ip}]. \quad \text{(B.12)}$$

Substituting Eq. (B.12) into Eq. (B.7) and using Eq. (B.6) leads to the final result expressing the elastic moduli **L** with respect to the deformation gradient **F** in terms of the dynamical coefficients **Z**, the first Piola–Kirchhoff stress, **Π**, and the deformation gradient, **F**,

$$L_i{}^{j}{}_{k}{}^{l} = Z_i{}^{j}{}_{k}{}^{l} + F_{iq} H^{jp} Z_p{}^{q}{}_{k}{}^{l} - F_{kq} H^{jp} Z_p{}^{q}{}_{i}{}^{l} - \Pi_i{}^{l} H^{j}{}_{k} + \Pi_k{}^{l} H^{j}{}_{i}, \quad \text{(B.13)}$$

where $H^{i}{}_{j} \equiv (F_i{}^{j})^{-1} = H^{ip} G_{pj}$.

References

Bhattacharya, K., 2003. Microstructure of martensite: Why it forms and how it gives rise to the shape-memory effect. Oxford Series on Materials Modelling, first ed. Oxford University Press, Oxford.

Born, M., Huang, K., 1962. Dynamical Theory of Crystal Lattices, first ed. Oxford University Press, Oxford.

Cowley, R.A., 1976. Acoustic phonon instabilities and structural phase transitions. Phys. Rev. B: Condensed Matter 13 (11), 4877–4885.

Davis, P.J., 1994. Circulant Matrices, second ed. Chelsea Publishing, New York, NY.

Dove, M.T., 1993. Introduction to Lattice Dynamics. Cambridge University Press, Cambridge.

Elliott, R.S., 2004. Lattice-level instabilities in bi-atomic alloys. Ph.D. thesis, Department of Aerospace Engineering, The University of Michigan, Ann Arbor, MI, USA.

Elliott, R.S., Shaw, J.A., Triantafyllidis, N., 2004. Stability of crystalline solids—II: Application to temperature-induced martensitic phase transformations in bi-atomic crystals. J. Mech. Phys. Solids, submitted for publication, doi:10.1016/j.jmps.2005.07.008.

Geymonat, G., Müller, S., Triantafyllidis, N., 1993. Homogenization of nonlinearly elastic materials, microscopic bifurcation and macroscopic loss of rank-one convexity. Arch. Ration. Mech. Anal. 122, 231–290.

Huang, X., Bungaro, C., Godlevsky, V., Rabe, K.M., 2002. Lattice instabilities of cubic NiTi from first principles. Phys. Rev. B: Condensed Matter 65(1):014108/1–5.

James, R.D., 1987. Metastability and incompletely posed problems. In: The stability and metastability of quartz. IMA Volumes in Mathematics and its Applications, vol. 3. Springer, Berlin, pp. 147–175.

Kastner, O., 2003. Molecular dynamics of a 2D model of the shape memory effect Part. i: Model and simulations. Continuum Mech. Thermodyn. 15 (5), 487–502.

Kittel, C., 1956. Introduction to Solid State Physics, second ed. Wiley, New York.

Miller, W., Jr., 1972. Symmetry Groups and Their Applications, Pure and Applied Mathematics, vol. 50, first ed. Academic Press, New York.

Otsuka, K., Wayman, C.M., (Eds), 1998. Shape Memory Materials. Cambridge University Press, Cambridge.

Pitteri, M., Zanzotto, G., 2002. Continuum models for phase transitions and twinning in crystals. Applied Mathematics, vol 19. Chapman & Hall, London.

Sands, D.E., 1993. Introduction to Crystallography. Dover, Mineola, NY.

Terhune, R.W., Kushida, T., Ford, G.W., 1985. Soft acoustic modes in trigonal crystals. Phys. Rev. B: Condensed Matter 32 (12), 8416–8419.

Wallace, D.C., 1998. Thermodynamics of Crystals. Dover, Mineola, NY.

Yu, Z.-Z., Clapp, P.C., 1989. Growth dynamics study of the martensitic transformation in Fe-30 Pct Ni alloys. Part II. Computer simulation of martensitic growth. Metall. Trans. A—Phys. Metall. Mat. Sci. 20A (9), 1617–1629.