

# A Theory of Anharmonic Lattice Statics for Analysis of Defective Crystals

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## Abstract

This paper develops a theory of anharmonic lattice statics for the analysis of defective complex lattices. This theory differs from the classical treatments of defects in lattice statics in that it does not rely on harmonic and homogenous force constants. Instead, it starts with an interatomic potential, possibly with infinite range as appropriate for situations with electrostatics, and calculates the equilibrium states of defects. In particular, the present theory accounts for the differences in the force constants near defects and in the bulk. The present formulation reduces the analysis of defective crystals to the solution of a system of nonlinear difference equations with appropriate boundary conditions. A harmonic problem is obtained by linearizing the nonlinear equations, and a method for obtaining analytical solutions is described in situations where one can exploit symmetry. It is then extended to the anharmonic problem using modified Newton-Raphson iteration. The method is demonstrated for model problems motivated by domain walls in ferroelectric materials.

**Keywords:** Lattice Statics, Defective Crystals, Discrete Mechanics.

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# 1 Introduction

The method of lattice statics introduced by Born and his co-workers (see Born and Huang (1988)) has been widely used to study various aspects of atomistic solids. In particular, it has been widely used to study equilibrium structure of defects. Based on a formulation by Matsubara (1952) and Kanazaki (1957), this version of lattice statics has been used for point defects by Flocken and Hardy (1969) and Flocken (1972), for interstitial by Flocken and Hardy (1968), for cracks by Hsieh and Thomson (1973) and Esterling (1978), for surfaces by Flocken (1977) and for dislocations by Maradudin (1958), Boyer and Hardy (1971), Esterling (1978), Esterling and Moriarty (1978), Shenoy et al. (1999) and Tewary (2000). Further reviews can be found in Flocken and Hardy (1970), Boyer and Hardy (1971), Tewary (1973), Bullough and Tewary (1979), Flocken and Hardy (1969), Maradudin et al. (1971), Mura (1982), King and Mura (1991), Thomson et al. (1992), Gallego and Ortiz (1993), Ortiz and Phillips (1999), Shenoy et al. (1999) and references therein. This method considers a harmonic defect-free crystal subjected to an eigendeformation chosen to represent the defect. The advantages of the method are that they provide analytic solutions and that they do not require any ad hoc cut-off or periodicity assumptions. However, they are harmonic and homogeneous, and importantly do not account for the strong nonlinear and heterogeneous behavior near the defect core. It should also be mentioned that none of the above-mentioned works solve the defect problem as a discrete boundary-value problem. In contrast, nonlinear treatments of defects are overwhelmingly computational and restrict themselves to finite domains or periodicity assumptions.

This paper is concerned with the formulation of a semi-analytical method of solution of fully nonlinear lattice statics problems for defective crystals. In particular, the method of solution takes as input an arbitrary interatomic potential, and is not restricted to interactions based on harmonic force constants. The solutions obtained represent equilibrium configurations for the input interatomic potential. The method of solution is based on a modified Newton-Raphson iteration. Each step in the iteration requires the solution of a harmonic problem with uniform force constants. The uniformity of the force constants ensures that methods of solution for difference equations, such as the discrete Fourier transform, can be applied to the linearized problem. The out-of-balance forces are computed from the full interatomic potential, thus ensuring that converged solutions represent equilibrium configurations of the anharmonic crystal. The iteration starts from a nominal configuration of the defective crystal. This initial configuration is not in equilibrium in general and the corresponding out-of-balance forces are not zero. The main purpose of the initial configuration is to place the crystal in the energy well corresponding to the defect of interest. General results then ensure that if the equilibrium defect is stable, i. e., if the corresponding force constants are coercive, and the initial nominal defect is sufficiently close to the equilibrium defect, the modified Newton-Raphson iteration converges linearly.

We demonstrate our methodology using a model problem motivated by the

study of domain walls in ferroelectric perovskites. Elsewhere we present detailed results of domain walls and defects on domain walls using more widely accepted potentials.

This paper is organized as follows. §2 reviews harmonic lattice statics. We reformulate lattice statics in a language as close to continuum mechanics as possible. In §3 the idea of symmetry reduction for defective crystals is presented and some subtleties in the linearized discrete governing equations are explained. §4 presents the idea of anharmonic lattice statics. Solution techniques for solving the linearized discrete governing equations are explained in §5. In §6 our formulation of lattice statics is generalized to a system of dipoles in which interactions are pairwise but not isotropic and atom position vectors are not the only degrees of freedom. The method of solution is illustrated by means of several examples concerned with the equilibrium structure of  $180^\circ$  and  $90^\circ$  domain walls in a two-dimensional lattice of dipoles. Conclusions are given in §7. In the appendix we show that with little modification our approach can be applied to three-body interactions.

## 2 Discrete Governing Equations

Consider a collection of atoms  $\mathcal{L}$  and assume that they interact through some interatomic potentials. Let  $\mathbf{x}^i$  denote the position of atom  $i \in \mathcal{L}$  and  $\mathcal{S}_i$  the set (list) of other atoms that it interacts with. We prohibit self interaction by assuming  $i \notin \mathcal{S}_i$ . The total energy is a function of the atomic positions,

$$\mathcal{E} = \mathcal{E}(\{\mathbf{x}^j\}_{j \in \mathcal{L}}), \quad (1)$$

and we assume that this may be written as the sum of the energy per atom

$$\mathcal{E} = \sum_{i \in \mathcal{L}} \mathcal{E}^i(\mathbf{x}^i, \{\mathbf{x}^j\}_{j \in \mathcal{S}_i}). \quad (2)$$

Note that this partitioning of energy cannot be done unambiguously in general. However, this is unambiguous in the case of pairwise interactions. Assuming that there are no discrete body forces, equilibrium requires \*

$$\frac{\partial \mathcal{E}}{\partial \mathbf{x}^i} = \mathbf{0} \quad \forall i \in \mathcal{L}. \quad (4)$$

It can be easily shown that this is equivalent to equilibrating energy of the atom  $\mathcal{E}^i$  with respect to  $\mathbf{x}^i$ , i.e.,

$$\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i}(\mathbf{x}^i, \{\mathbf{x}^j\}_{j \in \mathcal{S}_i}) = \mathbf{0}. \quad (5)$$

These equations (4) and (5) embody the main idea behind lattice or molecular statics. We seek a solution to equation (5) close to a given reference configuration  $\mathcal{B}_0 = (\mathbf{x}_0^i, \{\mathbf{x}_0^j\}_{j \in \mathcal{S}_i})$ . Therefore we expand the governing equations (5) about this reference configuration:

$$\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i} = \frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i}(\mathcal{B}_0) + \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^i \partial \mathbf{x}^i}(\mathcal{B}_0)(\mathbf{x}^i - \mathbf{x}_0^i) + \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0)(\mathbf{x}^j - \mathbf{x}_0^j) + \dots = \mathbf{0}. \quad (6)$$

We obtain the harmonic approximation by dropping the higher order terms.

It can be easily shown that because of translation invariance of the potential

$$\frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^i \partial \mathbf{x}^i}(\mathcal{B}_0) = - \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0). \quad (7)$$

This is trivially verified for pair-wise interactions and shown in the appendix to hold for three-body interactions. Using this, we rewrite the harmonic lattice

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\*When there is a discrete field of body forces this is written as

$$-\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i} \mathbf{x}^i, \{\mathbf{x}^j\}_{j \in \mathcal{S}_i} + \mathbf{F}^i = \mathbf{0}. \quad (3)$$

statics equations to be

$$\sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i} (\mathcal{B}_0) (\mathbf{x}^j - \mathbf{x}_0^j) - \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i} (\mathcal{B}_0) (\mathbf{x}^i - \mathbf{x}_0^i) = -\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i} (\mathcal{B}_0). \quad (8)$$

Setting

$$\begin{aligned} \mathbf{f}_i &= -\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i} (\mathcal{B}_0), \\ \mathbf{u}^i &= \mathbf{x}^i - \mathbf{x}_0^i, \\ \mathbf{K}_{ij} &= \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^i \partial \mathbf{x}^j} (\mathcal{B}_0), \end{aligned} \quad (9)$$

the harmonic lattice statics governing equations may be written as

$$\sum_{j \in \mathcal{S}_i} \mathbf{K}_{ij} (\mathbf{u}^j - \mathbf{u}^i) = \mathbf{f}_i \quad \forall i \in \mathcal{L}. \quad (10)$$

A couple of remarks are in order. The matrix of force constants  $\mathbf{K}_{ij}$  are derived from a potential about some reference configuration, and thus they depend on the reference configuration. In particular, they may depend explicitly on the indices  $i$  and  $j$ . In the classical formulation of harmonic lattice statics, the governing equations would be written for some periodic lattice and the force constants would depend only on the reference distance between atoms  $i$  and  $j$ .

The unbalanced force field  $\mathbf{f} = \{\mathbf{f}_i\}_{i \in \mathcal{L}}$  can be written as

$$\mathbf{f}_i = \sum_{j \in \mathcal{L}} \mathbf{K}_{ij} \Delta \mathbf{u}_e^j \quad \forall i \in \mathcal{L}. \quad (11)$$

Or

$$\mathbf{f} = \mathcal{T}(\Delta \mathbf{u}_e), \quad (12)$$

where the field of *eigen-deformations*  $\Delta \mathbf{u}_e = \{\Delta \mathbf{u}_e^i\}_{i \in \mathcal{L}}$  is formally defined as

$$\Delta \mathbf{u}_e = \mathcal{T}^{-1}(\mathbf{f}). \quad (13)$$

Thus we can rewrite Eq. (10) as

$$\sum_{j \in \mathcal{L}} \mathbf{K}_{ij} (\mathbf{u}^j - \Delta \mathbf{u}_e^j) = \mathbf{0} \quad \forall i \in \mathcal{L} \quad (14)$$

and recognize it to be exactly that as the classical equation of harmonic lattice statics (Ortiz and Phillips, 1999).

## 2.1 Linearized Discrete Governing Equations for Defective Crystals with No Symmetry Reduction

We now specialize to a (defective) complex lattice with a unit cell consisting of  $N$  atoms. Here by “defective” lattice we mean a collection of atoms that

is locally like a perfect complex lattice. We index the unit cells using integers  $(m, n, p) \in \mathbb{Z}^3$ . Let  $I\alpha\beta\gamma$  denote the  $I$ th atom in the  $(\alpha, \beta, \gamma)$  unit cell. Given an atom  $i$ , any other atom  $j$  can be specified uniquely by the unit cell it belongs to and its type, i.e.  $j = I\alpha\beta\gamma$ . The discrete harmonic governing equations (10) can now be written as

$$\sum_{\alpha, \beta, \gamma = -\infty}^{\infty} \prime \sum_{I=1}^N \mathbf{K}_{iI\alpha\beta\gamma} (\mathbf{u}_{\alpha\beta\gamma}^I - \mathbf{u}^i) = \mathbf{f}_i \quad \forall i \in \mathcal{L}, \quad (15)$$

where the prime on the summation means that the self-interaction term has been excluded.

Let us define unit cell displacement vectors as

$$\mathbf{U}_{mnp} = \begin{pmatrix} \mathbf{u}_{mnp}^1 \\ \vdots \\ \mathbf{u}_{mnp}^N \end{pmatrix} \quad (m, n, p) \in \mathbb{Z}^3. \quad (16)$$

Now the discrete governing equations can be written in terms of interaction of unit cells as

$$\sum_{\alpha, \beta, \gamma = -\infty}^{\infty} \mathbf{A}_{\alpha\beta\gamma}(m, n, p) \mathbf{U}_{m+\alpha, n+\beta, p+\gamma} = \mathbf{F}_{mnp} \quad (m, n, p) \in \mathbb{Z}^3, \quad (17)$$

where

$$\mathbf{A}_{\alpha\beta\gamma}(m, n, p) \in \mathbb{R}^{3N \times 3N}, \quad \mathbf{U}_{m+\alpha, n+\beta, p+\gamma}, \quad \mathbf{F}_{mnp} \in \mathbb{R}^{3N}. \quad (18)$$

This is a linear vector-valued partial difference equation with variable coefficient matrices of infinite order. The unit cell force vectors and the unit cell stiffness matrices are defined as

$$\mathbf{F}_{mnp} = \begin{pmatrix} \mathbf{F}_{1mnp} \\ \vdots \\ \mathbf{F}_{Nmnp} \end{pmatrix}, \quad \mathbf{A}_{\alpha\beta\gamma}(m, n, p) = \begin{pmatrix} \mathbf{K}_{11\alpha\beta\gamma} & \mathbf{K}_{12\alpha\beta\gamma} & \cdots & \mathbf{K}_{1N\alpha\beta\gamma} \\ \mathbf{K}_{21\alpha\beta\gamma} & \mathbf{K}_{22\alpha\beta\gamma} & \cdots & \mathbf{K}_{2N\alpha\beta\gamma} \\ \vdots & \vdots & \cdots & \vdots \\ \mathbf{K}_{N1\alpha\beta\gamma} & \mathbf{K}_{N2\alpha\beta\gamma} & \cdots & \mathbf{K}_{NN\alpha\beta\gamma} \end{pmatrix}. \quad (19)$$

To be able to solve such a difference equation one needs to assume a finite range of interaction and then numerically study the effect of the range of interaction. Assuming ranges of interaction  $r_1, r_2$  and  $r_3$  in  $m, n$  and  $p$  directions, respectively, we have

$$\sum_{\alpha=-r_1}^{r_1} \sum_{\beta=-r_2}^{r_2} \sum_{\gamma=-r_3}^{r_3} \mathbf{A}_{\alpha\beta\gamma}(m, n, p) \mathbf{U}_{m+\alpha, n+\beta, p+\gamma} = \mathbf{F}_{mnp} \quad (m, n, p) \in \mathbb{Z}^3, \quad (20)$$

which is a linear partial difference equation of order  $r = \max(2r_1, 2r_2, 2r_3)$ .

## 2.2 Linearized Discrete Governing Equations for Defective Crystals with 2-D Symmetry Reduction

Let us now consider a complex lattice with a defect that is extended in one dimension so that we can reduce the problem to two dimensions. In other words, we study a collection of atoms which have translation invariance in only one direction. In order to do so, we note that such a complex lattice may be written as the disjoint union of one dimensional complex lattices:

$$\mathcal{L}_2^d = \bigsqcup_{\alpha, \beta \in \mathbb{Z}} \mathcal{L}_2^d(\alpha, \beta), \quad (21)$$

where  $\mathcal{L}_2^d(\alpha, \beta)$  is a one-dimensional lattice or mathematically an equivalence class of atoms. Each one-dimensional complex lattice is a chain of unit cells. Because each unit cell is equivalent to any other unit cell in the chain, the decomposition (21) can be thought of as a partitioning of the defective complex lattice into some equivalence classes (chains). Choosing a representative from each equivalence class  $\mathcal{L}_2^d(\alpha, \beta)$ , the resulting two-dimensional lattice is called the reduced lattice and is denoted by  $\mathcal{L}^{d2}$ . Further the neighboring set  $\mathcal{S}_i$  can be partitioned as

$$\mathcal{S}_i = \bigsqcup_{\alpha, \beta \in \mathbb{Z}} \bigsqcup_{I=1}^N \mathcal{S}_{I\alpha\beta}(i), \quad (22)$$

where  $\mathcal{S}_{I\alpha\beta}(i)$  is an equivalence class of equivalent atoms which all would have the same displacement with respect to a given reference configuration. In other words,  $\mathcal{S}_{I\alpha\beta}(i)$  is the set of atoms of type  $I$  in the chain  $\mathcal{L}_2^d(\alpha, \beta)$  that interact with atom  $i$ . An example would be a lattice with broken atomic bonds on a half plane, i.e., a crack. In this example equivalence classes are sets of atoms lying on lines parallel to the crack edge (front). With this partitioning one can write

$$\sum_{j \in \mathcal{S}_i} \mathbf{K}_{ij} \mathbf{u}^j = \sum_{\alpha, \beta = -\infty}^{\infty} ' \sum_{I=1}^N \mathbf{K}_{iI\alpha\beta} \mathbf{u}_{\alpha\beta}^I, \quad (23)$$

where

$$\mathbf{K}_{iI\alpha\beta} = \sum_{j \in \mathcal{S}_{I\alpha\beta}(i)} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^{I\alpha\beta} \partial \mathbf{x}^i} (\mathcal{B}_0), \quad (24)$$

and prime on the summation means that the self-interaction term has been excluded. It is seen that in a defective lattice there is a partial symmetry and a given atom  $i$  interacts with equivalence classes and this is why each substiffness matrix is defined in terms of a lattice sum. Thus the discrete governing equations can now be written as

$$\sum_{\alpha, \beta = -\infty}^{\infty} ' \sum_{I=1}^N \mathbf{K}_{iI\alpha\beta} (\mathbf{u}_{\alpha\beta}^I - \mathbf{u}^i) = \mathbf{f}_i \quad \forall i \in \mathcal{L}. \quad (25)$$



Let us define unit cell displacement vectors as

$$\mathbf{U}_{mn} = \begin{pmatrix} \mathbf{u}_{mn}^1 \\ \vdots \\ \mathbf{u}_{mn}^N \end{pmatrix}. \quad (26)$$

Now the governing equations can be written in terms of interaction of unit cells as

$$\sum_{\alpha, \beta = -\infty}^{\infty} \mathbf{A}_{\alpha\beta}(m, n) \mathbf{U}_{m+\alpha, n+\beta} = \mathbf{F}_{mn} \quad (m, n) \in \mathbb{Z}^2, \quad (27)$$

where

$$\mathbf{A}_{\alpha\beta}(m, n) \in \mathbb{R}^{3N \times 3N}, \quad \mathbf{U}_{m+\alpha, n+\beta}, \mathbf{F}_{mn} \in \mathbb{R}^{3N}. \quad (28)$$

This is a linear vector-valued partial difference equation with variable coefficient matrices in two independent variables. The unit cell force vectors and the unit cell stiffness matrices are defined as

$$\begin{aligned} \mathbf{F}_{mn} &= \begin{pmatrix} \mathbf{F}_{1mn} \\ \vdots \\ \mathbf{F}_{Nmn} \end{pmatrix}, \\ \mathbf{A}_{\alpha\beta}(m, n) &= \begin{pmatrix} \mathbf{K}_{11\alpha\beta} & \mathbf{K}_{12\alpha\beta} & \cdots & \mathbf{K}_{1N\alpha\beta} \\ \mathbf{K}_{21\alpha\beta} & \mathbf{K}_{22\alpha\beta} & \cdots & \mathbf{K}_{2N\alpha\beta} \\ \vdots & \vdots & \cdots & \vdots \\ \mathbf{K}_{N1\alpha\beta} & \mathbf{K}_{N2\alpha\beta} & \cdots & \mathbf{K}_{NN\alpha\beta} \end{pmatrix}. \end{aligned} \quad (29)$$

### 2.3 Linearized Discrete Governing Equations for Defective Crystals with 1-D Symmetry Reduction

We now consider a collection of atoms that has translation invariance in two directions. In other words,  $\mathcal{L}$  is a collection of two-dimensional perfect lattices. Thus let us assume that  $\mathcal{L}$  can be partitioned into two-dimensional equivalence classes:

$$\mathcal{L}_1^d = \bigsqcup_{\alpha \in \mathbb{Z}} \mathcal{L}_1^d(\alpha) \quad (30)$$

or infinite sets of atoms  $\mathcal{L}_1^d(\alpha)$  that lie on some planes. Each  $\mathcal{L}_1^d(\alpha)$  is a two-dimensional periodic collection of unit cells, i.e., a perfect two-dimensional complex lattice. Choosing a representative from each equivalence class, the resulting chain is called the reduced lattice  $\mathcal{L}^{d_1}$ . The neighboring set  $\mathcal{S}_i$  can be partitioned as

$$\mathcal{S}_i = \bigsqcup_{\alpha \in \mathbb{Z}} \bigsqcup_{I=1}^N \mathcal{S}_{I\alpha}(i), \quad (31)$$

where  $\mathcal{S}_{I\alpha}(i)$  is the equivalence class of all the atoms of type  $I$  and index  $\alpha$  with respect to atom  $i$ . In other words,  $\mathcal{S}_{I\alpha}(i)$  is the set of all atoms of type  $I$  in the

two dimensional lattice  $\mathcal{L}_1^d(\alpha)$  that interact with atoms  $i$ . For a domain wall, for example, each equivalence class is a set of atoms lying on a plane parallel to the domain wall.

With this partitioning one can write the linearized discrete governing equations as

$$\sum_{\alpha=-\infty}^{\infty} \prime \sum_{I=1}^N \mathbf{K}_{iI\alpha} \mathbf{u}_\alpha^I + \left( - \sum_{\alpha=-\infty}^{\infty} \prime \sum_{I=1}^N \mathbf{K}_{iI\alpha} \right) \mathbf{u}^i = \mathbf{f}_i, \quad (32)$$

where the prime on the first sum means that the term  $\alpha = 0, I = i$  is excluded to avoid self-interaction and

$$\begin{aligned} \mathbf{K}_{iI\alpha} &= \sum_{j \in \mathcal{S}_{I\alpha(i)}} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0), \\ \mathbf{f}_i &= - \frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i}(\mathcal{B}_0), \\ \mathbf{u}_\alpha^I &= \mathbf{x}^{I\alpha} - \mathbf{x}_0^{I\alpha} = \mathbf{x}^j - \mathbf{x}_0^j \quad \forall j \in \mathcal{S}_{I\alpha(i)}. \end{aligned} \quad (33)$$

Let us define unit cell displacement vectors as

$$\mathbf{U}_m = \begin{pmatrix} \mathbf{u}_m^1 \\ \vdots \\ \mathbf{u}_m^N \end{pmatrix}. \quad (34)$$

Now the governing equations can be written in terms of interaction of unit cells as

$$\sum_{\alpha=-\infty}^{\infty} \mathbf{A}_\alpha(m) \mathbf{U}_{m+\alpha} = \mathbf{F}_m \quad m \in \mathbb{Z}, \quad (35)$$

where

$$\mathbf{A}_\alpha(m) \in \mathbb{R}^{3N \times 3N}, \quad \mathbf{U}_\alpha, \mathbf{F}_m \in \mathbb{R}^{3N}. \quad (36)$$

This is a linear vector-valued ordinary difference equation with variable coefficient matrices. The unit cell force vectors and the unit cell stiffness matrices are defined as

$$\mathbf{F}_m = \begin{pmatrix} \mathbf{F}_{1m} \\ \vdots \\ \mathbf{F}_{Nm} \end{pmatrix}, \quad \mathbf{A}_\alpha(m) = \begin{pmatrix} \mathbf{K}_{11\alpha} & \mathbf{K}_{12\alpha} & \cdots & \mathbf{K}_{1N\alpha} \\ \mathbf{K}_{21\alpha} & \mathbf{K}_{22\alpha} & \cdots & \mathbf{K}_{2N\alpha} \\ \vdots & \vdots & \cdots & \vdots \\ \mathbf{K}_{N1\alpha} & \mathbf{K}_{N2\alpha} & \cdots & \mathbf{K}_{NN\alpha} \end{pmatrix}. \quad (37)$$

Note that, in general,  $\mathbf{A}_\alpha(m)$  need not be symmetric as will be explained shortly. The above system of difference equations is a Volterra system of difference equations (see Elaydi (1996))<sup>†</sup>.

<sup>†</sup>Lattice statics analysis of defective crystals with 1-D symmetry reduction leads to the solution of vector-valued ordinary difference equations with variable coefficient matrices. Inhomogeneities are localized and the idea is to treat the inhomogeneous region as boundary and transition regions. This will result in two vector-valued difference equations with constant coefficient matrices one forward and one backward. In the end, the original difference equation will be solved by matching the solutions of these two ordinary difference equations.

The above governing equations can be written in terms of a discrete convolution operator as<sup>‡</sup>

$$\mathbf{A}\mathbf{X} = \mathbf{F}, \quad (38)$$

where  $\mathbf{X} = \{\mathbf{X}_n\}$ ,  $\mathbf{F} = \{\mathbf{F}_n\}$  and the discrete convolution operator is defined as

$$\mathbf{A}\mathbf{X} = \{(\mathbf{A}\mathbf{X})_n\}, \quad (39)$$

and

$$(\mathbf{A}\mathbf{X})_n = \sum_{m=-\infty}^{\infty} A_{n-m} \mathbf{X}_m. \quad (40)$$

## 2.4 Some remarks

For the case of  $N \geq 2$ , there are some subtleties in calculating the  $\mathcal{A}_\alpha$  matrices. This is also the case for defective crystals with 2-D and no symmetry reductions but for the sake of simplicity we explain this subtlety only for defective crystals with a 1-D symmetry reduction. One subtlety is that some interactions should be ignored. One is the interaction of an atom of type I and index n with all atoms of type I and index n, i.e., there are no interactions within a given equivalence class (this is a consequence of Eq. (7)). This means that  $\mathcal{A}_0$  has a special structure. When position of atom  $i$  of type I changes, all its equivalent atoms, i.e., those with  $\alpha = 0$  undergo the same perturbation. Atoms of the same type as  $i$  do not contribute to energy of  $i$  because the potential is pairwise and their relative distances from the atom  $i$  are always the same. This means that

$$\mathbf{K}_{II0} = - \sum_{\alpha=-\infty}^{\infty} ' \sum_{\substack{J=1 \\ J \neq I}}^N \mathbf{K}_{IJ\alpha}. \quad (41)$$

The same thing is true for forcing terms. The reason for this is that the distance between the equivalent atoms is fixed and atoms in the equivalence class of  $i$  do not contribute to  $-\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i}$  and its derivatives. For a defective crystal with a 2-D symmetry reduction the above property implies that

$$\mathbf{K}_{II00} = - \sum_{\alpha,\beta=-\infty}^{\infty} ' \sum_{\substack{J=1 \\ J \neq I}}^N \mathbf{K}_{IJ\alpha\beta}. \quad (42)$$

The other subtlety is when a finite number of interactions is considered for representative unit cells. Consider atoms with index n and project the whole defective crystal on a line perpendicular to the two-dimensional defect. This would be the reduced lattice  $\mathcal{L}^{d1}$ . As an example, we have the picture shown in Fig. 1 for A and O2 atoms in a perovskite multilattice  $\text{ABO}_3$ . Suppose

<sup>‡</sup>This is the approach that Babuška (1959) chooses in his treatment of difference equations. We do not use this notation in this paper but it would be useful to know that the discrete governing equations have a discrete convolution form.

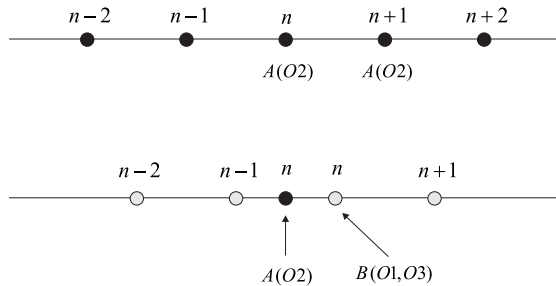


Figure 1: Nearest neighbors of A and O2 atoms and their indices in an  $ABO_3$  defective crystal with a 1-D symmetry reduction.

a given representative unit cell interacts with its first  $m$ th nearest neighbor (representative) unit cells. We consider the interaction of A and O2 atoms with other A and O2 atoms of indices  $\{n - m, \dots, n + m\}$  (except the ones that have already been excluded). Looking at Fig. 1, one can see that symmetry of interactions dictates that interactions of A and O2 atoms with O1, O2 and O3 atoms with index  $n + m$  should be ignored. Similarly, consider atoms B, O2 or O3 with index  $n$  and their nearest neighbors as shown in Fig. 2. Every atom B (O1 or O3) interacts with B, O1 and O3 atoms with index  $\{n - m, \dots, n + m\}$  (except the ones that have already been excluded). Again, symmetry implies that the interactions of B, O1 and O3 atoms with A and O2 atoms with index  $n - m$  should be ignored.

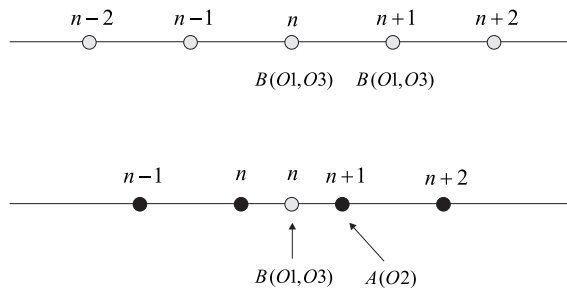


Figure 2: Nearest neighbors of B, O1 and O3 atoms and their indices in an  $ABO_3$  defective crystal with a 1-D symmetry reduction.

Another interesting subtlety is the symmetry of  $\mathcal{A}_\alpha$  matrices. It should be noted that each  $\mathbf{K}_{iI\alpha}$  is symmetric but the matrices  $\mathcal{A}_\alpha$  ( $\alpha = -m, \dots, m$ ) are not symmetric, in general. This can be seen more clearly in a simple 2-D model. Consider a 2-D rectangular multi-lattice composed of two simple lattices each

with lattice parameters  $a$  and  $c$  and the shift vector  $\mathbf{p} = (p_1, p_2)$ . This system has three coefficient matrices  $\mathcal{A}_1, \mathcal{A}_0, \mathcal{A}_1 \in \mathbb{R}^{4 \times 4}$ . We now compare  $\mathbf{K}_{12-1}$  and  $\mathbf{K}_{21-1}$  to see if  $\mathcal{A}_1$  is symmetric. It can be easily shown that

$$\mathbf{K}_{12-1} = \sum_{Y\{n-1\}} \frac{\partial^2 E}{\partial \mathbf{x}_{n-1} \partial \mathbf{y}_{n-1}} (\mathcal{B}_0), \quad (43)$$

$$\mathbf{K}_{21-1} = \sum_{X\{n-1\}} \frac{\partial^2 E}{\partial \mathbf{y}_{n-1} \partial \mathbf{x}_{n-1}} (\mathcal{B}_0), \quad (44)$$

where  $X\{n-1\}$  is the set of atoms of type 1 which have index  $n-1$  relative to the atom  $n$  of type 2 (these are the circles in Fig. 3). Similarly,  $Y\{n-1\}$  is the set of atoms of type 2 which have index  $n-1$  relative to the atom  $n$  of type 1 (these are the squares in Fig. 3).  $\mathbf{x}_{n-1}$  and  $\mathbf{y}_{n-1}$  are position vectors of atoms of types 1 and 2 with index  $n-1$ , respectively.

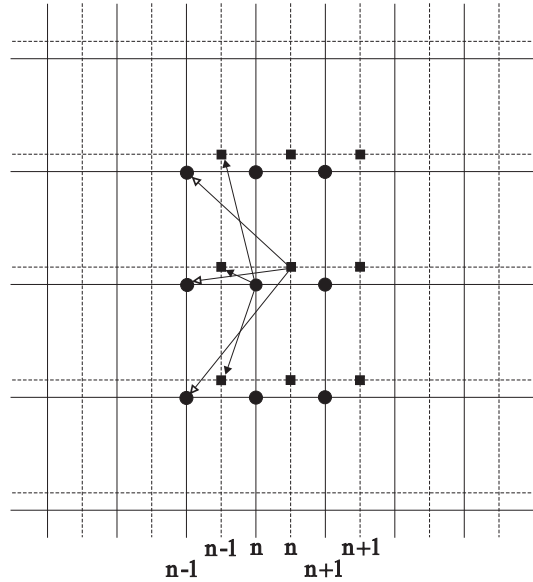


Figure 3: Non-symmetry of  $\mathcal{A}_i$  matrices.

As it is seen in Fig. 3, these two matrices are not equal as the length of the corresponding relative position vectors are not equal. It should be noted that the lose of symmetry in the reduced 1-D system is just a consequence of symmetry reduction and still the underlying 3-D physical system is symmetric.

Suppose all the atoms in the lattice have the same displacements, i.e.,

$$\mathbf{X}_n = \mathbf{C} = (\underbrace{\mathbf{c}, \dots, \mathbf{c}}_{N \text{ copies}})^\top, \quad \mathbf{c} \in \mathbb{R}^3. \quad (45)$$

Using Eq. (41), it can be easily shown that

$$\sum_{\alpha=-\infty}^{\infty} \mathcal{A}_\alpha \mathbf{C} = \mathbf{0} \quad \forall \mathbf{c} \in \mathbb{R}^3, \quad (46)$$

i.e.,

$$\mathcal{N} \left( \sum_{\alpha=-\infty}^{\infty} \mathcal{A}_\alpha \right) = 3 \quad (47)$$

as was expected, where  $\mathcal{N}(\mathbf{A})$  is the nullity of the linear transformation represented by the matrix  $\mathbf{A}$ . Note that the above statement is formal because there is an infinite sum and in general one should worry about convergence. This is not an issue for short-range interactions but long-range interactions should be treated carefully. We will come back to the convergence issue in the sequel. For the case of a defective crystal with a 2-D symmetry reduction the above property states that

$$\mathcal{N} \left( \sum_{\alpha, \beta=-\infty}^{\infty} \mathcal{A}_{\alpha\beta} \right) = 3. \quad (48)$$

There is another symmetry relating  $\mathcal{A}_{-\gamma}$  to  $\mathcal{A}_\gamma$ . It can be easily shown that reciprocity implies that

$$\mathbf{K}_{IJ-\gamma} = \mathbf{K}_{JI\gamma}. \quad (49)$$

This means that

$$\mathcal{A}_{-\gamma} = \mathcal{A}_\gamma^\top. \quad (50)$$

Convergence of infinite sums raise their own delicate issues. In the analysis of defective crystals with 1-D and 2-D symmetry reductions, we need to calculate stiffness matrices that are defined in terms of lattice sums of square matrices. Discrete field of unbalanced forces is also defined in terms of lattice sums. Since we will be interested in dipole-dipole interactions, we will find that substiffness matrices for defective crystals with 1-D and 2-D symmetry reductions are absolutely convergent. However, the forces are obtained as conditionally convergent sums and thus require care. In our examples, we look at systems of dipoles lying on a plane and thus force is also defined in terms of absolutely convergent lattice sums.

Finally, our lattice statics model forces are always calculated exactly. However, to be able to solve the governing discrete equations for an infinite lattice we need to have a system of difference equations of finite order. It would be interesting to know how sensitive the solutions are to the range of interaction of representative unit cells. This is problem dependent and should be carefully studied for a given interatomic potential.

In all the existing lattice statics calculations a fixed number of nearest neighbor interactions (usually only the first and second nearest-neighbor interactions) are considered. Our formulation of lattice statics can consider any number of nearest-neighbor interactions and this enables us to numerically study the effect of range of interactions with no difficulty. In §6 we present a numerical study of the effect of range of interaction for a lattice of dipoles. It will be seen that the effective potential is highly localized and increasing the range of interaction does not change the displacements, i.e., the displacements are independent of the range of interaction.

Our formulation of anharmonic lattice statics starts with choosing a reference configuration. Here a comment is in order regarding the choice of reference configuration. For a given defect, reference configuration is chosen to be a nominal defect. By ‘nominal’ defect, we mean a configuration that is locally like the bulk crystal but in some region(s) is close to the relaxed configuration of the defect. Of course, a nominal defect is not unique. An example is shown in Fig. 4 for an edge dislocation. In this figure we show the reduced lattice, i.e., representative atoms of lines of atoms perpendicular to the plane. We know that a dislocation can be understood as an extra half plane of atoms inserted in the bulk lattice (in the reduced lattice a half line of extra atoms). Fig. 4a shows a nominal defect that has been obtained by inserting a half plane of extra atoms between two crystallographic planes. In Fig. 4b the configuration (a) has been modified to make it exactly like the bulk crystal except in the region bounded by the broken lines. These two nominal defect reference configurations are both acceptable choices but configuration (b) is preferable because its unbalanced force field is localized and this makes the numerical calculations more efficient.

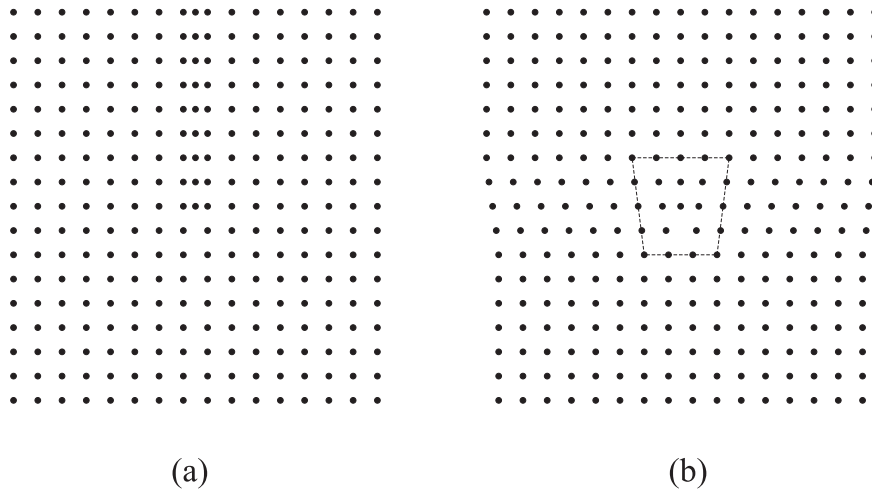


Figure 4: (Two possible reference configurations for a dislocation.

In the previous lattice statics models of dislocations (Maradudin, 1958; Boyer and Hardy, 1971; Gallego and Ortiz, 1993) always some cutting and pasting process is used. In the present formulation all is needed is a reference configuration. The unit cell numbering for an edge dislocation is shown in Fig. 5. Note that the  $n$ -axis is ‘curved’ but still the governing linearized equations are

$$\sum_{\alpha,\beta} \mathcal{A}_{\alpha\beta}(m, n) \mathbf{X}_{m+\alpha, n+\beta} = \mathbf{F}_{mn} \quad (m, n) \in \mathbb{Z}^2. \quad (51)$$

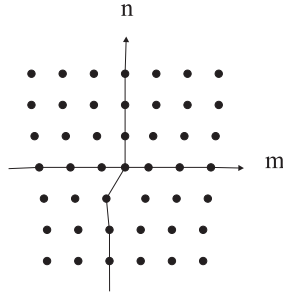


Figure 5: Unit cell numbering for the reference configuration of an edge dislocation.

We close this section by stating our harmonic lattice statics algorithm:

- Input data: defective crystal geometry, interatomic potential
- ▷ Initialization
    - ▷ Construct  $\mathcal{B}_0$ , calculate force and substiffness matrix moduli
    - ▷ Do for all  $\alpha \in \mathbb{K}^s$ 
      - ▷ Assemble substiffness matrices and construct  $\mathcal{A}_\alpha$
    - ▷ End Do
  - ▷ Calculate unbalanced forces  $\mathbf{F}^0 = \mathbf{F}(\mathcal{B}_0)$
  - ▷ Solve the governing linear difference equations
  - ▷ End



### 3 Anharmonic Lattice Statics

The classical harmonic lattice statics is not appropriate where displacements from the initial configuration are large. There have been modifications of the harmonic lattice statics in the past (Esterling, 1978; Esterling and Moriarty, 1978; Gallego and Ortiz, 1993). The idea of these and similar works is to consider the fully nonlinear equations close to defects. These works, however, do not solve a nonlinear defect problem as a discrete boundary-value problem; instead all these and similar works are more or less heuristic. In this section we present a formulation of anharmonic lattice statics in which one solves a nonlinear discrete defect problem by solving discrete linear boundary-value problems. Anharmonic lattice statics is based on Newton-Raphson (NR) method for solving nonlinear equations. The basic idea of NR method is to look at a quadratic approximation to the nonlinear equations in each step. Suppose  $\mathbf{f} : \mathbb{R}^n \rightarrow \mathbb{R}^n$  is continuously differentiable and that  $\mathbf{f}(\mathbf{x}^*) = \mathbf{0}$  for some  $\mathbf{x}^* \in \mathcal{D} \subset \mathbb{R}^n$ . We know that derivative of  $\mathbf{f}$  is a linear map defined as

$$\mathbf{f}(\mathbf{x} + \mathbf{u}) = \mathbf{f}(\mathbf{x}) + D\mathbf{f}(\mathbf{x})\mathbf{u} + o(\|\mathbf{u}\|). \quad (52)$$

Let us start from an initial guess  $\mathbf{x}_0 \in \mathcal{D}$ . The linear approximation of  $\mathbf{f}$  about  $\mathbf{x}_0$  calculated at a point  $\mathbf{x}_1 \in \mathcal{D}$  is

$$\mathbf{f}(\mathbf{x}_1) \approx \mathbf{f}(\mathbf{x}_0) + D\mathbf{f}(\mathbf{x}_0)(\mathbf{x}_1 - \mathbf{x}_0). \quad (53)$$

Assuming that  $\mathbf{f}(\mathbf{x}_1) \approx \mathbf{0}$  we have

$$\mathbf{x}_1 = \mathbf{x}_0 - D\mathbf{f}(\mathbf{x}_0)^{-1}\mathbf{f}(\mathbf{x}_0). \quad (54)$$

Similarly, in the  $k$ th step

$$\mathbf{x}_{k+1} = \mathbf{x}_k - D\mathbf{f}(\mathbf{x}_k)^{-1}\mathbf{f}(\mathbf{x}_k). \quad (55)$$

It can be shown that this algorithm has a quadratic convergence (see Dennis and Schnabel (1996)), i.e.,

$$\|\mathbf{x}_{k+1} - \mathbf{x}^*\| \leq C\|\mathbf{x}_k - \mathbf{x}^*\|^2 \quad \text{for some positive number } C. \quad (56)$$

The modified NR method is based on a similar idea. In the  $k$ th iteration one defines

$$\mathbf{x}_{k+1} = \mathbf{x}_k - D\mathbf{f}(\mathbf{x}_0)^{-1}\mathbf{f}(\mathbf{x}_k), \quad (57)$$

i.e., the only difference is that in all the steps the derivative of the initial guess is used. This is however slower than the usual NR iteration.

By modifying the proof presented in (Dennis and Schnabel, 1996), it can be shown that the convergence of modified NR method is linear, i.e.,

$$\|\mathbf{x}_{k+1} - \mathbf{x}^*\| \leq c\|\mathbf{x}_k - \mathbf{x}^*\| \quad \text{for some } c \in (0, 1). \quad (58)$$

The idea of anharmonic lattice statics is to find the nonlinear solutions by a modified Newton-Raphson iteration. In modified Newton-Raphson method

the Hessian matrix is not updated in each iteration and the initial Hessian is used. Modified Newton-Raphson method is slowly and linearly convergent and a large number of iterations should be performed to get good results. In our lattice statics calculations this is an efficient method as the most expensive part of the calculations is the computation of substiffness matrices (very slowly converging lattice sums). It is important to note that the Hessian at  $\mathbf{x} = \mathbf{x}_0$  should be positive-definite for the modified NR to converge to a local minimum of the energy.

Here we explain the idea for all three types of defective crystals. Let  $\alpha \in \mathbb{Z}^3, \mathbb{Z}^2, \mathbb{Z}$  for defective crystals with no symmetry reduction, with a 2-D symmetry reduction and with a 1-D symmetry reduction, respectively. The linearized governing equations have the following form

$$\sum_{\alpha \in \mathbb{Z}^s} \mathbf{A}_\alpha \mathbf{U}_{n+\alpha} = \mathbf{F}_n \quad n \in \mathbb{Z}^s \quad (s = 1, 2, \text{ or } 3). \quad (59)$$

Note that in general  $\mathbf{A}_\alpha = \mathbf{A}_\alpha(n)$  and are evaluated with respect to a given reference configuration  $\mathcal{B}_0$ . Given a reference configuration  $\mathcal{B}_0$ , we calculate the discrete field of unbalanced forces exactly. Let us denote this by  $\mathbf{F}^0 = \{\mathbf{F}_n^0\}_{n \in \mathbb{Z}^s}$ . Note that  $\mathbf{F}^0 : \mathcal{L}^{ds} \rightarrow \mathbb{R}^3$ , where  $\mathcal{L}^{ds}$  is the reduced defective lattice.\* Having  $\mathbf{F}^0$ , one has the following discrete boundary value problem (DBVP)

$$\sum_{\alpha \in \mathbb{Z}^s} \mathbf{A}_\alpha \mathbf{U}_{n+\alpha}^0 = \mathbf{F}_n^0 \quad n \in \mathbb{Z}^s$$

Boundary Conditions (B.C.) (60)

The boundary conditions are problem dependent. For infinite defective crystals we require boundedness of displacements at infinity. Solving the above DBVP one obtains  $\mathbf{U}^0 = \{U_n^0\}_{n \in \mathbb{Z}^s}$ . Now the reference configuration is updated as follows.

$$\mathcal{B}_0^1 = \mathcal{B}_0 + \mathbf{U}^0. \quad (61)$$

In the case of a system with  $\mathcal{B}_0 = \{\mathbf{X}_n^0\}_{n \in \mathbb{Z}^s}$ , i.e., when the only degrees of freedom are position vectors of the lattice points, this means that

$$\{\mathbf{X}_n^1\}_{n \in \mathbb{Z}^s} = \{\mathbf{X}_n^0 + \mathbf{U}_n^0\}_{n \in \mathbb{Z}^s}. \quad (62)$$

Now having a new reference configuration one can calculate the discrete field of unbalanced forces  $\mathbf{F}^1 = \{\mathbf{F}_n^1\}_{n \in \mathbb{Z}^s}$ . In the second step one has the following DBVP

$$\sum_{\alpha \in \mathbb{Z}^s} \mathbf{A}_\alpha \mathbf{U}_{n+\alpha}^1 = \mathbf{F}_n^1 \quad n \in \mathbb{Z}^s$$

B.C. (63)

Note that the stiffness matrices are not updated and in all the steps the original stiffness matrices are used. Solving the above DBVP one obtains  $\mathbf{U}^1 =$

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\*In the case of a defective crystal with no symmetry reduction  $\mathcal{L}^{d3} = \mathcal{L}$ .

$\{U_n^1\}_{n \in \mathbb{Z}^s}$  and  $\mathcal{B}_0^2 = \mathcal{B}_0^1 + \mathbf{U}^1$ . This process at step  $k$  requires solving the following BVP

$$\sum_{\alpha \in \mathbb{Z}^s} \mathbf{A}_\alpha \mathbf{U}_{n+\alpha}^{k-1} = \mathbf{F}_n^{k-1} \quad n \in \mathbb{Z}^s$$

*B.C.*

(64)

where

$$\mathbf{F}^{k-1} = \mathbf{F}(\mathcal{B}_0^{k-1}) \quad \text{and} \quad \mathcal{B}_0^{k-1} = \mathcal{B}_0^{k-2} + \mathbf{U}^{k-2}. \quad (65)$$

Depending on the problem the fields  $\mathbf{U}^k$  are localized or localized modulo some rigid translation fields. This means that the fields  $\mathbf{F}^k$  are localized. This is problem dependent and one should carefully study the rate of decay of unbalanced forces for a given defective crystal. The following is our anharmonic lattice statics algorithm:

Input data:  $\mathcal{B}_0, \mathcal{A}_\alpha, \mathbf{U}^0$

- ▷ Initialization
  - ▷  $\mathcal{B}_0^1 = \mathcal{B}_0 + \mathbf{U}^0$
- ▷ Do until convergence is achieved
  - ▷  $\mathbf{F}^k = \mathbf{F}(\mathcal{B}_0^k)$
  - ▷ Calculate  $\mathbf{U}^k$  by solving the harmonic problem
  - ▷  $\mathcal{B}_0^{k+1} = \mathcal{B}_0^k + \mathbf{U}^k$
- ▷ End Do
- ▷ End

## 4 Solution Methods for the Linearized Discrete Governing Equations

In this section we present analytic methods for solving the discrete governing equations for defective crystals with 1-D, 2-D and no symmetry reductions. In anharmonic lattice statics the first step in solving a nonlinear problem is to solve the linearized governing equations. Linearized governing equations with respect to a given reference configuration are vector-valued partial difference equations. In this section we briefly review the theory of ordinary and partial difference equations.

### 4.1 Theory of Difference Equations

Difference equations arise in many problems of mathematical physics. They also appear in discretization of boundary value problems and also in combinatorics. In this subsection we review a few facts and theorems from theory of difference equations. For more details see Agarwal (2000); Elaydi (1996); Lakshmikantham and Trigiante (1988) and references therein.

#### 4.1.1 Ordinary Difference Equations

An ordinary difference equation is the discrete analogue of an ordinary differential equation. Difference equations can be defined on bounded or unbounded discrete domains. For us all difference equations are defined on unbounded domains. Consider a sequence  $\{u_n\}_{n \in \mathbb{N}} \subset \mathbb{R}$ . A difference equation in the independent variable  $n$  is an equation of the form

$$f(n, u_n, \dots, u_{n+p}) = 0. \quad (66)$$

The order of a difference equation is the difference between the largest and smallest arguments explicitly involved in the equation. A linear (scalar-valued) difference equation has the following form

$$\sum_{j=0}^p K_j(n) u_{n+j} = b_n \quad n \in \mathbb{N}. \quad (67)$$

Here, we are interested in linear difference equations with constant coefficients. These equations show up in discrete systems with uniform physical properties. Consider a  $p$ th order difference equation with constant coefficients

$$u_{n+p} + a_1 u_{n+p-1} + a_2 u_{n+p-2} + \dots + a_p u_n = b_n. \quad (68)$$

Similar to differential equations, we solve the corresponding homogeneous equation. Assuming that solutions are of the form  $\lambda^n$ ,  $\lambda \in \mathbb{C}$ , one obtains

$$\lambda^p + a_1 \lambda^{p-1} + \dots + a_p = 0. \quad (69)$$

This is the characteristic polynomial of the difference equation (68). There are several possibilities for characteristic roots. If all the roots are real and distinct, for example, the general solution is of the form

$$u_n^c = c_1 \lambda_1^n + c_2 \lambda_2^n + \dots + c_p \lambda_p^n. \quad (70)$$

For details on other possibilities see Elaydi (1996). The general solution of Eq. (68) can be written as

$$u_n = u_n^c + u_n^p, \quad (71)$$

where  $u_n^p$  is a particular solution of the nonhomogeneous equation.

A system of linear difference equations of first order has the following form<sup>||</sup>

$$\mathbf{u}_{n+1} = \mathcal{A}(n)\mathbf{u}_n + \mathbf{b}_n, \quad \mathbf{u}_n, \mathbf{b}_n \in \mathbb{R}^p, \quad \mathcal{A}(n) \in \mathbb{R}^{p \times p}, \quad n \in \mathbb{N} \cup \{0\}. \quad (73)$$

If  $\mathcal{A}$  does not depend on  $n$  the system (73) is called a system with constant coefficients.

$$\mathbf{u}_{n+1} = \mathcal{A}\mathbf{u}_n + \mathbf{b}_n. \quad (74)$$

For the homogeneous system with constant coefficients corresponding to (74), i.e.,

$$\mathbf{u}_{n+1} = \mathcal{A}\mathbf{u}_n \quad (75)$$

the general solution is

$$\mathbf{u}_n = \mathcal{A}^n \mathbf{c}, \quad \mathbf{c} \in \mathbb{R}^p, \quad \forall n \in \mathbb{N}. \quad (76)$$

Here,  $\mathcal{A}^n$  is called the fundamental matrix of the system (74). This is the analogue of  $e^{\mathcal{A}t}$  in a linear system of differential equations. System of difference equations (74) has  $p$  linearly independent solutions and the general solution can be written as

$$\mathbf{u}_n = \mathcal{A}^n \mathbf{c} + \mathbf{u}_n^p, \quad (77)$$

where  $\mathbf{u}_n^p$  is a particular solution. Using the method of variation of constants the general solution can be expressed as

$$\mathbf{u}_n = \mathcal{A}^n \mathbf{c} + \sum_{j=0}^{n-1} \mathcal{A}^{n-j-1} \mathbf{b}_j. \quad (78)$$

Note that a system of difference equations can be thought of as a vector-valued ordinary difference equation.

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<sup>||</sup>It should be noted that this is not the most general form of a linear vector-valued difference equation. The most general first-order linear vector-valued difference equation has the following form

$$\mathcal{A}_1(n)\mathbf{u}_{n+1} + \mathcal{A}_2(n)\mathbf{u}_n = \mathbf{b}_n, \quad \mathbf{u}_n, \mathbf{b}_n \in \mathbb{R}^p, \quad \mathcal{A}_1(n), \mathcal{A}_2(n) \in \mathbb{R}^{p \times p}, \quad n \in \mathbb{N} \cup \{0\}. \quad (72)$$

The matrix  $\mathcal{A}_1$  can be singular in general. A direct solution of this equation in the case of constant coefficient matrices can be found in Yavari (2004).

### 4.1.2 Partial Difference Equations

Partial difference equations are discrete analogues of partial differential equations. Let  $\mathbb{Z}^p$  be the set of all p-tuples of integers ( $p \geq 2$ ). A linear partial difference equation has the following form

$$L\mathbf{X}_\alpha = \sum_{\beta \in \Omega} \mathbf{A}(\beta)\mathbf{X}_{\alpha+\beta} = \mathbf{F}_\alpha, \quad (79)$$

where  $\Omega \subset \mathbb{Z}^p$ ,  $\alpha, \beta \in \mathbb{Z}^p$  and

$$\mathbf{X}, \mathbf{F} : \Omega \rightarrow \mathbb{R}^q, \quad \mathbf{A} : \Omega \rightarrow \mathbb{R}^q \times \mathbb{R}^q. \quad (80)$$

For  $p = 2$ , a linear partial difference equation has the following form

$$\sum_{(r,s) \in \mathbb{Z}^2} \mathbf{A}_{rs}\mathbf{X}_{m+r,n+s} = \mathbf{F}_{mn} \quad (m, n) \in \Omega. \quad (81)$$

It is known that (Gregor, 1998) solution space of a partial difference equation is, in general, infinite dimensional. This means that explicit solutions of partial difference equations cannot be as simple as those of ordinary difference equations. The most common techniques for solving linear partial difference equations are integral transforms. For solving partial difference equations on bounded rectangular domains there are direct methods using matrix tensor product methods (Lynch et al., 1964). However, these methods are not applicable to the problems we have in mind for defective crystals. There are also some direct methods for solving simple partial difference equations (see Mickens (1990)). However, these methods are not applicable for general vector-valued partial difference equations.

### 4.1.3 Discrete Fourier Transform

Discrete Fourier Transform (DFT) is a powerful technique for solving systems of linear difference equations. In the literature there are two different types of discrete Fourier transform both known as DFT. The first type, which is the one we use in this paper, transforms a sequence (or more precisely a lattice function) to a function of a continuous variable(s). This is sometimes called *continuous discrete Fourier transform* (CDFT). Theory of CDFT was developed in (Babůška, 1959; Babůška et al., 1960; Vitásek, 1959). The other type of DFT, which we call *discrete DFT* (DDFT), transforms a sequence to another sequence (Briggs and Hendon, 1995), (Benedetto, 1997) and is usually useful for solving periodic difference equations or difference equations on bounded domains. We will briefly review DDFT and its applications in solving difference equations with periodic boundary conditions at the end of this section. In this work by DFT we mean CDFT, i.e., the one that maps a lattice function to a continuous function in k-space.

Consider a lattice  $\mathcal{L}$  and a lattice function  $f : \mathcal{L} \rightarrow \mathbb{R}^3$ . The discrete Fourier transform of  $f$  is defined formally as

$$\hat{f}(\mathbf{k}) = V \sum_{j \in \mathcal{L}} f(j) e^{i\mathbf{k} \cdot \mathbf{x}^j} \quad \mathbf{k} \in \mathbf{B}, \quad (82)$$

where  $V$  is the volume of the unit cell and  $\mathbf{B}$  is the first Brillouin zone. For a chain of atoms of unit lattice spacing this definition reduces to the usual definition of DFT of a sequence in  $\mathbb{R}$ , where  $V = 1$ ,  $\mathbf{B} = [-\pi, \pi]$ . Let us denote by  $\mathcal{U}$  the set of all discrete Fourier transformable lattice functions. Let us also denote by  $\mathcal{R}$  the set of those lattice functions such that

$$|f(\mathbf{x})| \leq C \prod_{i=1}^3 (1 + |x_i|^p) \quad \forall \mathbf{x} = (x_1, x_2, x_3) \in \mathcal{L}, \quad (83)$$

for some integer  $p \geq 0$  and constant  $C \geq 0$ . It can be shown (Vitásek, 1959) that there is a one-to-one correspondence between the spaces  $\mathcal{R}$  and  $\mathcal{U}$ . It should be noted that in the definition of DFT the convergence should be understood in the sense of distributions.

Inverse DFT is defined as

$$f(j) = \frac{1}{(2\pi)^3} \int_{\mathbf{B}} \hat{f}(\mathbf{k}) e^{-i\mathbf{k} \cdot \mathbf{x}^j} d^3k. \quad (84)$$

DFT has many nice properties and here we mention a few of them. DFT is a linear operator, i.e.,

$$(\alpha f + \beta g)^\wedge = \alpha \hat{f} + \beta \hat{g} \quad \forall \alpha, \beta \in \mathbb{R}, \forall f, g \in \mathcal{U}. \quad (85)$$

Shifting property of DFT is essential in solving difference equations. Suppose

$$\widehat{\mathbf{X}}_{\mathbf{n}} = \mathbf{Y}(\mathbf{k}). \quad (86)$$

Then

$$\widehat{\mathbf{X}}_{\mathbf{n}+\mathbf{m}} = e^{-i\mathbf{m} \cdot \mathbf{k}} \mathbf{Y}(\mathbf{k}). \quad (87)$$

Discrete convolution of two lattice functions  $f$  and  $g$  is defined as

$$(f * g)(i) = V \sum_{j \in \mathcal{L}} f(i - j)g(j). \quad (88)$$

Note that the multiplication  $f(i - j)g(j)$  is defined componentwise. If  $f, g \in \mathcal{U}$ , then

$$\widehat{(f * g)}(\mathbf{k}) = \hat{f}(\mathbf{k})\hat{g}(\mathbf{k}). \quad (89)$$

Discrete Fourier Transform is a powerful tool in solving partial difference equations but should be used carefully in numerical calculations as the integrands in inverse DFT may be extremely oscillatory.

#### 4.1.4 DFT and Difference Equations

Consider the following ordinary difference equation.

$$x_{p+1} - 2x_p + x_{p-1} = f_p \quad p \in \mathbb{Z}. \quad (90)$$

Note that this difference equation is translation invariant, i.e., if the sequence  $\{x_p\}_{p \in \mathbb{Z}}$  is a solution so is the sequence  $\{x_p + c\}_{p \in \mathbb{Z}}$ ,  $\forall c \in \mathbb{R}$ . Applying DFT to this difference equation we obtain

$$(e^{-ik} - 2 + e^{ik}) \widehat{x}_p(k) = \widehat{f}_p(k). \quad (91)$$

Or

$$\widehat{x}_p(k) = \frac{1}{2(\cos k - 1)} \widehat{f}_p(k). \quad (92)$$

Thus formally

$$x_p = \frac{1}{2\pi} \int_{-\pi}^{\pi} e^{-ipk} \frac{1}{2(\cos k - 1)} \widehat{f}_p(k) dk. \quad (93)$$

Note that this integral is not convergent in general because there is a singularity at  $k = 0$ , i.e.,

$$\frac{1}{2(\cos k - 1)} = -\frac{1}{k^2} + O(1). \quad (94)$$

This is a consequence of translation invariance of the difference equation. In other words for this difference equation the solution can be obtained up to a rigid translation and this shows up in the inverse discrete Fourier transform as a singularity. One can make the integral convergent by adding a suitable rigid translation. The following would be a rigid translation that removes the singularity.

$$x_p = \frac{1}{2\pi} \int_{-\pi}^{\pi} e^{-ipk} \left[ \frac{1}{2(\cos k - 1)} \widehat{f}_p(k) - \frac{e^{ipk}}{2(\cos k - 1)} \right] dk. \quad (95)$$

For  $\mathbb{R}$ -valued difference equations there are rigorous treatments of this problem in the literature (see De Boor et al. (1989) and Veit (2003)). In a special case when the loading sequence is symmetric about  $p = 0$  the inverse DFT is convergent. An example would be the following.

$$f_{-p} = f_p \quad \forall p \in \mathbb{N}, \quad f_0 = 0. \quad (96)$$

In this case  $\widehat{f}_p(0) = 0$  and the inverse DFT is convergent.

Consider the following linear vector-valued partial difference equation with constant coefficient matrices.

$$\sum_{\alpha=-r_1}^{r_1} \sum_{\beta=-r_2}^{r_2} \sum_{\gamma=-r_3}^{r_3} \mathbf{A}_{\alpha\beta\gamma} \mathbf{U}_{m+\alpha, n+\beta, p+\gamma} = \mathbf{F}_{mnp} \quad (m, n, p) \in \mathbb{Z}^3. \quad (97)$$

Taking DFT from both sides of the above equation, we obtain

$$\mathcal{Z}(\mathbf{k}) \widehat{\mathbf{U}}_{mnp}(\mathbf{k}) = \widehat{\mathbf{F}}_{mnp}(\mathbf{k}) \quad \mathbf{k} \in \mathbf{B} = [-\pi, \pi]^3, \quad (98)$$



where

$$\mathcal{Z}(\mathbf{k}) = \sum_{\alpha=-r_1}^{r_1} \sum_{\beta=-r_2}^{r_2} \sum_{\gamma=-r_3}^{r_3} e^{-i(\alpha k_1 + \beta k_2 + \gamma k_3)} \mathbf{A}_{\alpha\beta\gamma} \quad (99)$$

is the symbol of the difference equation. Assuming that  $\mathcal{Z}(\mathbf{k})$  is invertible in  $\mathbf{B}$  the solution can be written as

$$\mathbf{U}_{mnp} = \frac{1}{(2\pi)^3} \int_{\mathbf{B}} e^{-i\mathbf{m}\cdot\mathbf{k}} \mathcal{Z}^{-1}(\mathbf{k}) \widehat{\mathbf{F}}_{mnp}(\mathbf{k}) d\mathbf{k}, \quad (100)$$

where  $\mathbf{m} = (m, n, p)$ . Symbol of a difference equation is not invertible, in general. An example would be singularity of the symbol at  $\mathbf{k} = \mathbf{0}$  for a translation-invariant difference equation. Assuming that origin is the only singularity point, solution of the difference equation can be obtained by imposing a suitable rigid translation. The following is a suitable choice.

$$\mathbf{U}_{mnp} = \frac{1}{(2\pi)^3} \int_{\mathbf{B}} e^{-i\mathbf{m}\cdot\mathbf{k}} [\mathcal{Z}^{-1}(\mathbf{k}) - e^{i\mathbf{m}\cdot\mathbf{k}} \mathbf{D}(\mathbf{k})] \widehat{\mathbf{F}}_{mnp}(\mathbf{k}) d\mathbf{k}, \quad (101)$$

where

$$\mathbf{D}(\mathbf{k}) = \begin{pmatrix} \mathbf{U}(\mathbf{k}) & \dots & \mathbf{U}(\mathbf{k}) \\ \vdots & & \vdots \\ \mathbf{U}(\mathbf{k}) & \dots & \mathbf{U}(\mathbf{k}) \end{pmatrix}, \quad \mathbf{U}(\mathbf{k}) = \begin{pmatrix} d_1(\mathbf{k}) & 0 & 0 \\ 0 & d_2(\mathbf{k}) & 0 \\ 0 & 0 & d_3(\mathbf{k}) \end{pmatrix},$$

$$d_1(\mathbf{k}) = (\mathcal{Z}^{-1}(\mathbf{k}))_{11}, d_2(\mathbf{k}) = (\mathcal{Z}^{-1}(\mathbf{k}))_{22}, d_3(\mathbf{k}) = (\mathcal{Z}^{-1}(\mathbf{k}))_{33}. \quad (102)$$

An alternative approach to remove the singularity is as follows. Let us first introduce the following change of variables

$$\bar{\mathbf{U}}_{mnp} = (-1)^{m+n+p} \mathbf{U}_{mnp}, \quad \bar{\mathbf{F}}_{mnp} = (-1)^{m+n+p} \mathbf{F}_{mnp}, \quad \bar{\mathbf{A}}_{\alpha\beta\gamma} = (-1)^{\alpha+\beta+\gamma} \mathbf{A}_{\alpha\beta\gamma}. \quad (103)$$

The governing equations in terms of the new variables are

$$\sum_{\alpha=-r_1}^{r_1} \sum_{\beta=-r_2}^{r_2} \sum_{\gamma=-r_3}^{r_3} \bar{\mathbf{A}}_{\alpha\beta\gamma} \bar{\mathbf{U}}_{m+\alpha, n+\beta, p+\gamma} = \bar{\mathbf{F}}_{mnp} \quad (m, n, p) \in \mathbb{Z}^3. \quad (104)$$

The above system of difference equations is not translation invariant. As an example, let us look at the difference equation (90). Defining  $\bar{x}_p = (-1)^p x_p$  and  $\bar{f}_p = (-1)^p f_p$ , the difference equation is rewritten as

$$-\bar{x}_{p+1} - 2\bar{x}_p - \bar{x}_{p-1} = (-1)^p \bar{f}_p \quad p \in \mathbb{Z}. \quad (105)$$

It is seen that this equation is not translation invariant. The solution of the original difference equation can be written as

$$x_p = \frac{(-1)^p}{2\pi} \int_{-\pi}^{\pi} \frac{e^{-ipk}}{2(i \sin k - 1)} \widehat{f}_p(k) dk. \quad (106)$$

#### 4.1.5 Difference Equations on Finite Domains: Periodic Boundary Conditions

In any numerical treatment of defects in crystals, e.g., molecular dynamics, ab initio calculations, etc., one takes a supercell large enough to be a representative of the defective crystal and then extends it to the whole space periodically. A useful tool for solving difference equations with periodic boundary conditions is the discrete DFT. Here we explain some of its details for applications to ordinary difference equations. But the results can be extended to partial difference equations with no difficulty.

Consider a function  $f : I \rightarrow \mathbb{R}$ , where  $I = \{0, 1, \dots, N-1\}$ ,  $N \in \mathbb{N}$  and  $f$  is periodic, i.e.,

$$f(m + kN) = f(m) \quad \forall k \in \mathbb{Z}. \quad (107)$$

$f$  can be thought of as a finite sequence with  $N$  elements. Discrete DFT of  $f$  is defined as

$$\hat{f}(k) = \sum_{m=0}^{N-1} f(m) \omega_N^{mk} \quad \forall k \in I, \quad (108)$$

where  $\omega_N = e^{-\frac{2\pi i}{N}}$ . The inverse of discrete DFT has the following representation

$$f(n) = \frac{1}{N} \sum_{k=0}^{N-1} \hat{f}(k) \omega_N^{-nk} \quad \forall n \in I. \quad (109)$$

For a sequence  $\{x_n\}_{n=1}^{N-1}$  it can be easily shown that

$$\widehat{x_{n+1}}(k) = \omega_N^{-k} \widehat{x_n}(k) + \omega_N^{-k} (x_n - x_0). \quad (110)$$

Similarly

$$\widehat{x_{n-1}}(k) = \omega_N^k \widehat{x_n}(k) + x_{-1} - x_{N-1}. \quad (111)$$

Let us consider the following discrete boundary-value problem

$$\begin{aligned} x_{n+1} - \alpha x_n + x_{n-1} &= f_n & n = 0, 1, \dots, N-1, \\ x_0 &= x_N \quad (x_{-1} = x_{N-1}). \end{aligned} \quad (112)$$

Taking DFT from both sides one obtains

$$Z(k) \widehat{x_n}(k) = \widehat{f_n}(k), \quad (113)$$

where  $Z(k) = \omega_N^k + \omega_N^{-k} - \alpha$ . Thus

$$\widehat{x_n}(k) = Z(k)^{-1} \widehat{f_n}(k) \quad (114)$$

and hence

$$x_n = \frac{1}{N} \sum_{k=0}^{N-1} Z(k)^{-1} \widehat{f_n}(k) \omega_N^{-nk} \quad n \in I. \quad (115)$$

This would be the solution sequence as long as  $Z(k) \neq 0$ . Suppose  $\alpha = 2$ , e.g. the governing equation of a chain of atoms with harmonic interactions between nearest neighbors. In this case the difference equation is translation invariant and hence  $Z(0) = 0$ . In (115) let us remove the  $k = 0$  term and define

$$\bar{x}_n = \frac{1}{N} \sum_{k=1}^{N-1} Z(k)^{-1} \widehat{f}_n(k) \omega_N^{-nk} \quad n \in I. \quad (116)$$

Note that

$$x_n - \bar{x}_n = \frac{1}{N} Z(0)^{-1} \sum_{m=0}^{N-1} f_m, \quad (117)$$

which is a constant, i.e.  $\bar{x}_n$  is equal to  $x_n$  up to a rigid translation. Note also that if the system is self-equilibrated, i.e. if  $\sum_{m=0}^{N-1} f_m = 0$  then the singularity of  $Z(k)$  at  $k = 0$  causes no problem.

Let us now consider a vector-valued difference equation and define DFT componentwise. Thus for  $\mathbf{X}_n \in \mathbb{R}^d$  define

$$\widehat{\mathbf{X}}_n(k) = \sum_{m=0}^{N-1} \omega_N^{mk} \mathbf{X}_m \quad n \in I. \quad (118)$$

Therefore one can show the following two relations easily.

$$\widehat{\mathbf{X}}_{n+1}(k) = \omega_N^{-k} \widehat{\mathbf{X}}_n(k) + \omega_N^{-k} (\mathbf{X}_N - \mathbf{X}_0), \quad (119)$$

$$\widehat{\mathbf{X}}_{n-1}(k) = \omega_N^k \widehat{\mathbf{X}}_n(k) + \mathbf{X}_{-1} - \mathbf{X}_{N-1}. \quad (120)$$

Consider the following discrete boundary-value problem with a periodic boundary condition.

$$\begin{aligned} \mathbf{A}_{-1} \mathbf{X}_{n-1} + \mathbf{A}_0 \mathbf{X}_n + \mathbf{A}_1 \mathbf{X}_{n+1} &= \mathbf{F}_n & n = 0, \dots, N-1, \\ \mathbf{X}_N = \mathbf{X}_0, \mathbf{X}_{-1} &= \mathbf{X}_{N-1}. \end{aligned} \quad (121)$$

Taking DFT from both sides and using the boundary conditions, formally one has

$$\widehat{\mathbf{X}}_n(k) = \mathbf{Z}(k)^{-1} \widehat{\mathbf{F}}_n(k) \quad (122)$$

and hence

$$\mathbf{X}_n = \frac{1}{N} \sum_{k=0}^{N-1} \mathbf{Z}(k)^{-1} \widehat{\mathbf{F}}_n(k) \omega_N^{-nk} \quad n = 0, \dots, N-1, \quad (123)$$

where  $\mathbf{Z}(k) = \omega_N^k \mathbf{A}_{-1} + \mathbf{A}_0 + \omega_N^{-k} \mathbf{A}_1$ . Note that for a translation-invariant difference equation  $\mathbf{Z}(0)$  is singular and the solution is obtained by removing the term  $k = 0$ .

## 5 Lattice Statics Analysis of a Defective Lattice of Point Dipoles

In this section we consider a two-dimensional defective lattice of dipoles with lattice parameter  $a$ . Each lattice point represents a unit cell and the corresponding dipole is somehow a measure of the distortion of the unit cell with respect to the high symmetry phase. This system is interesting in the sense that its potential energy is not only a function of atom (unit cell) positions; it depends on polarization vectors too. This means that the potential energy is partially translation invariant. Total energy of the lattice is assumed to have the following three parts

$$\mathcal{E}(\{\mathbf{x}^i\}_{i \in \mathcal{L}}, \{\mathbf{P}^i\}_{i \in \mathcal{L}}) = \mathcal{E}^d(\{\mathbf{x}^i, \mathbf{P}^i\}_{i \in \mathcal{L}}) + \mathcal{E}^{\text{short}}(\{\mathbf{x}^i\}_{i \in \mathcal{L}}) + \mathcal{E}^a(\{\mathbf{P}^i\}_{i \in \mathcal{L}}), \quad (124)$$

where,  $\mathcal{E}^d$ ,  $\mathcal{E}^{\text{short}}$  and  $\mathcal{E}^a$  are the dipole energy, short-range energy and anisotropy energy, respectively. These energies have the following forms. The first term is

$$\begin{aligned} \mathcal{E}^d = \frac{1}{2} \sum_{\substack{i, j \in \mathcal{L} \\ j \neq i}} \left[ \frac{\mathbf{P}^i \cdot \mathbf{P}^j}{|\mathbf{x}^i - \mathbf{x}^j|^3} - \frac{3\mathbf{P}^i \cdot (\mathbf{x}^i - \mathbf{x}^j) \mathbf{P}^j \cdot (\mathbf{x}^i - \mathbf{x}^j)}{|\mathbf{x}^i - \mathbf{x}^j|^5} \right] \\ + \sum_{i \in \mathcal{L}} \frac{1}{2\alpha} \mathbf{P}^i \cdot \mathbf{P}^i, \end{aligned} \quad (125)$$

where  $\alpha$  is the electric polarizability and is assumed to be the same for all the lattice points (molecules).\*\* The short-range energy is modelled by a Lennard-Jones potential with the following form

$$\mathcal{E}^{\text{short}} = \frac{1}{2} \sum_{\substack{i, j \in \mathcal{L} \\ j \neq i}} 4\epsilon \left[ \left( \frac{a}{|\mathbf{x}^i - \mathbf{x}^j|} \right)^{12} - \left( \frac{a}{|\mathbf{x}^i - \mathbf{x}^j|} \right)^6 \right]. \quad (126)$$

The anisotropy energy quantifies the tendency of the lattice to remain in some energy wells. We assume the following form for this energy

$$\mathcal{E}^a = \sum_{i \in \mathcal{L}} K_A |\mathbf{P}^i - \mathbf{P}_1|^2 \dots |\mathbf{P}^i - \mathbf{P}_s|^2. \quad (127)$$

This means that the dipoles prefer to have values in the set  $\{\mathbf{P}_1, \dots, \mathbf{P}_s\}$ . Note that this is a self-energy.

Let  $\mathcal{S} = (\{\mathbf{x}^i\}_{i \in \mathcal{L}}, \{\mathbf{P}^i\}_{i \in \mathcal{L}})$  be the equilibrium configuration (a local minimum of the energy), i.e.,

$$\frac{\partial \mathcal{E}}{\partial \mathbf{x}^i} = \frac{\partial \mathcal{E}}{\partial \mathbf{P}^i} = \mathbf{0} \quad \forall i \in \mathcal{L}. \quad (128)$$

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\*\*Note that the last part of this energy is a self-energy.

Linearizing the governing equations (128) about a reference configuration  $\mathcal{B}_0 = (\{\mathbf{x}_0^i\}_{i \in \mathcal{L}}, \{\mathbf{P}_0^i\}_{i \in \mathcal{L}})$  we obtain

$$\begin{aligned} & \frac{\partial \mathcal{E}}{\partial \mathbf{x}^i}(\mathcal{B}_0) + \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{x}^i}(\mathcal{B}_0)(\mathbf{x}^i - \mathbf{x}_0^i) + \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0)(\mathbf{x}^j - \mathbf{x}_0^j) \\ & + \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^i \partial \mathbf{x}^i}(\mathcal{B}_0)(\mathbf{P}^i - \mathbf{P}_0^i) + \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{x}^i}(\mathcal{B}_0)(\mathbf{P}^j - \mathbf{P}_0^j) + \dots = \mathbf{0}, \end{aligned} \quad (129)$$

$$\begin{aligned} & \frac{\partial \mathcal{E}}{\partial \mathbf{P}^i}(\mathcal{B}_0) + \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{P}^i}(\mathcal{B}_0)(\mathbf{x}^i - \mathbf{x}_0^i) + \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i}(\mathcal{B}_0)(\mathbf{x}^j - \mathbf{x}_0^j) \\ & + \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^i \partial \mathbf{P}^i}(\mathcal{B}_0)(\mathbf{P}^i - \mathbf{P}_0^i) + \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{P}^i}(\mathcal{B}_0)(\mathbf{P}^j - \mathbf{P}_0^j) + \dots = \mathbf{0} \end{aligned} \quad (130)$$

where  $\mathcal{S}_i$  is the neighboring set of atom  $i$ . Note that the only contribution to the term  $\frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^i \partial \mathbf{P}^i}(\mathcal{B}_0)$  comes from the anisotropy energy and the polarizability part of the dipole-dipole energy and has the following form for the case of  $s = 2^*$

$$\begin{aligned} \mathbf{K}_0^{pp} & := \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^i \partial \mathbf{P}^i}(\mathcal{B}_0) \\ & = 2K_A (|\mathbf{P}_0^i - \mathbf{P}_1|^2 + |\mathbf{P}_0^i - \mathbf{P}_2|^2) \mathbf{I} + 4K_A (\mathbf{P}_0^i - \mathbf{P}_1) \otimes (\mathbf{P}_0^i - \mathbf{P}_2) \\ & \quad + 4K_A (\mathbf{P}_0^i - \mathbf{P}_2) \otimes (\mathbf{P}_0^i - \mathbf{P}_1) + \frac{1}{\alpha} \mathbf{I}, \end{aligned} \quad (131)$$

where  $\mathbf{I}$  is the  $2 \times 2$  identity matrix and  $\otimes$  denotes tensor product. Assuming interactions of order  $m$ , for a defective crystal with a 1-D symmetry reduction the set  $\mathcal{S}_i$  can be partitioned as follows

$$\mathcal{S}_i = \bigsqcup_{\alpha=-m}^m \bigsqcup_{I=1}^N \mathcal{S}_{I\alpha}(i), \quad (132)$$

where  $\mathcal{S}_{I\alpha}(i)$  is the set of atoms of type  $I$  lying on the line parallel to the  $y$ -axis and  $\alpha a$  away from the atom  $i$ .<sup>†</sup> Let us define

$$\mathbf{u}^i = \mathbf{x}^i - \mathbf{x}_0^i, \quad \mathbf{q}^i = \mathbf{P}^i - \mathbf{P}_0^i, \quad (133)$$

$$\mathbf{f}_i^x = -\frac{\partial \mathcal{E}}{\partial \mathbf{x}^i}(\mathcal{B}_0), \quad \mathbf{f}_i^p = -\frac{\partial \mathcal{E}}{\partial \mathbf{P}^i}(\mathcal{B}_0), \quad (134)$$

\*But note that  $\mathbf{K}_0^{pp}$  has contributions from dipole-dipole interactions.

<sup>†</sup>Here we have assumed that each unit cell has  $N$  dipoles and the defect lies on the line  $x = 0$ .

and

$$\begin{aligned}
\mathbf{K}_{iI\alpha}^{xx} &= \delta_{\alpha 0} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{x}^i}(\mathcal{B}_0) + \sum_{j \in \mathcal{S}_{I\alpha}(i)} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0), \\
\mathbf{K}_{iI\alpha}^{xp} &= \delta_{\alpha 0} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^i \partial \mathbf{x}^i}(\mathcal{B}_0) + \sum_{j \in \mathcal{S}_{I\alpha}(i)} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{x}^i}(\mathcal{B}_0), \\
\mathbf{K}_{iI\alpha}^{px} &= \delta_{\alpha 0} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{P}^i}(\mathcal{B}_0) + \sum_{j \in \mathcal{S}_{I\alpha}(i)} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i}(\mathcal{B}_0), \\
\mathbf{K}_{iI\alpha}^{pp} &= \delta_{\alpha 0} \mathbf{K}_0^{pp} + \sum_{j \in \mathcal{S}_{I\alpha}(i)} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{P}^i}(\mathcal{B}_0). \tag{135}
\end{aligned}$$

With the above definitions the linearized governing equations can be written as

$$\sum_{\alpha=-m}^m \sum_{I=1}^N \mathbf{K}_{iI\alpha}^{xx} \mathbf{u}_\alpha^I + \sum_{\alpha=-m}^m \sum_{I=1}^N \mathbf{K}_{iI\alpha}^{xp} \mathbf{q}_\alpha^I = \mathbf{f}_i^x, \tag{136}$$

$$\sum_{\alpha=-m}^m \sum_{I=1}^N \mathbf{K}_{iI\alpha}^{px} \mathbf{u}_\alpha^I + \sum_{\alpha=-m}^m \sum_{I=1}^N \mathbf{K}_{iI\alpha}^{pp} \mathbf{q}_\alpha^I = \mathbf{f}_i^p. \tag{137}$$

Now by simply looking at the linearized equations, one would expect to see translation-invariance for the variable  $\mathbf{u}_\alpha^I$ . This means that the sum of matrices that act on  $\mathbf{u}_\alpha^I$  cannot be full rank, i.e., in  $\mathbb{R}^2$

$$\mathcal{N} \left( \sum_{\alpha=-m}^m \sum_{I=1}^N \mathbf{K}_{iI\alpha}^{xx} \right) = \mathcal{N} \left( \sum_{\alpha=-m}^m \sum_{I=1}^N \mathbf{K}_{iI\alpha}^{px} \right) = 2 \tag{138}$$

Note that this is not the case for matrices that act on  $\mathbf{q}_\alpha^I$  variables.

One should note that dipole-dipole energy is pairwise but not isotropic. In other words, the energy is the sum of pairwise interaction of dipoles but for each pair the energy is not only a function of the relative distance of dipoles; in addition to relative distances it depends on the dot product of the relative position vectors and the polarization vectors of the two dipoles.

It is easy to show that for dipole-dipole energy the following holds (as a consequence of translation invariance)

$$\frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{x}^i}(\mathcal{B}_0) = - \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0). \tag{139}$$

Note also that

$$\sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0) = \sum_{j \in \mathcal{S}_{i0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0) + \sum_{j \in \mathcal{S}_i \setminus \mathcal{S}_{i0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0), \tag{140}$$

where  $\mathcal{S}_{i_0}$  is the equivalence class with  $i$  as its representative. Thus

$$\frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{x}^i}(\mathcal{B}_0) + \sum_{j \in \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0) = - \sum_{j \in \mathcal{S}_i \setminus \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0). \quad (141)$$

It should be noted that only the dipole-dipole energy contributes to  $\frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i}$ .

It is an easy exercise to show that for dipole-dipole energy the following holds

$$\frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{P}^i} = \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^i \partial \mathbf{x}^i} = - \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i}. \quad (142)$$

This can be restated as

$$\frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^i \partial \mathbf{P}^i} + \sum_{j \in \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i} = - \sum_{j \in \mathcal{S}_i \setminus \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i}. \quad (143)$$

As a consequence of (141) and (143) we have

$$\mathbf{K}_{II0}^{xx} = - \sum_{\alpha=-m}^m \sum_{\substack{J=1 \\ J \neq I}}^N \mathbf{K}_{IJ\alpha}^{xx} \quad \text{and} \quad \mathbf{K}_{II0}^{px} = - \sum_{\alpha=-m}^m \sum_{\substack{J=1 \\ J \neq I}}^N \mathbf{K}_{IJ\alpha}^{px}. \quad (144)$$

The substiffness matrix  $\mathbf{K}_{II0}^{xp}$  has a more complicated structure. Note that

$$\begin{aligned} \mathbf{K}_{II0}^{xp} &= \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^i \partial \mathbf{x}^i} + \sum_{j \in \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{x}^i} \\ &= - \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i} + \sum_{j \in \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{x}^i} \\ &= - \sum_{j \in \mathcal{S}_i \setminus \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i} - \sum_{j \in \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i} + \sum_{j \in \mathcal{S}_{i_0}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{x}^i} \\ &= \mathbf{K}_{II0}^{px} + \sum_{j \in \mathcal{S}_{i_0}} \left( \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{x}^i} - \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^i} \right). \end{aligned} \quad (145)$$

The linearized governing equations can be written in a more compact form in terms of interaction of unit cells as

$$\sum_{\alpha=-m}^m \mathcal{A}_\alpha \mathbf{U}_{n+\alpha} = \mathbf{F}_n, \quad (146)$$

where

$$\begin{aligned} \mathbf{A}_\alpha &= \begin{pmatrix} \mathbf{A}_\alpha^{xx} & \mathbf{A}_\alpha^{xp} \\ \mathbf{A}_\alpha^{px} & \mathbf{A}_\alpha^{pp} \end{pmatrix} \in \mathbb{R}^{4N \times 4N}, \quad \mathbf{U}_m = \{\mathbf{u}_m^1 \dots \mathbf{u}_m^N \mathbf{q}_m^1 \dots \mathbf{q}_m^N\}^\top, \\ \mathbf{F}_m &= \{\mathbf{f}_{m1}^x \dots \mathbf{f}_{mN}^x \mathbf{f}_{m1}^p \dots \mathbf{f}_{mN}^p\}^\top, \\ \mathbf{A}_\alpha^{**} &= \begin{pmatrix} \mathbf{K}_{11\alpha}^{**} & \dots & \mathbf{K}_{1N\alpha}^{**} \\ \vdots & & \vdots \\ \mathbf{K}_{N1\alpha}^{**} & \dots & \mathbf{K}_{NN\alpha}^{**} \end{pmatrix} \in \mathbb{R}^{N \times N} \quad *, \star = x, p. \end{aligned} \quad (147)$$

For a defective crystal with a 2-D symmetry reduction discrete governing equations can be obtained similarly.

### 5.1 Hessian for the Bulk Lattice

The linearized governing equations in the bulk can be written as

$$\sum_{J=1}^N \mathbf{K}_{IJ}^{xx} \mathbf{u}^J + \sum_{J=1}^N \mathbf{K}_{IJ}^{xp} \mathbf{q}^J = \mathbf{f}_I^x \quad I = 1, \dots, N, \quad (148)$$

$$\sum_{J=1}^N \mathbf{K}_{IJ}^{px} \mathbf{u}^J + \sum_{J=1}^N \mathbf{K}_{IJ}^{pp} \mathbf{q}^J = \mathbf{f}_I^p \quad I = 1, \dots, N, \quad (149)$$

where

$$\begin{aligned} \mathbf{f}_I^x &= -\frac{\partial \mathcal{E}}{\partial \mathbf{x}^I}(\mathcal{B}_0), \quad \mathbf{f}_I^p = -\frac{\partial \mathcal{E}}{\partial \mathbf{P}^I}(\mathcal{B}_0), \\ \mathbf{K}_{IJ}^{xx} &= \delta_{IJ} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^I \partial \mathbf{x}^J}(\mathcal{B}_0) + \sum_{j \in \mathcal{L}_I \setminus \{I\}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{x}^I}(\mathcal{B}_0), \\ \mathbf{K}_{IJ}^{xp} &= \delta_{IJ} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^J \partial \mathbf{x}^I}(\mathcal{B}_0) + \sum_{j \in \mathcal{L}_I \setminus \{I\}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{x}^I}(\mathcal{B}_0), \\ \mathbf{K}_{IJ}^{px} &= \delta_{IJ} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^J \partial \mathbf{P}^I}(\mathcal{B}_0) + \sum_{j \in \mathcal{L}_I \setminus \{I\}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{x}^j \partial \mathbf{P}^I}(\mathcal{B}_0), \\ \mathbf{K}_{IJ}^{pp} &= \delta_{IJ} \mathbf{K}_0^{pp} + \sum_{j \in \mathcal{L}_I \setminus \{I\}} \frac{\partial^2 \mathcal{E}}{\partial \mathbf{P}^j \partial \mathbf{P}^I}(\mathcal{B}_0) \quad I, J = 1, \dots, N. \end{aligned} \quad (150)$$

Assuming that  $s = 2(\text{number of proffered polarizations})$ ,  $\mathbf{P}_1 = -\mathbf{P}_2 = \mathbf{P}_0$ , one has  $\mathbf{K}_0^{pp} = (8K_A P_0^2 + \frac{1}{\alpha}) \mathbf{I}$ . Now the Hessian is written as

$$\mathbf{H} = \begin{pmatrix} \mathbf{K}_{11} & \dots & \mathbf{K}_{1N} \\ \vdots & & \vdots \\ \mathbf{K}_{N1} & \dots & \mathbf{K}_{NN} \end{pmatrix}, \quad \mathbf{K}_{IJ} = \begin{pmatrix} \mathbf{K}_{IJ}^{xx} & \mathbf{K}_{IJ}^{xp} \\ \mathbf{K}_{IJ}^{px} & \mathbf{K}_{IJ}^{pp} \end{pmatrix}, \quad I, J = 1, \dots, N \quad (151)$$



Note that  $\mathbf{H}$  has a zero eigenvalue of multiplicity two that represents the  $\mathbf{x}$ -translation invariance of the governing equations.

It is known that (Tosi and Doyama, 1967; Faux, 1971) point dipole model does not describe the true physics of polarizable molecules, especially in small distances. One example of breakdown of this model is ‘polarization catastrophe’ which is an instability in energy minimization of systems governed by point-dipole interactions. In our calculations, we observed that the Hessian of the dipole-dipole potential is not positive-definite. However, by adding a short-range energy and an anisotropy energy the total Hessian can be positive-definite. We do not argue that our model potential represents any physical system. Our goal here is to demonstrate the power of our theory of anharmonic lattice statics for analysis of a defective crystal governed by a stable potential.

## 5.2 Example 1: A 180° Domain Wall in a 2-D Lattice of Dipoles

Let us look at a 180° domain wall and consider the reference configuration shown in Fig. 6. In a 180° domain wall, polarization vector changes from  $-\mathbf{P}_0$  on the left side of the domain wall to  $\mathbf{P}_0$  on the right side of the domain wall. We are interested in understanding the structure of the defective lattice close to the domain wall. In this example, each equivalent class is a set of atoms lying on a line parallel to the domain wall, i.e., we have a defective crystal with a 1-D symmetry reduction. As we will see shortly, this is a simple but rich example. For index  $n$  in the reduced lattice (see Fig. 6), the vector of unknowns is

$$\mathbf{U}_n = \{\mathbf{u}_n, \mathbf{q}_n\}^T \in \mathbb{R}^4. \quad (152)$$

Consider a square lattice with lattice vectors  $\mathbf{e}_1 = \{a, 0\}^T$  and  $\mathbf{e}_2 = \{0, a\}^T$  with polarization vectors  $\mathbf{P} = P_0\{0, 1\}^T$ . In the bulk, because of symmetry  $\mathbf{f}^x = \mathbf{0}$ . It can be easily shown that

$$\mathbf{f}^p = - \sum_{\substack{j \in \mathcal{L} \\ j \neq i}} \left\{ \frac{\mathbf{P}}{|\mathbf{x}^i - \mathbf{x}^j|^3} - 3 \frac{\mathbf{P} \cdot (\mathbf{x}^i - \mathbf{x}^j)(\mathbf{x}^i - \mathbf{x}^j)}{|\mathbf{x}^i - \mathbf{x}^j|^5} \right\} - \frac{1}{\alpha} \mathbf{P}. \quad (153)$$

Multiplying both sides by  $\mathbf{P}$  and enforcing  $\mathbf{f}^p = \mathbf{0}$  we obtain

$$\frac{P_0^2}{\alpha} = - \sum_{\substack{j \in \mathcal{L} \\ j \neq i}} \left\{ \frac{P_0^2}{|\mathbf{x}^i - \mathbf{x}^j|^3} - 3 \frac{[\mathbf{P} \cdot (\mathbf{x}^i - \mathbf{x}^j)]^2}{|\mathbf{x}^i - \mathbf{x}^j|^5} \right\}. \quad (154)$$

Thus for the bulk polarization  $\mathbf{P} = P_0\{0, 1\}^T$  we have

$$\alpha = \left[ \sum_{\substack{(m,n) \in \mathbb{Z}^2 \\ (m,n) \neq (0,0)}} \frac{1}{a^3} \left\{ \frac{3n^2}{(m^2 + n^2)^{\frac{5}{2}}} - \frac{1}{(m^2 + n^2)^{\frac{3}{2}}} \right\} \right]^{-1}. \quad (155)$$

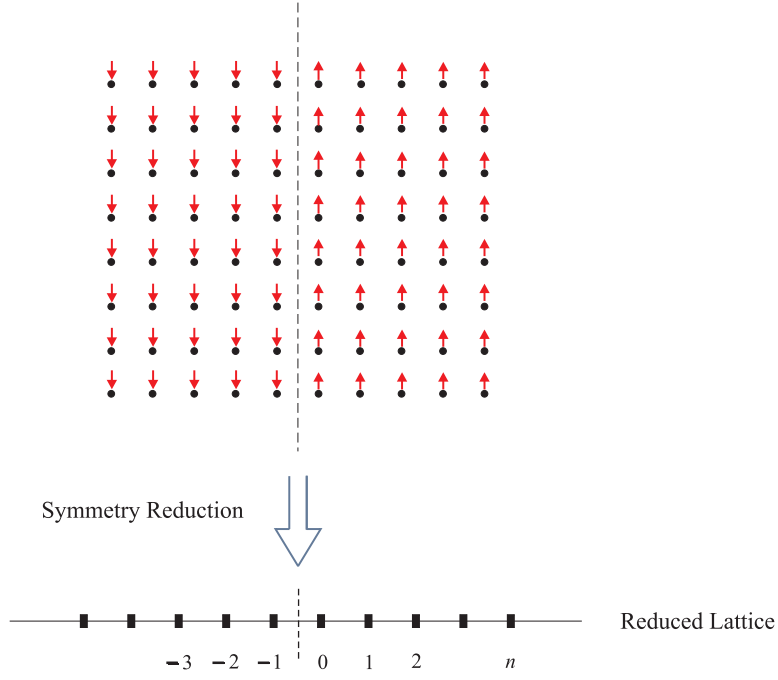


Figure 6: Reference configuration for a  $180^\circ$  domain wall in the 2-D lattice of dipoles, its symmetry reduction and its reduced lattice. Note that domain wall is not a crystallographic plane.

Given the lattice parameter  $a$  and the bulk polarization  $\mathbf{P}_0$ , polarizability  $\alpha$  is uniquely determined for the bulk lattice. We can now check positive-definiteness of the Hessian for a the bulk lattice.

### Solution of The Governing Difference Equation

Let us assume that  $m = 1$ , i.e. a given unit cell interacts only with its nearest neighbor equivalence classes. Solution for an arbitrary  $m$  can be found similarly. Because of symmetry it is possible to work with only one half of the lattice. Bulk equations for the right half lattice are

$$\mathbf{A}_{-1}\mathbf{U}_{n-1} + \mathbf{A}_0\mathbf{U}_n + \mathbf{A}_1\mathbf{U}_{n+1} = \mathbf{F}_n \quad n \geq 1. \quad (156)$$

Governing equations for  $n = 0$  are boundary equations. These can be written as (note that  $\mathbf{U}_{-1} = -\mathbf{U}_0$ )

$$(\mathbf{A}_0^b - \mathbf{A}_{-1}^b)\mathbf{U}_0 + \mathbf{A}_1^b\mathbf{U}_1 = \mathbf{F}_0, \quad (157)$$

where the superscript  $b$  is to emphasize that the boundary stiffness matrices are in general different from the bulk ones. Let us define the following variable

$$\mathbf{X}_n = \begin{pmatrix} \mathbf{U}_{n-1} \\ \mathbf{U}_n \end{pmatrix} \quad n \geq 1. \quad (158)$$

Now the governing equation for  $\mathbf{X}_n$  is

$$\mathbf{X}_{n+1} = \mathcal{A}\mathbf{X}_n + \mathbf{G}_n \quad n \geq 1, \quad (159)$$

where

$$\mathcal{A} = \begin{pmatrix} \mathbf{0} & \mathbf{1} \\ -\mathbf{A}_1^{-1}\mathbf{A}_{-1} & -\mathbf{A}_1^{-1}\mathbf{A}_0 \end{pmatrix} \in \mathbb{R}^{8 \times 8}, \quad \mathbf{G}_n = \begin{pmatrix} \mathbf{0} \\ \mathbf{A}_1^{-1}\mathbf{F}_n \end{pmatrix} \in \mathbb{R}^8. \quad (160)$$

Note that because of symmetry

$$\mathbf{U}_n = -\mathbf{U}_{-n-1} \quad n \leq -1, \quad (161)$$

Assuming that  $\mathbf{F}_n = 0$  for  $n > M$  we have

$$\begin{aligned} \mathbf{X}_2 &= \mathcal{A}\mathbf{c} + \mathbf{G}_1, \\ \mathbf{X}_3 &= \mathcal{A}^2\mathbf{c} + \mathcal{A}\mathbf{G}_1 + \mathbf{G}_2, \\ &\vdots \\ \mathbf{X}_{M+1} &= \mathcal{A}^M\mathbf{c} + \mathcal{A}^{M-1}\mathbf{G}_1 + \dots + \mathbf{G}_M = \mathcal{A}^M\mathbf{c} + \mathbf{d}, \\ \mathbf{X}_{M+2} &= \mathcal{A}(\mathcal{A}^M\mathbf{c} + \mathbf{d}), \\ &\vdots \\ \mathbf{X}_n &= \mathcal{A}^{n-(M+2)}(\mathcal{A}^M\mathbf{c} + \mathbf{d}) \quad n \geq M+1, \end{aligned} \quad (162)$$

where  $\mathbf{c} = \mathbf{X}_1 = \{\mathbf{U}_0, \mathbf{U}_1\}^\top$  and  $\mathbf{d} = \mathcal{A}^{M-1}\mathbf{G}_1 + \dots + \mathbf{G}_M$ . For the potential we use it turns out that  $M = 5$  (see Fig. 7).

A physically meaningful solution should be bounded at infinity. The matrix  $\mathcal{A}$  is not diagonalizable because of translation invariance of the governing equations.<sup>††</sup> However, it has the following Jordan decomposition

$$\mathcal{A} = \mathbf{X}\mathbf{\Lambda}\mathbf{X}^{-1}, \quad (163)$$

where  $\mathbf{X}$  is the matrix of generalized eigenvectors and  $\mathbf{\Lambda}$  has the following form

$$\mathbf{\Lambda} = \begin{pmatrix} \mathbf{\Lambda}_1 & & \\ & \mathbf{J} & \\ & & \mathbf{\Lambda}_2 \end{pmatrix} \in \mathbb{R}^8, \quad \mathbf{\Lambda}_1, \mathbf{\Lambda}_2 \in \mathbb{R}^2. \quad (164)$$

Here  $\mathbf{\Lambda}_1$  and  $\mathbf{\Lambda}_2$  are diagonal matrices of eigenvalues of modulus greater than and smaller than 1, respectively and  $\mathbf{J} \in \mathbb{R}^{4 \times 4}$  is the Jordan block corresponding to the eigenvalue  $\lambda = 1$  with multiplicity four. Now for  $n \geq M+1$

$$\mathbf{X}_n = \mathbf{X}\mathbf{\Lambda}^{n-(M+2)}(\mathbf{\Lambda}^M\mathbf{X}^{-1}\mathbf{c} + \mathbf{X}^{-1}\mathbf{d}). \quad (165)$$

---

<sup>††</sup>This is the case only for  $\mathbf{u}_n$ .

Boundedness equations can be written as

$$(\mathbf{\Lambda}^M \mathbf{X}^{-1} \mathbf{c})_{\{1, \dots, 4\}} = -(\mathbf{X}^{-1} \mathbf{d})_{\{1, \dots, 4\}}, \quad (166)$$

where  $(\cdot)_{\{1, \dots, 4\}}$  means the first four rows of the matrix  $(\cdot)$ . Boundary and boundedness equations give us the vector of unknowns  $\mathbf{c}$ .

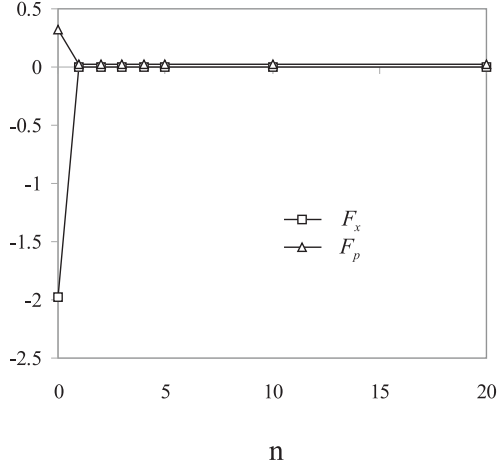


Figure 7: Unbalanced forces in the reference configuration of a  $180^\circ$  domain wall in the 2-D lattice of dipoles.  $F_x$  is the component of  $\mathbf{f}^x$  perpendicular to the domain wall and  $F_p$  is the component of  $\mathbf{f}^p$  parallel to the domain wall. Other force components are zero because of symmetry.

The harmonic and anharmonic solution for the numerical values of  $a = 1.0$ ,  $P_0 = 1.0$ ,  $\epsilon = \frac{1.0}{8}$ ,  $K_A = 2.0$  are shown in Fig. 8. Note that because of symmetry for a given unit cell number  $n$ ,  $\mathbf{U}_n = \{u_x \ 0 \ 0 \ q_y\}^T$ . Anharmonic lattice statics iterations converged after almost ten iterations. For convergence tolerance for displacement and polarization unbalanced forces are  $10^{-4} \frac{\epsilon}{a}$  and  $10^{-4} \frac{P_0}{\alpha}$ , respectively. The harmonic solution for the range of interaction  $m = 2$  differs from that of the range of interaction  $m = 1$  by less than 0.5% and the anharmonic displacements are the same. This means that the effective potential is highly localized and considering  $m = 1$  is enough.<sup>†</sup> However, in each step unbalanced forces are calculated exactly. It is seen that a  $180^\circ$  domain wall is two lattice spacings thick. Interestingly, this is in qualitative agreement with our calculations with shell potentials for  $\text{BaTiO}_3$  and  $\text{PbTiO}_3$  (Yavari, 2004) and also with ab initio calculations (Meyer and Vanderbilt, 2001). To understand the effect of different parameters of the potential on the domain wall structure,

<sup>†</sup>In all the following numerical examples  $m = 1$  is chosen.

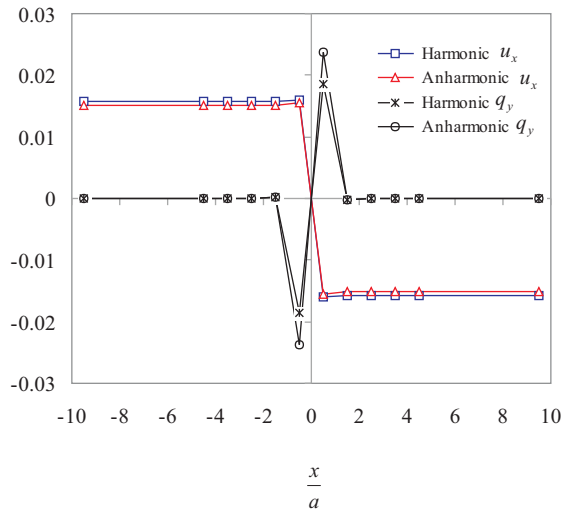


Figure 8: Harmonic and anharmonic displacements in  $180^\circ$  domain wall in the 2-D lattice of dipoles.

we consider the following four systems

$$S1 : a = 1.0, P_0 = 1.0, \epsilon = \frac{10.0}{8}, K_A = 2.0 \quad (167)$$

$$S2 : a = 1.0, P_0 = 1.0, \epsilon = \frac{1.0}{8}, K_A = 2.0 \quad (168)$$

$$S3 : a = 1.0, P_0 = 1.0, \epsilon = \frac{1.0}{8}, K_A = 10.0 \quad (169)$$

$$S4 : a = 1.0, P_0 = 0.5, \epsilon = \frac{1.0}{8}, K_A = 2.0 \quad (170)$$

S2 is the system we just discussed. S1 has a short energy ten times larger than that of S2 and S3 has an anisotropic energy five times larger than that of S1. S4 has a polarization with half of the magnitude of that of S1. The anharmonic displacements of these four systems are compared in Fig. 9. It is seen that S1 has the smallest  $u_x$  displacements and the other three systems have almost the same  $u_x$  displacements. This shows that the  $\mathbf{x}$  displacements are controlled by short-range energy and the more dominant the short-range energy the smaller the  $\mathbf{x}$  displacements. S1 and S2 have the same  $u_q$  and this is not surprising as they have the same polarizations and the same anisotropic energies. S3, which has the largest anisotropic energy, has the smallest  $u_q$ . S4 has the largest  $u_q$  which means that the smaller the dipole-dipole contribution the larger the polarization displacements. An important observation is that all the four systems have the same domain wall thickness. This is not surprising as a simple dimensional

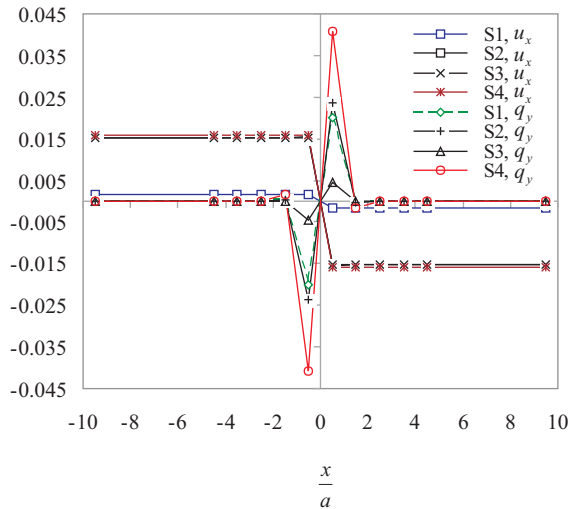


Figure 9: Harmonic and anharmonic displacements for four different choices of the interatomic potential.

analysis shows that the domain wall thickness is proportional to lattice spacing  $a$ .

**Remark.** In this example, we presented the exact harmonic solutions. One can solve an approximate harmonic problem by using a homogenized system in terms of the stiffness matrices. In general, stiffness matrices on the left and right sides of the wall (and also stiffness matrices for the unit cells close to the wall) are different. One can average the stiffness matrices and then use DFT for solving the resulting homogeneous vector-valued difference equation. The harmonic solutions are different from the exact solutions but as the unbalanced forces are calculated exactly both exact and approximate harmonic solutions lead to the same anharmonic solutions.

### Sensitivity of Solutions to the Choice of Reference Configuration: One and Two-Parameter Families of Reference Configurations

Now one may wonder what would happen if one starts with a different reference configuration. We studied the effect of choice of reference configuration on the solutions by looking at the one and two-parameter families of reference configurations shown in Fig. 10. In Fig. 10 (a), we assume that polarization vectors in the two layers adjacent to the wall have magnitude  $KP_0$ . We solved the governing equations for different  $K$  values ( $K \in [0.25, 1.75]$ ) and did not observe any new equilibrium configuration for any of these large perturbations

form the original nominal defect. In Fig. 10 (b) a two-parameter family of reference configurations is shown. In this case, polarization vectors in the two layers adjacent to the wall have magnitudes  $K_1 P_0$  or  $K_2 P_0$ . To be able to reduce the governing partial difference equations to an ordinary difference equation one needs to choose a larger unit cell as shown in the figure. Again for any choice of  $K_1, K_2 \in [0.25, 1.75]$ , we obtained the same equilibrium configuration for the domain wall.

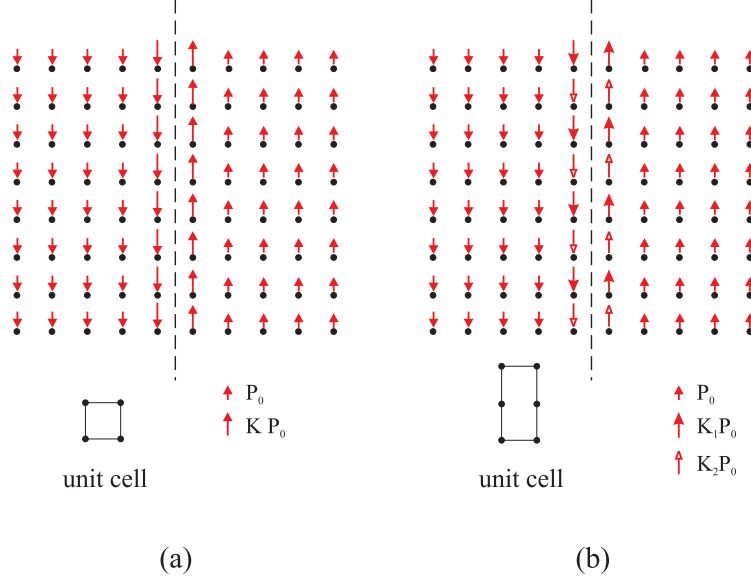


Figure 10: (a) A one-parameter family of reference configurations for the  $180^\circ$  domain wall. (b) A two-parameter family of reference configurations for the same  $180^\circ$  domain wall.

### 5.3 Example 2: A Second Type of $180^\circ$ Domain Wall

The reference configuration for this type of  $180^\circ$  domain wall is shown in Fig. 11. In this case lattice vectors are  $\mathbf{e}_1 = \{\frac{a}{\sqrt{2}}, -\frac{a}{\sqrt{2}}\}^\top$  and  $\mathbf{e}_2 = \{-\frac{a}{\sqrt{2}}, \frac{a}{\sqrt{2}}\}^\top$  with polarization vectors  $\mathbf{P} = P_0\{0, 1\}^\top$ . In the bulk, again because of symmetry  $\mathbf{f}^x = \mathbf{0}$  and one can show that

$$\alpha = \left[ \frac{1}{a^3} \sum_{\substack{(m,n) \in \mathbb{Z}^2 \\ (m,n) \neq (0,0)}} \left\{ \frac{3(m-n)^2}{2(m^2+n^2)^{\frac{5}{2}}} - \frac{1}{(m^2+n^2)^{\frac{3}{2}}} \right\} \right]^{-1}. \quad (171)$$

The form of governing equations are exactly similar to the previous example

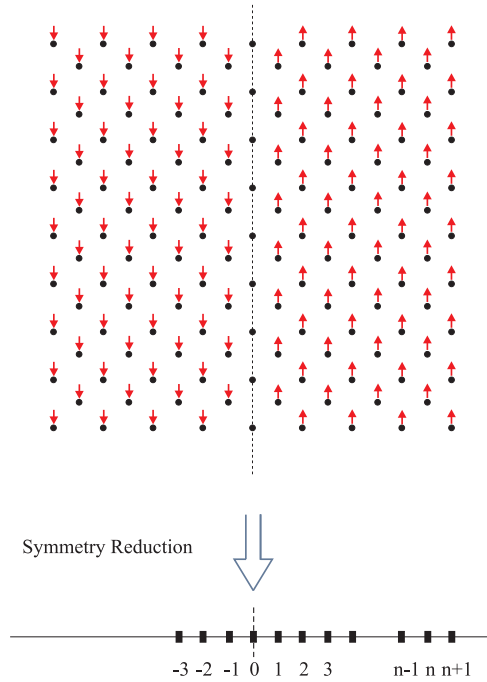


Figure 11: Reference configuration for a  $180^\circ$  domain wall in the 2-D lattice of dipoles, its symmetry reduction and its reduced lattice. In this example domain wall passes through some atoms, i.e., it is a crystallographic line.

and again because of symmetry we can reduce the problem to a half lattice. In this example equivalence classes are lines of atoms parallel to the domain wall and  $\frac{a}{\sqrt{2}}$  apart from one another. Unbalanced forces are again highly localized. Fig. 12 shows the harmonic and anharmonic solutions for both position vectors and polarization for the numerical values of  $a = 1.0$ ,  $P_0 = 0.5$ ,  $\epsilon = 1.0$ ,  $K_A = 2.0$ . Again, displacements are perpendicular to the domain wall and polarization displacements are parallel to the wall, i.e., for unit cell number  $n$ ,  $\mathbf{U}_n = \{u_x \ 0 \ 0 \ q_y\}^T$ . It is seen that the first harmonic solution is dramatically different from the next iterations and the anharmonic solutions. This is, in general, not surprising and shows the inadequacy of harmonic solutions for a chosen reference configuration.

#### 5.4 Example 3: A $90^\circ$ Domain Wall in a 2-D Lattice of Dipoles

In this example a  $90^\circ$  domain wall is considered. The reference configuration is shown in Fig. 13. Governing equations have a form similar to that of  $180^\circ$



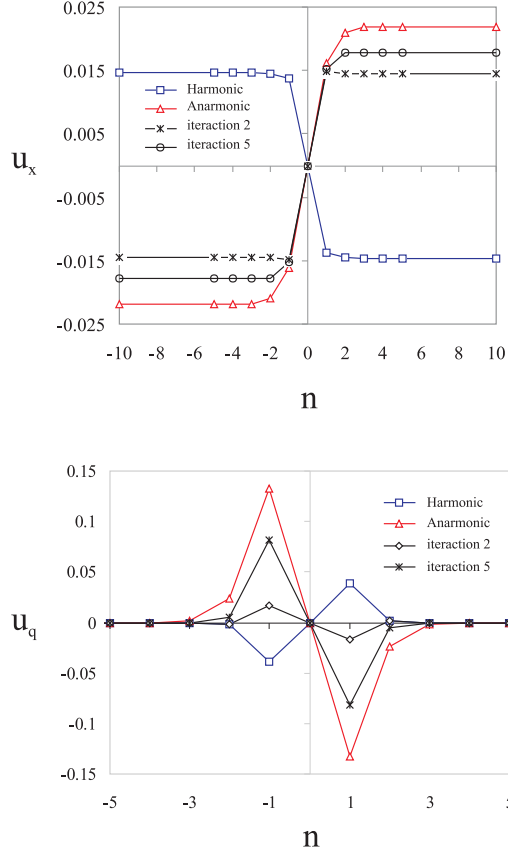


Figure 12: Harmonic and anharmonic displacements in  $180^\circ$  domain wall in the 2-D lattice of dipoles.

domain walls. Symmetry of the domain wall implies that polarization force is nonzero only parallel to the domain wall, i.e., because of symmetry for the  $n$ th unit cell  $\mathbf{U}_n = \{u_x \ u_y \ 0 \ q_y\}^T$ . We also have the following symmetry

$$\mathbf{U}_{-n} = \mathbf{R}\mathbf{U}_{-n-1}, \quad \mathbf{R} = \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \quad n \geq 1. \quad (172)$$

This reduces the problem to a half lattice similar to what we saw for  $180^\circ$  domain walls. For the numerical values of  $a = 1.0$ ,  $P_0 = 1.0$ ,  $\epsilon = 1.0$ ,  $K_A = 2.0$  displacements are shown in Fig. 14. Polarization displacements are shown in Fig. 15. It is seen that the domain wall is very sharp and its thickness

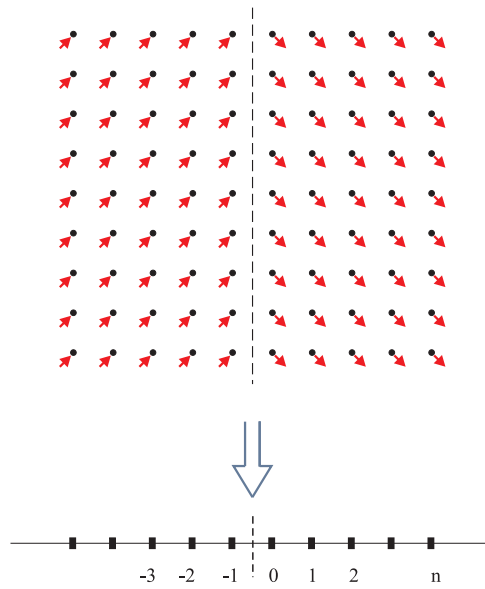


Figure 13: Reference configuration for a  $90^\circ$  domain wall in the 2-D lattice of dipoles, its symmetry reduction and its reduced lattice. Note that domain wall is not a crystallographic line.

is comparable to those of  $180^\circ$  domain walls. Interestingly, this is similar to what has been seen for ferroelectric  $180^\circ$  and  $90^\circ$  domain walls in tetragonal  $\text{PbTiO}_3$  using shell potentials (Yavari, 2004) and ab initio calculations (Meyer and Vanderbilt, 2001).

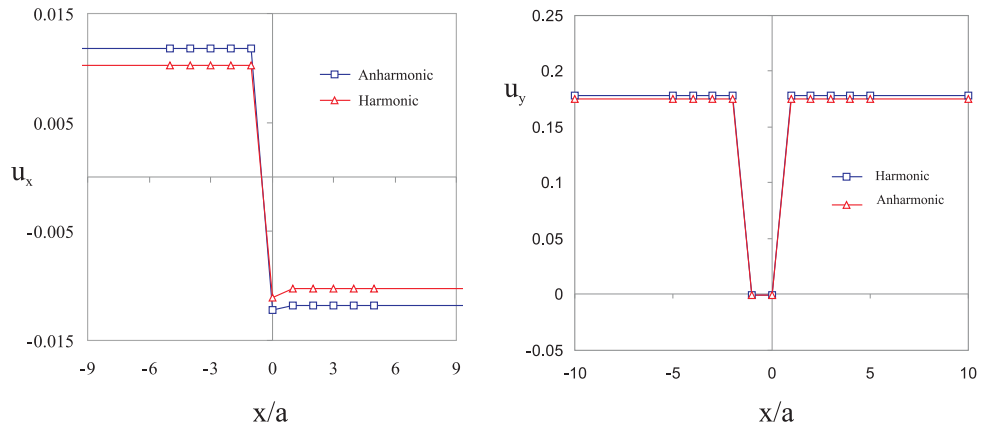


Figure 14: Unit cell displacements in a  $90^\circ$  domain wall in the 2-D lattice of dipoles.

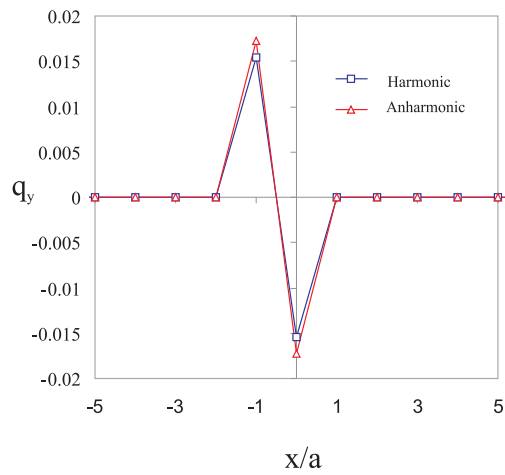


Figure 15: Polarization displacement in a  $90^\circ$  domain wall in the 2-D lattice of dipoles.

## 6 Conclusions

In this paper we developed a general theory of anharmonic lattice statics that can be used in systematic analysis of a defective crystal given an interatomic potential. This differs from all the existing treatments in that it does not apply only to Bravais lattices and does not rely on a knowledge of force constants. Instead, it can be used for arbitrary defective lattices and all is needed is an interatomic potential describing the interaction of atoms. We started by looking at deformation of a crystal from a given reference configuration as a discrete deformation mapping and presented all the developments in a language very similar to continuum mechanics.

We explained how one should construct the discrete governing equations for a given complex lattice. The discrete governing equations are linearized about a reference configuration. The reference configuration is arbitrary and problem dependent and in general not force-free and perhaps not translation invariant. Our experience shows that a nominal defect structure could be a good reference configuration. Linearizing the (nonlinear) discrete governing equations about the reference configuration leads to a nonhomogeneous system of linear difference equations with variable coefficient matrices. The forcing terms are a result of the fact that the reference configuration is not a local minimum of the energy, in general. We call these forces the unbalanced forces. We classified defective complex lattices into three groups, namely defective crystals with 1-D, 2-D and no symmetry reductions. Exploiting a symmetry reduces the dimensionality of the discrete governing equations and this leads to numerically more efficient solution techniques. Having analytic solutions for linearized governing equations, the anharmonic solution can be obtained by modified Newton-Raphson iterations. The idea is to keep the initial stiffness matrices and update the reference configuration by calculating the unbalanced forces in each step. We explained the convergence issue for long range interactions and our presentation is not just formal.

For solving the harmonic displacements we used methods from theory of difference equations. Our solution technique for an infinite defective crystal with a 1-D symmetry reduction is novel. For more complicated defective crystals with 2-D and no symmetry reductions we use discrete Fourier transform (DFT) for solving the governing partial difference equations. We explained the subtleties in using DFT for translation-invariant difference equations.

As an example of a non-isotropic pairwise potential in which atom position vectors are not the only degrees of freedom, we considered a lattice of point dipoles. Dipole-dipole interactions are pairwise but anisotropic as the potential energy of two dipoles depends on the dot product of the relative position vector and the polarization vectors in addition to the relative distance of the two dipoles. It was shown that our general formulation of lattice statics can easily handle such a system. We were able to solve two types of  $180^\circ$  domain wall problems. It was observed that the domain wall thickness is about two lattice spacings. Interestingly, this is in quantitative agreement with our calculations with shell potentials for the ferroelectric tetragonal  $\text{PbTiO}_3$  and also ab initio

calculations. In one domain wall problem, it was observed that harmonic and anharmonic solutions are dramatically different. This shows the importance of anharmonic effects close to defects. We also solved a  $90^\circ$  domain wall problem. It was observed that domain wall thickness is again about two lattice spacings. This is again similar to what has been observed for ferroelectric domain walls in  $\text{PbTiO}_3$  using shell potentials and ab initio calculations.

We believe this method can be useful in generating semi-analytical solutions for many different systems with defects. The semi-analytical solutions can be very useful for validating numerical techniques. Semi-analytical solutions can also be useful in studying different interatomic potentials. We believe this development is a step forward in rationalizing lattice scale calculations.

## Acknowledgments

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## 7 Appendix: Three-Body Interactions

In this appendix we consider three-body interactions and discuss some of the modifications that should be made in the theory that we developed in the bulk of this paper. As the effect of pairwise interactions can be studied separately, let us assume that a collection of atoms  $\mathcal{L}$  is governed by only three-body interactions. Generalizing the results of this appendix to arbitrary N-body interactions is straightforward. The energy of the system can be written as

$$\mathcal{E} = \frac{1}{6} \sum_{\substack{i,j,k \in \mathcal{L} \\ (j,k) \neq (i,i)}} \phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k). \quad (173)$$

Note that  $\phi$  is invariant under permutations of  $i, j, k$ . For example,  $\phi(\mathbf{x}^k, \mathbf{x}^j, \mathbf{x}^i) = \phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k)$ . Because of material-frame-indifference  $\phi$  has the following dependence on the position vectors (Keating, 1966)

$$\phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k) = \psi(r^{ij}, r^{jk}, r^{ki}, \omega^{ijk}, \omega^{jik}, \omega^{kij}), \quad (174)$$

where

$$r^{pq} = |\mathbf{x}^p - \mathbf{x}^q|, \quad \omega^{pqs} = (\mathbf{x}^p - \mathbf{x}^q) \cdot (\mathbf{x}^p - \mathbf{x}^s). \quad (175)$$

Force on an atom  $i$  comes from interactions of  $i$  with pairs of atoms  $j, k$ . Contribution of the triplet  $(i, j, k)$  to this force is

$$\mathbf{f}_i(i, j, k) = -\frac{\partial \phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k)}{\partial \mathbf{x}^i}. \quad (176)$$

$\mathbf{f}_j(i, j, k)$  and  $\mathbf{f}_k(i, j, k)$  are defined similarly. It is an easy exercise to show that

$$\mathbf{f}_i(i, j, k) + \mathbf{f}_j(i, j, k) + \mathbf{f}_k(i, j, k) = \mathbf{0}. \quad (177)$$

This is the analogue of the relation  $\mathbf{f}_{ji} = -\mathbf{f}_{ij}$  for pairwise interactions. It is easy to show that balance of angular momentum is trivially satisfied provided that balance of linear momentum is already satisfied.

Neighboring set  $\mathcal{S}_i$  of atom  $i \in \mathcal{L}$  is the set of all the atoms that interact with  $i$ . By definition,  $i \notin \mathcal{S}_i$ . Neighboring set  $\mathcal{S}_{ij}$  of the pair of atoms  $(i, j)$ ,  $i \neq j$  is the set of atoms in  $\mathcal{L}$  that interact with the pair  $(i, j)$ . By definition,  $i, j \notin \mathcal{S}_{ij}$ . Note also that  $\mathcal{S}_{ij} = \mathcal{S}_i \setminus \{j\}$ .

Atom energy  $\mathcal{E}^i$  can be defined as one sixth of the energy of all the triplets of atoms adjacent to  $i$ . Pair-atom energy  $\mathcal{E}^{ij}$  is one half of the energy of all the triplets of atoms adjacent to the pair  $(i, j)$ . Note that energy of the triplet  $(i, j, k)$  is trivially defined as

$$\mathcal{E}^{ijk} = \phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k). \quad (178)$$

Let us consider a discrete system of atoms without any external body forces. Linearization of the governing equations about a reference configuration  $\mathcal{B}_0$  can be expressed as

$$\frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^i \partial \mathbf{x}^i}(\mathcal{B}_0) \mathbf{u}^i + \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i}(\mathcal{B}_0) \mathbf{u}^j = -\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i}(\mathcal{B}_0). \quad (179)$$

This can be simplified to read

$$\frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^i \partial \mathbf{x}^i} (\mathcal{B}_0) \mathbf{u}^i + \sum_{j \in \mathcal{S}_i} \sum_{k \in \mathcal{S}_{ij}} \frac{\partial^2 \phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k)}{\partial \mathbf{x}^j \partial \mathbf{x}^i} (\mathcal{B}_0) \mathbf{u}^j = -\frac{\partial \mathcal{E}^i}{\partial \mathbf{x}^i} (\mathcal{B}_0). \quad (180)$$

Now suppose the defective crystal has a 1-D symmetry reduction, i.e.,

$$\mathcal{S}_i = \bigsqcup_{\alpha=-\infty}^{\infty} \bigsqcup_{l=1}^N \mathcal{S}_{I\alpha}(i). \quad (181)$$

Thus

$$\begin{aligned} \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i} (\mathcal{B}_0) \mathbf{u}^j &= \sum_{\alpha=-\infty}^{\infty} ' \sum_{l=1}^N \sum_{j \in \mathcal{S}_{I\alpha}(i)} \sum_{k \in \mathcal{S}_{ij}} \frac{\partial^2 \phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k)}{\partial \mathbf{x}^j \partial \mathbf{x}^i} (\mathcal{B}_0) \mathbf{u}^{I\alpha} \\ &= \sum_{\alpha=-\infty}^{\infty} ' \sum_{l=1}^N \mathbf{K}_{iI\alpha} \mathbf{u}^{I\alpha}, \end{aligned} \quad (182)$$

where

$$\mathbf{K}_{iI\alpha} = \sum_{j \in \mathcal{S}_{I\alpha}(i)} \sum_{k \in \mathcal{S}_{ij}} \frac{\partial^2 \phi(\mathbf{x}^i, \mathbf{x}^j, \mathbf{x}^k)}{\partial \mathbf{x}^j \partial \mathbf{x}^i} (\mathcal{B}_0). \quad (183)$$

For three-body interactions the following relation holds (as a consequence of translation invariance)

$$\frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^i \partial \mathbf{x}^i} = - \sum_{j \in \mathcal{S}_i} \frac{\partial^2 \mathcal{E}^i}{\partial \mathbf{x}^j \partial \mathbf{x}^i}. \quad (184)$$

This means that for a defective crystals with 1-D and 2-D symmetry reductions, respectively, we have

$$\mathbf{K}_{II0} = - \sum_{\alpha=-\infty}^{\infty} ' \sum_{\substack{J=1 \\ J \neq I}}^N \mathbf{K}_{IJ\alpha}, \quad (185)$$

$$\mathbf{K}_{II0} = - \sum_{\alpha, \beta=-\infty}^{\infty} ' \sum_{\substack{J=1 \\ J \neq I}}^N \mathbf{K}_{IJ\alpha\beta}, \quad (186)$$

which is exactly what we had for pairwise interactions. This is not surprising as the above relations are a consequence of translation invariance of the lattice irrespective of the form of interactions. In conclusion, it is seen that in our formulation of lattice statics, the only modification needed for three-body interactions is in the definition of substiffness matrices.

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