Optimality of General Lattice Transformations

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December 5, 2017

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1. Introduction

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In his seminal article on "The nature of martensite" Bain [Bai24] proposed a mechanism that transforms the face-centred cubic (fcc) lattice to the body-centred cubic (bcc) lattice, a phase transformation most importantly manifested in low carbon steels. He writes:

> *"It is reasonable, also, that the atoms themselves will rearrange* [...] *by a method that will require least temporary motion.* [...] *A mode of atomic shift requiring minimum motion was conceived by the author* [...] *"*
>
> Bain, 1924

The key observation that led to his famous correspondence was that *"If one regards the centers of faces as corners of a new unit, a body-centered structure is already at hand; however, it is tetragonal instead of cubic"*. He remarks that this is not surprising *"as it is the only easy method of constructing a bcc atomic structure from the fcc atomic structure"*.

Even though now widely accepted, his mechanism, which he illustrated with a model made of cork balls and needles (see Fig. 1), was not without criticism from his contemporaries. In their fundamental paper Kurdjumov & Sachs [KS30] wrote [free translation from German] that "nothing certain about the mechanism of the martensite transformation is known. Bain imagines that a tetragonal unit cell within the fcc lattice transforms into a bcc unit cell through compression along one direction and expansion along the two other. However a proof of this hypothesis is still missing".[1] Interestingly, without being aware of it, the authors implicitly used the Bain mechanism in their derivation of the Kurdjumov & Sachs orientation relationships (see [KM] for details).

In subsequent years, the determination of the transformation mechanism remained of great interest. In their paper on "Atomic Displacements in the Austenite-Martensite Transformation" [JW48] Jaswon and Wheeler again acknowledged that

> *"Off all the possible distortions of a primitive unit cell of the face-centred cubic structure, which could generate a body-centred cubic structure of the given relative orientation, the one which actually occurs is the smallest"*
>
> Jaswon and Wheeler, 1948

[1] *"Über den Mechanismus dieser ,,Martensitumwandlung" ist bisher nichts Sicheres bekannt. Bain stellt sich vor, daß eine tetra- gonalkörperzentrierte Elementarzelle des Austenits durch Schrumpfung in der einen Richtung und Ausdehnung in den beiden anderen in die kubischraumzentrierte des α-Eisens übergeht. Eine Bestätigung für diese Anschauung konnte bisher nicht erbracht werden."*

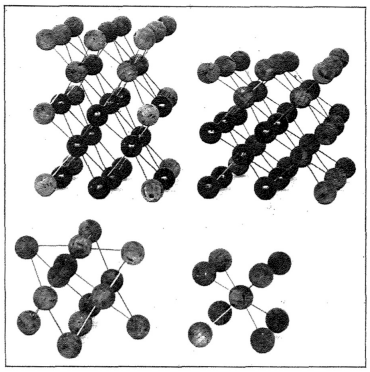

Figure 1: From E.C. Bain: the small models show the fcc and bcc unit cells; the large models represent 35 atoms in an fcc and bcc arrangement respectively.

By combining it with experimental observations of the orientation relationships they devised an algorithm to derive the strain tensor. However their approach is only applicable to cases where the orientation relationship is known a-priori.

With the years passing and a number of supporting experimental results (for a discussion see e.g. [BW72]) the Bain mechanism rose from a conjecture to a widely accepted fact. Nevertheless, almost a century after Bain first announced his correspondence a rigorous proof based on the assumption of minimal atom movement has been missing. Of course, the transformation from fcc-to-bcc is not the only instance where the determination of the transformation strain is of interest. The overall question remains the same: Which transformation strain(s), out of all the possible deformations mapping the lattice of the parent phase to the lattice of the product phase, require(s) the least atomic movement?

To provide a definite answer to this question one first needs to quantify the notion of least atomic movement in such a way that it does not require additional input from experiments. Then one needs to establish a framework that singles out the optimal transformation among the infinite number of possible lattice transformation strains. One way to appropriately quantify least atomic movement is the criterion of smallest principal strains as suggested by Lomer in [Lom55]. In his paper, Lomer compared 1600 different lattice correspondences for the β to α phase transition in Uranium and concluded that only one of them involved strains of less than 10%. More recently, in [CSTR16] an algorithm is proposed to determine the transformation strain based on a similar minimality criterion (see Remark **??**) that also allows for the consideration of different sublattices. The present paper considers a criterion of least atomic movement in terms of a family of different strain measures and, for each such strain measure, rigorously proves the existence of an optimal lattice transformation between *any* two Bravais lattices.[2] As a main application, it is

[2]In particular, no assumptions are made on the type of lattice points (e.g. atoms, molecules) or on the relation between the point groups of the two lattices.

shown that the Bain strain is the optimal lattice transformation from fcc-to-bcc with respect to three of the most commonly used strain measures.

The structure of the paper is as follows: after stating some preliminaries in Section **??** we explore in more depth some mathematical aspects of lattices in Section **??**. This section is mainly intended for the mathematically inclined reader and may be skipped on first reading without inhibiting the understanding of Section **??**, which constitutes the main part of this paper. In this section we establish a geometric criterion of optimality and prove the existence of optimal lattice transformations for any displacive phase transition between two Bravais lattices. Additionally a precise algorithm to compute these optimal strains is provided. In the remaining subsections, the general theory is applied to prove the optimality of the Bain strain in an fcc-to-bcc transformation, to show that the Bain strain remains optimal in an fcc to body-centred tetragonal (bct) transformation and finally to derive the optimal transformation strain between two triclinic phases of Terephthalic Acid. Similarly to the fcc-to-bcc transition, this phase transformation is of particular interest as it involves large stretches and thus the lattice transformation requiring least atomic movement is not clear.

The following notations and definitions are standard and will be used throughout.

Notation. Let $\mathcal{R} \in \{\mathbb{Z}, \mathbb{R}\}$ denote the sets of integers or reals respectively. The set $\mathcal{R}^{m,n}$ denotes the space of $m \times n$ matrices over $\mathcal{R}$ and the set $\mathcal{R}^n$ denotes the space of vectors over $\mathcal{R}$. We equip $\mathcal{R}^n$ with the standard Euclidean basis $\hat{e}_1, \dots, \hat{e}_n$, where $\hat{e}_i = (0, \dots, 1, \dots, 0)$ is the normalized vector whose i-th entry is 1. For a given matrix $A \in \mathcal{R}^{m,n}$ with column vectors $\mathbf{a}_i = A\hat{e}_i$ we write $A = [\mathbf{a}_1, \dots, \mathbf{a}_n]$. Furthermore, we define the multiplication and addition of a matrix F with a set of matrices $\mathcal{S}$ by $F.\mathcal{S} := \{FS : S \in \mathcal{S}\}$ and $F + \mathcal{S} := \{F + S : S \in \mathcal{S}\}$, respectively. The invertible matrices over $\mathcal{R}$ are denoted by $\mathrm{GL}(3, \mathcal{R}) := \mathcal{R}^{3,3} \cap \{A \in \mathcal{R}^{3,3} \text{ is invertible}\}$ and we use the subscript + to denote orientation preserving matrices, i.e. matrices with positive determinant. Furthermore, we denote by $\mathrm{SO}(3) = \{R \in \mathbb{R}^{3,3} : R^{\mathrm{T}}R = \mathbb{I}, \det R = 1\}$ the group of proper rotations.

For a comprehensive review of definitions and lemmata in the context of (simple) lattice transformations we refer the reader to [KM16] and we shall adapt its notation henceforth. We recall the definition of the $\|A\|_{p,q}$ matrix norm of a $m \times n$ matrix

$$\|A\|_{p,q} = \left(\sum_{j=1}^{n} \left(\sum_{i=1}^{m} |a_{ij}|^p \right)^{q/p} \right)^{1/q} = \left(\sum_{j=1}^{n} \|A\hat{e}_j\|_p^q \right)^{1/q},$$

where $p, q \in [1, \infty]$. One norm of particular importance will be the *column max norm* given by $\|A\|_{2,\infty} = \max_{i=1,\dots,n} |A\hat{e}_i|$. Unless otherwise specified, here and throughout the rest of the paper $|\cdot|$ always denotes the Euclidean norm if the argument is a vector in $\mathbb{R}^3$ and the Frobenius norm if the argument is a matrix in $\mathbb{R}^{3,3}$. The

following subsets of $\mathbb{Z}^{3,3}$ will be of particular importance in the development of the framework.

Definition 1. ($\mathrm{SL}_{\mathrm{i}}^{\mathrm{k}}(3,\mathbb{Z})$)
For $k \in \mathbb{N}$ define

$$\mathrm{SL}_{\mathrm{i}}^{\mathrm{k}}(3,\mathbb{Z}) := \{A \in \mathbb{Z}^{3,3} : \det A = i,\ |A_{mn}| \leq k\ \forall\, m,n \in \{1,2,3\}\}$$

and set $\mathrm{SL}_{\mathrm{i}}(3,\mathbb{Z}) = \bigcup_{k=1}^{\infty} \mathrm{SL}_{\mathrm{i}}^{\mathrm{k}}(3,\mathbb{Z})$. Clearly $\mathrm{SL}_{\mathrm{i}}^{\mathrm{k}}(3,\mathbb{Z}) \subset \mathrm{SL}_{\mathrm{i}}^{\mathrm{l}}(3,\mathbb{Z})$ for all $0 \leq k \leq l$ and $i,k,l \in \mathbb{N}$. Table A1 gives an overview of the sizes of $\mathrm{SL}_{\mathrm{i}}^{\mathrm{k}}(3,\mathbb{Z})$ for $i = 1,\ldots,8$ and $k = 1,\ldots,7$.

Furthermore, we recall that, following the arguments of [KM16], a lattice transformation $H : \mathcal{L}_0 \to \mathcal{L}_1$ between two Bravais lattices is optimal with respect to the pseudometric $d_r, r \neq 0$ if it minimizes

$$d_r(H,\mathbb{I}) = |(H^{\mathrm{T}}H)^{r/2} - \mathbb{I}| = \left(\sum_{i=1}^{3}(\nu_i^r - 1)^2\right)^{1/2}, \tag{1}$$

where ν_i, $i = 1,2,3$ are the principal stretches/singular values of H.

1.1. Review of Multilattices

We first recall the definition of a simple Bravais lattice

Definition 2. (Bravais lattice, [Bha03] Ch. 3)
Let $F = [f_1,f_2,f_3] \in \mathrm{GL}_+(3,\mathbb{R})$. We define the *Bravais lattice* $\mathcal{L}(F)$ *generated by* F as the lattice generated by $\mathrm{col}[F]$, i.e.

$$\mathcal{L}(F) := \mathrm{col}[F.\mathbb{Z}_+^{3,3}],$$

where $\mathrm{col}[A]$ denotes the column vectors of A. Thus by definition a Bravais lattice is $\mathrm{span}_{\mathbb{Z}}\{f_1,f_2,f_3\}$ together with an orientation.

Following [PZ02, Ch. 11] an n-multilattice is the union of affine simple lattices and can always be represented in an *essential* form, i.e. the unit cell can be chosen to have minimal volume. We will distinguish between the skeletal (Bravias) lattice $\mathcal{L} = \mathcal{L}(F)$ with $F = [\mathbf{f}_1,\mathbf{f}_2,\mathbf{f}_3] \in \mathrm{GL}_+(3,\mathbb{R})$ that fixes the unit cell and the shift vectors $\{\mathbf{p}_1,\ldots,\mathbf{p}_{n-1}\}$ within that unit cell. To ensure uniqueness, we require that the shift vectors are not related by multiples of the skeletal lattice vectors, i.e.

$$\mathbf{p}_i \neq \mathbf{p}_j + F.\mathbf{l} \tag{2}$$

for all $i,j = 1,\ldots,n-1$ with $i \neq j$ and any $\mathbf{l} \in \mathbb{Z}^3$ (see also Figure 2). We denote the space of vectors $\{\mathbf{f}_1,\mathbf{f}_2,\mathbf{f}_3,\mathbf{p}_1,\ldots,\mathbf{p}_{n-1}\}$ satisfying these conditions by $\mathcal{D}_{n+2}$ and we

view it as a single matrix in $\mathbb{R}^{3,n+2}$, i.e.

$$\mathcal{E} = [\mathbf{f}_1, \mathbf{f}_2, \mathbf{f}_3, \mathbf{p}_1, \ldots, \mathbf{p}_{n-1}] \in \mathcal{D}_{n+2} \subset \mathbb{R}^{3,n+2},$$

where $F = [\mathbf{f}_1, \mathbf{f}_2, \mathbf{f}_3] \in \mathrm{GL}_+(3, \mathbb{R})$ is the generating skeletal matrix and $P = [\mathbf{p}_1, \ldots, \mathbf{p}_{n-1}] \in \mathbb{R}^{3,n-1}$ are the shifts. In analogy to simple lattices we write

$$\mathcal{L}(\mathcal{E}) = \mathcal{L}(F, P) = \bigcup_{i=0}^{n-1} (\mathbf{p}_i + \mathcal{L}(F)), \tag{3}$$

where $\mathbf{p}_0 := (0, 0, 0)$.

Notation. For brevity we will henceforth simply write *essential lattice* to denote a multilattice that is described in an essential form and *non-essential lattice* to denote a multilattice that is described in a non-essential form. Furthermore, we will drop the prefix multi- and simply write lattice instead of multilattice.

2. Essential, monoatomic n-lattices

Each n-lattice $\mathcal{L} = \mathcal{L}(\mathcal{E})$ can be described by several equivalent sets of essential parameters $\mathcal{E}'$. Following [PZ02], if $\mathcal{L} = \mathcal{L}([F, P])$ is an essential monoatomic n-lattice, any equivalent essential parameter $\mathcal{E}' = [F', P']$ is obtained by choosing equivalent lattice vectors for its skeletal lattice, any shift vector as its the base point, any permutation of the shift vectors $P = [\mathbf{p}_1, \ldots, \mathbf{p}_{n-1}] \in \mathbb{R}^{3,n-1}$ and by adding any multiple of the skeletal lattice to the shift vectors. Thus, we have $F' = F.\mu$ with $\mu \in \mathrm{GL}(3, \mathbb{Z})$ and

$$P' = P\alpha + FL,$$

where $L \in \mathbb{Z}^{3,n-1}$ and $\alpha = (a_i^j) \in \mathrm{A}(n) = \mathrm{Perm}(n-1) \cup \bigcup_{k=1}^{(} n-1)(\kappa_k . \mathrm{Perm}(n-1))$. Here, $\mathrm{Perm}(n)$ is a permutation matrix and $\kappa_k := \mathbb{I} - \hat{e}_k \otimes \hat{e}_k - \hat{e}_k \otimes (1, \ldots, 1)$.[3] We denote the group of all transformations between equivalent descriptors by $\Gamma_{n+2} < \mathrm{GL}(n+2, \mathbb{Z})$ and all elements in $\gamma \in \Gamma_{n+2}$ are of the form

$$\gamma \in \Gamma_{n+2} \Leftrightarrow \gamma = \left(\begin{array}{c|c} \mu & L \\ \hline \mathbb{O}_{n-1,3} & \alpha \end{array}\right) \in \mathbb{Z}^{n+2,n+2}, \tag{4}$$

where $L \in \mathbb{Z}^{3,n-1}, \alpha \in \mathrm{A}(n)$ and $\mu \in \mathrm{SL}(3, \mathbb{Z})$.[4]

Lemma 1. *(A criterion for essentialness ([PZ02, p. 312])*
Let the descriptors $\mathcal{E} \in \mathcal{D}_{n+2}$ have lattice metric $K := \mathcal{E}^{\mathrm{T}}\mathcal{E}$. Then $\mathcal{E}$ is an essential

[3] The matrices κ_k allow the interchange of the basepoint of the skeletal lattice with any of the shift vectors.

[4] We note that from a computational point of view it is advantageous to replace L by μL in (4). Thus, when e.g. restricting L to only have values between −1 and 1, it is still possible for all shift vectors to stay inside the (equivalent) unit cell.

descriptors if and only if $\mathbb{I} \in \Gamma_{n+2}$ *is the only matrix* $\gamma \in \Gamma_{n+2}$*, among those satisfying* $\gamma^{\mathrm{T}} K \gamma = K$*, such that its* μ*-component (cf.* (4)*) is the identity in* $\mathrm{SL}(3,\mathbb{Z})$*.*

2.1. Lattice transformations between essential, monoatomic n-lattices

Let $\mathcal{L}(\mathcal{E}) = \mathcal{L}(F,P)$ and $\mathcal{L}(\Sigma) = \mathcal{L}(G,Q)$, where $F,G \in \mathrm{GL}_+(3,\mathbb{R})$ and $P,Q \in \mathbb{R}^{3,n-1}$, be two essential monoatomic n-lattices. Owing to essentialness, mapping one essential, monoatomic n-lattices to another requires

1. mapping the skeletal lattices to each other and
2. mapping the shift vectors to each other.

Generically, it is not possible to find a single transformation strain that would satisfy both conditions. Since the shift vectors are tied to the periodic structure of the skeletal lattice, we will henceforth always enforce that the skeletal lattices are mapped to each other and introduce a penalty for the mismatch between the shift vectors (shuffle movement within the unit cell). We note that we only measure the total distance each atom moves and are not interested in the individual paths.

The resulting minimization problem then reads

$$\mathcal{I}_{r,p,q,\alpha}(\mathcal{E},\Sigma) \coloneqq \min_{\substack{\mu\in\mathrm{SL}(3,\mathbb{Z}),\\ \pi\in\mathrm{A}(n-1),\\ L\in\mathbb{Z}^{3,n-1}}} \Big(\underbrace{d_r(H_\mu,\mathbb{I})}_{\text{skeletal}} + \underbrace{\alpha(\det F)^{-\frac{1}{3}} \|H_\mu P\pi - (Q+GL)\|_{p,q}}_{\text{shift}} \Big), \tag{5}$$

where $p,q \geq 1$, $r \in \mathbb{R}\backslash\{0\}$ and $\alpha \in \mathbb{R}^+$ is some suitable weight. We will refer to the first summand as the *skeletal part* and the second summand as the *shift part*. Details about the derivation of this expression can be found in Appendix A.

3. Non-Essential, monoatomic n-lattices

Since every lattice has an essential description, any non-essential lattice is always the sublattice of some essential lattice with additional shift vectors. Thus we can obtain all non-essential lattices by building them up from essential lattices.

3.1. From essential to non-essential lattices

Let $\mathcal{L}_0 = \mathcal{L}(\mathcal{E}_0)$, $\mathcal{E}_0 = [F_0,P_0] \in \mathbb{R}^{3,n+2}$ be an essential n-lattice. Our goal is to determine all generators $\mathcal{E} \in \mathbb{R}^{3,m+2}$ of non-essential m-lattices that generate the same lattice, i.e. $\mathcal{L}_0 = \mathcal{L}(\mathcal{E})$. The main idea is that the skeletal lattice vectors of $\mathcal{L}_0$ which are not skeletal lattice vectors of $\mathcal{L}$ become shift vectors. Figure 2 illustrates the overall construction.

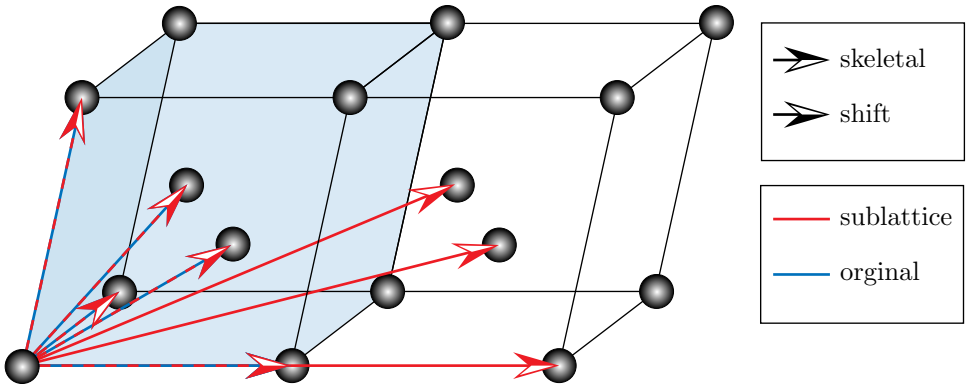

Figure 2: The unit cell spanned by the red skeletal sublattice vectors has 5 shift vectors and spans the same lattice as the essential blue unit cell with 2 shift vectors.

Converting skeletal vectors to shift vectors

For simplicity we start with a simple Bravais lattice $\mathcal{L}_0 = \mathcal{L}(F_0)$. By definition a (Bravais) sublattice $\mathcal{L} = \mathcal{L}(F)$ is a subgroup of $\mathcal{L}_0$ and thus its generator F is related to F_0 by an integral matrix μ, i.e. $F = F_\mu = F_0\mu$, where $\mu \in \mathrm{SL_i}(3,\mathbb{Z})$ for some $i \in \mathbb{N}$. In particular, the unit cell of the sublattice $\mathcal{L} = \mathcal{L}(F_\mu)$ is i times bigger than that of the original lattice $\mathcal{L}_0 = \mathcal{L}(F_0)$.

To preserve the number of atoms per unit volume we ought to introduce $i-1$ additional shift vectors $P^s = P^s_\mu = [\mathbf{p}^s_1, \ldots, \mathbf{p}^s_{i-1}] \in \mathbb{R}^{3,i-1}$ in the sublattice unit cell.[5] As for the regular shift vectors, these new shift vectors should not be related by integer multiples of the skeletal vectors of $\mathcal{L}$, i.e. $\mathbf{p}^s_i \neq \mathbf{p}^s_j + F_\mu.\mathbf{l}$ for all $i \neq j$ and $\mathbf{l} \in \mathbb{Z}^3$. A convenient choice for the new shift vectors is

$$\{\mathbf{p}^s_k\}_{k=1,\ldots,i-1} = \{\sum_{k=1}^{3} \lambda_k \mathbf{f}_k, 0 \leq \lambda_k < 1\} \cap \mathcal{L}(F_0) = \{F.\boldsymbol{\lambda} : \boldsymbol{\lambda} \in [0,1)^3\} \cap \mathcal{L}(F_0), \qquad (6)$$

i.e. we require them to lie in the interior of the unit cell of $\mathcal{L}$. Recalling that $F_\mu = F_0.\mu, \mu \in \mathrm{SL_i}(3,\mathbb{Z})$, we can factor out F_0 to obtain

$$\{\mathbf{p}^s_k\}_{k=1,\ldots,i-1} = F_0.\{\mu.\boldsymbol{\lambda} : \mu.\boldsymbol{\lambda} \in \mathbb{Z}^3, \boldsymbol{\lambda} \in [0,1)^3\}.$$

Using e.g. Cramer's rule this condition can be written as $\{0 \leq (\det \mu^j)/i < 1\}_{j=1,2,3}$, where μ^j denotes the matrix μ with it's j-th column replaced by $\mu\boldsymbol{\lambda}$. In particular, the condition for interior vectors only depends on μ.

[5] We use the superscript s to indicate that the shifts are obtained from the previous skeletal vectors.

Copying the original shift vectors

If $\mathcal{L}_0$ is not a Bravais lattice, we additionally need to copy the original shift vectors P_0 of $\mathcal{L}_0$ in each of the i copies of the original unit cell that compromise the sublattice unit cell. A possible choice is to add the original shift vectors to the shift vectors that were obtained from the previous skeletal vectors, i.e.

$$\{\mathbf{p}_{k,l}\} = \mathbf{p}_k^s + \mathbf{p}_{0l},$$

where $k = 1, \dots, i-1$ and $\mathbf{p}_{0l}$, $l = 1, \dots, n-1$ are the original shift vectors. We note that this choice does not guarantee that all non-skeletal shift vectors lie in the interior of the sublattice unit cell. However, when considering transformations we will allow equivalent descriptions of $\mathcal{L}(F_\mu)$ (cf. (4)) and thus in particular we allow the addition of skeletal vectors from the sublattice unit cell.

In summary, given any essential n-lattice $\mathcal{L}_0 = \mathcal{L}(\mathcal{E}_0) = \mathcal{L}(F_0, P_0)$ and any integral matrix $\mu \in \mathrm{SL_i}(3, \mathbb{Z})$ the generator

$$\mathcal{E}_\mu = [F_\mu, P_\mu] = [\mathbf{f}_1, \mathbf{f}_2, \mathbf{f}_3, \mathbf{p}_1^s, \dots, \mathbf{p}_{\det\mu-1}^s, \mathbf{p}_{1,1}, \dots, \mathbf{p}_{i,n-1}],$$

generates the same lattice, i.e. $\mathcal{L}(\mathcal{E}_\mu) = \mathcal{L}(\mathcal{E}_0)$. In particular, $\mathcal{L}(\mathcal{E}_\mu)$ is an *in*-lattice.

3.2. Lattice transformations between monoatomic n and m lattices

Using the above derivation we are now able to determine optimal lattice transformations between any two monoatomic lattices. As before let us start from the essential descriptions of both lattices. We denote the essential generator of the parent n-lattice by $\mathcal{E}_0 = [\mathbf{f}_{01}, \mathbf{f}_{02}, \mathbf{f}_{03}, \mathbf{p}_{01}, \dots, \mathbf{p}_{0n-1}]$ and the generator of the product m-lattice by $\Sigma_0 = [\mathbf{g}_{01}, \mathbf{g}_{02}, \mathbf{g}_{03}, \mathbf{q}_{01}, \dots, \mathbf{q}_{0m-1}]$. Since the number of atoms cannot change during transformation, we at least need to consider transformations between (non-)essential c-lattices, where $c = \mathrm{lcm}(m, n)$ is the least common multiple of m and n. Thus, compatible sublattice cells of the two lattices are generated by

$$F_\mu = F_0.\mu \text{ and } G_\nu = G_0.\nu, \tag{7}$$

where $\mu \in \mathrm{SL}_{\mathrm{l\cdot lcm(m,n)/n}}(3, \mathbb{Z})$, $\nu \in \mathrm{SL}_{\mathrm{l\cdot lcm(m,n)/m}}(3, \mathbb{Z})$ and $l \in \mathbb{N}$ is any positive integer. Particularly, if $m = n$ and $l = 1$ this case reduces to the previous case of transformations between essential monoatomic n-lattices. Defining $H_\nu^\mu := G_\nu F_\mu^{-1} = G_0(\nu\mu^{-1})F_0^{-1}$ the resulting minimization problem reads

$$\mathcal{I}_{r,p,q,\alpha}(\mathcal{E}, \Sigma) := \min_{\mu,\nu,\pi,L} \left(d_r(H_\nu^\mu, \mathbb{I}) + \alpha(\det F)^{-\frac{1}{3}} \| H_\nu^\mu P_\mu \pi - (Q_\nu + G_\nu L) \|_{p,q} \right), \tag{8}$$

where $p, q \geq 1$, $r \in \mathbb{R}\backslash 0$ and $\alpha \in \mathbb{R}^+$ is some suitable weight (cf. (5)).

Remark 1. In (8) the number l is fixed. However, generally we may not want to prescribe the size of the sublattice unit cell and instead consider the total minimization problem

$$\mathcal{I}_{r,p,q,\alpha}(\mathcal{E},\Sigma) \coloneqq \min_{l\in\mathbb{N}} \min_{\mu_l,\nu_l,\pi,L} \left(d_r(H_{\nu_l}^{\mu_l},\mathbb{I}) + \alpha(\det F)^{-\frac{1}{3}} \|H_{\nu_l}^{\mu_l} P_{\mu_l}\pi - (Q_\nu + G_{\nu_l}L)\|_{p,q} \right), \tag{9}$$

where $\mu_l \in \mathrm{SL}_{\mathrm{l\cdot lcm(m,n)/n}}(3,\mathbb{Z})$ and $\nu_l \in \mathrm{SL}_{\mathrm{l\cdot lcm(m,n)/m}}(3,\mathbb{Z})$. We note that the skeletal term favors larger and larger l. In the limit $l \to \infty$ the material becomes a continuum and thus H approaches $\mathbb{I}$ causing the skeletal term $d_r(H_{\nu_l}^{\mu_l},\mathbb{I})$ to vanish. The shift term on the other hand is proportional to l and thus favors smaller sublattice cells. In particular, generically, both terms balance each other and will result in an optimal size of the sublattice unit cell.

4. Towards solving the minimization problem

So far we have only established the form of the minimization problem but not attempted to solve it. We seek to address the following two questions:

- Can an optimal transformation be found in finitely many steps?
- Can an optimal transformation be found within a feasible amount of time by a computer?

In the case of simple Bravais lattices (cf. [KM16]) both questions had a positive answer and the criterion was provided by the following theorem.

Theorem 1. *Given two lattices $\mathcal{L}_0 = \mathcal{L}(F)$ and $\mathcal{L}_1 = \mathcal{L}(G)$ generated by $F, G \in \mathrm{GL}_+(3,\mathbb{R})$ respectively, there exists a d_r-optimal lattice transformation $H_{\mu_{\min}} = G\mu_{\min}F^{-1}$, for any $r \in \mathbb{R}\backslash\{0\}$. For $r > 0$ all optimal changes of basis are contained in the finite compact sets*

$$d_r: \quad \left\{ \mu \in \mathrm{SL}(3,\mathbb{Z}) : \|\mu\|_{2,\infty}^r \leq \frac{\|F\|_{2,\infty}^r}{\nu_{\min}^r(G)} (m_{0,r}+1) \right\}, \tag{10}$$

where $\nu_{\min}(A)$ denotes the smallest principal stretch/singular value of A and $m_{0,r} \coloneqq d_r(H_{\mathbb{I}},\mathbb{I}) = d_r(GF^{-1},\mathbb{I})$.

Compared to the case of transformations between Bravais lattices our (multi-)lattice minimization problems (5), (8), (9) additionally involve the sets $\mathbb{Z}^{3,n-1}$ and $\mathrm{A}(n-1)$. Since $|\mathbb{Z}^{3,n-1}| = \infty$, we cannot expect to find a solution in a compact set. However, allowing the addition of any multiple of skeletal lattice vectors to the shift vectors results in shift vectors that lie far outside the transformation cell. This behavior seems unphysical and thus we may, and henceforth will, restrict $L \in \mathbb{Z}^{3,n-1}$ to have

values between -1 and 1 and denote this set by $\Lambda(n-1) \coloneqq \{L \in \mathbb{Z}^{3,n-1} : |L_{ij}| \leq 1\}$. For a comparison between the sizes of the individual sets see Figure A1.

4.1. Estimates for essential transformations

By (5) we have $\mathcal{I}_{r,p,q,\alpha}(\mathcal{E}, \Sigma) \geq \min_{\mu \in \mathrm{SL}(3,\mathbb{Z})} d_r(H_\mu, \mathbb{I})$ and thus we can apply (10) trivially with

$$m_{0,r,p,q,\alpha} \coloneqq d_r(GF^{-1}, \mathbb{I}) + \alpha(\det F)^{-\frac{1}{3}} \|GF^{-1}P - Q\|_{p,q}.$$

In particular, the optimal μ is contained in a compact (finite) set. Furthermore, we may reduce the number of cases by conducting a preliminary minimization to find a smaller $m_{0,r}$, i.e.

$$m_{0,r,p,q,\alpha} \coloneqq \min_{\mu_0 \in \mathrm{M}, \pi_0 \in \mathrm{P}, L_0 \in \mathrm{L}} \left(d_r(G\mu_0 F^{-1}, \mathbb{I}) + \alpha(\det F)^{-\frac{1}{3}} \|G\mu_0 F^{-1} P\pi_0 - (Q + GL_0)\|_{p,q} \right),$$

where $\mathrm{M} \subset \mathrm{SL}(3,\mathbb{Z})$, $\mathrm{P} \subset \mathrm{A}(n-1)$ and $\mathrm{L} \subset \Lambda(n-1)$ are some suitably chosen subset. If the n-lattice approaches a Bravais lattice, the shift term $\|Q + G.L - H_\mu P\pi\|_{p,q}$ approaches 0 and thus we cannot find general lower bounds on the shift term. However, when considering a specific phase transformation, the matrices F and G are known explicitely and thus lower bounds can be found and used improve the estimates. In summary, for small enough n the minimization problem can be solved within a feasible time by a computer.

4.2. Estimates for non-essential transformations

Let us first consider a transformation between an n and an m lattice for a fixed l, i.e. the transformation is between $l \cdot \mathrm{lcm}(n,m)$ lattices. By only considering the transformation between the skeletal lattices we again arrive at an estimate of the form (10) with μ replaced by $\nu\mu^{-1}$, where $\mu \in \mathrm{SL}_{\mathrm{l\cdot lcm(m,n)/n}}(3,\mathbb{Z})$ and $\nu \in \mathrm{SL}_{\mathrm{l\cdot lcm(m,n)/m}}(3,\mathbb{Z})$ (cf. (7)). To facilitate our estimates we introduce the following sets.

Definition 3. ($\mathrm{SL}_{\mathrm{m',n'}}(3,\mathbb{Z})$)
Let $m', n' \in \mathbb{N}$ and set

$$\mathrm{SL}_{\mathrm{m',n'}}(3,\mathbb{Z}) \coloneqq \{AB^{-1} : A \in \mathrm{SL}_{\mathrm{m'}}(3,\mathbb{Z}), B \in \mathrm{SL}_{\mathrm{n'}}(3,\mathbb{Z})\}.$$

Using this definition our estimate for the skeletal term becomes

$$\|\eta\|_{2,\infty}^s \leq \frac{\|F\|_{2,\infty}^s}{\nu_{\min}^s(G)}(m_{0,s,p} + 1), \tag{11}$$

where $\eta := \nu\mu^{-1} \in \mathrm{SL}_{\mathrm{l\cdot lcm(m,n)/m,l\cdot lcm(m,n)/n}}(3,\mathbb{Z})$. It is easy to see that $\mathrm{SL}_{m',n'}(3,\mathbb{Z}) \subsetneq \frac{1}{n'} \cdot \mathrm{SL}_{m'n'^2}(3,\mathbb{Z})$ and thus the minimization problem is again finite. However, the set $\mathrm{SL}_{m'n'^2}(3,\mathbb{Z})$ is significantly bigger than $\mathrm{SL}^{k}_{m/n}(3,\mathbb{Z})$ and thus the resulting estimates are not useful from a computational point of view. For computational aspects see Appendix C.2.

By Remark 1, it is clear that in the most general case (9), i.e. if we do not prescribe the size of the sublattice cell, the naive estimate (11), which only takes into account the skeletal term, will fail. However, in practice, the energy penalty from the shift term will result in a finite minimization problem.

5. Appplications

- Terfenol (has big strains)
- tranformation strains from Khausik's book
- fcc to bcc in non-essential form (x)
- pressure induced NaCl (fcc to simple cubic) - see Stokes and Hatch: Phy Rev B
- beryllium titanium
- alloys from Cherry's paper
- ... ?

Databases

- Materials genome project: https://materialsproject.org/
- crystallography open database http://www.crystallography.net/cod/

5.1. HCP to FCC

A common generator for the HCP 2-lattice is given by

$$\mathcal{E} = \left[\begin{pmatrix} \frac{1}{\sqrt{2}} & 0 & 0 \\ \frac{1}{2\sqrt{2}} & \frac{\sqrt{3}}{2\sqrt{2}} & 0 \\ 0 & 0 & \frac{2}{\sqrt{3}} \end{pmatrix}, \begin{pmatrix} \frac{1}{2\sqrt{2}} \\ \frac{1}{2\sqrt{6}} \\ \frac{1}{\sqrt{3}} \end{pmatrix} \right] \tag{12}$$

and for the 1-lattice of the FCC we have

$$\Sigma = \left[\left(\begin{array}{ccc} 0 & \frac{1}{2} & \frac{1}{2} \\ \frac{1}{2} & 0 & \frac{1}{2} \\ \frac{1}{2} & \frac{1}{2} & 0 \end{array} \right)\right] \tag{13}$$

and the coefficients are chosen such that both lattices have the same atom density. Using our algorithm for transformation from 2 to 1 lattices we obtain:

6. Summary and Outlook

A. Calculations

The expression (5) can be derived as follows. First, recall that by [KM16, Lemma 5] all possible lattice transformation between $\mathcal{L}(F)$ and $\mathcal{L}(G)$ are given by $H_\mu = G\mu F^{-1}$. For the shift term, we are allowed to chose any of the equivalent descriptions for the shift vectors in both the parent and the product lattice, i.e. by (4) we need to measure the difference

$$H_\mu(F.L' + P.\pi') - (G.L'' + Q.\pi''), \tag{14}$$

where $L', L'' \in \mathbb{Z}^{3,n-1}$ and $\pi', \pi'' \in \mathrm{A}(n-1)$. Using that $H_\mu F = G.\mu$ and that $\mu.L \in \mathbb{Z}^{3,n-1}$ we can simplify this difference to $H_\mu P.\pi' - G.(L'' - \mu.L') - Q.\pi'') =: H_\mu P.\pi - G.L''' - Q.\pi'$. Furthermore, $L'''\pi'' \in \mathbb{Z}^{3,n-1}$ and $(\pi'')^2 = \mathbb{I}_{n-1}$, so that post-multiplication with π'' yields $H_\mu P.\pi'.\pi'' - G.L'''.\pi'' - Q$. And finally, since $L \coloneqq L'''.\pi'' \in \mathbb{Z}^{3,n-1}$ and $\pi \coloneqq \pi'.\pi'' \in \mathrm{A}(n-1)$ we arrive at $H_\mu P\pi - (Q + GL)$ as claimed. We can now equip this difference of points with any suitable measure of distance, e.g. the $L_{p,q}$ matrix norms.

Finally, the factor $(\det F)^{-\frac{1}{3}}$ is included to ensure scaling invariance. This is necessary since the skeletal part is of multiplicative form, so that e.g. a doubling of both product and parent lattice does not change its value, whereas the shift part is of additive form and thus a doubling of the side length would double it's value. The factor $(\det F)^{-\frac{1}{3}}$ accounts for this effect. To simplify computations one can without loss of generality assume that the unit cell of the parent lattice has unit volume.

k	$i=1$	$i=2$	$i=3$	$i=4$	$i=5$	$i=6$	$i=7$	$i=8$
1	3480	1896	288	240	0	0	0	0
2	67 704	147 696	63 792	145 632	33 360	95 184	18 336	78 228
3	640 824	986 352	1 060 920	1041 312	596 544	1 423 080	501 408	855 536
4	2597 208	4 929 936	3 727 512	6 721 896	2 798 880	6 545 160	2 449 872	7 020 120
5	10 426 488	16 399 536	13 917 240	20 306 568	13 942 248	21 144 168	10 833 264	21 051 192
6	23 527 320	45 022 992	36 455 544	56 593 704	30 165 864	70 632 144	25 548 624	60 653 208
7	65 641 560	107 900 880	92 699 352	129 006 120	80 631 816	155 690 928	80 211 096	136 858 584

Table A1: Comparison of sizes for $\mathrm{SL}_i^k(3,\mathbb{Z})$.

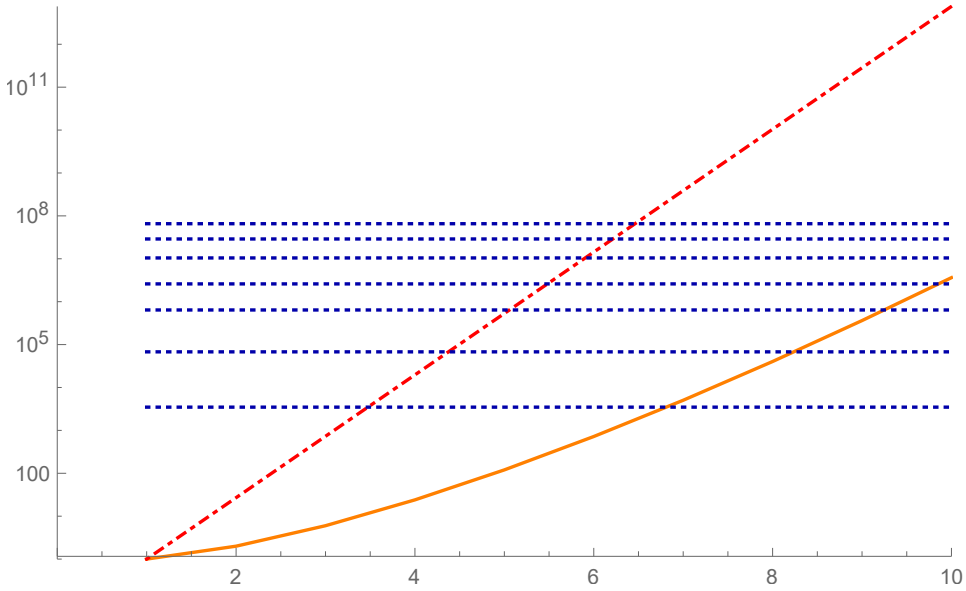

Figure A1: Log-plot of $|\mathrm{A}(n-1)|$ (orange), $|\Lambda(n-1)|$ (red) for $n=1,\ldots,10$ and $|\mathrm{SL}_1^k(3,\mathbb{Z})|$ for $k=1,2,3,4,5,6,7$ (blue, independent of n).

B. Tables and Figures

C. Multi-atomic lattices

In the most general case there are K species of atoms in the unit cell. We shall always assume that one species spans the skeletal lattice and assign it the shift vector $P^1 \in \mathbb{R}^{3,n_1-1}$, where n_1 is the number of atoms of species 1 in the unit cell. The remaining species $2,\ldots,K$ are identified with a shift vector $P^k \in \mathbb{R}^{3,n_k}$ yielding a total shift vector

$$\mathbf{P} = [P^1, P^2, \ldots, P^K] \in \mathbb{R}^{3,-1+\Sigma_1^K n_k}.$$

As before (cf. (3)) we define the multi-atomic $\{(k,n_k)\}_{k=1}^K$-lattice by

$$\mathcal{L}(\mathcal{E}) = \mathcal{L}(F,\mathbf{P}) = \bigcup_{i=0}^{n_1-1}(\mathbf{p}_i^1 + \mathcal{L}(F)) \cup \bigcup_{k=2}^{K}\bigcup_{i=1}^{n_k}(\mathbf{p}_i^k + \mathcal{L}(F)),$$

where $\mathbf{p}_0^1 = (0,0,0)$ and each upper index denotes a different atomic species.

C.1. Lattice transformations between multi-atomic lattices

As for transformations between monoatomic lattices we distinguish between transformation between the same numbers of atoms and different numbers of atoms.

Let us start with a transformation between two $\{(k,n_k)\}_{k=1}^K$ lattices generated by $\mathcal{E} = [F,\mathbf{P}]$ and $\Sigma = [G,\mathbf{Q}]$, respectively. Requiring that each atomic species gets mapped to itself and assuming that the atoms of species 1 span the skeletal lattice we arrive at the essential multi-atomic minimization problem

$$\mathcal{I}^1_{r,p,q,\boldsymbol{\alpha}}(\mathcal{E},\Sigma) := \min_{\substack{\mu\in \mathrm{SL}(3,\mathbb{Z}),\\ \pi_k, L_k,\\ k=1,\dots,K}} \left(d_r(H_\mu,\mathbb{I}) + \sum_{k=1}^{K} \alpha^k \|H_\mu P^k \pi^k - (Q^k + GL^k)\|_{p,q} \right). \tag{15}$$

Here, $\boldsymbol{\alpha} = (\alpha^1,\dots,\alpha^K) \in \mathbb{R}_+^K$ are some suitable weights for each atomic species and $H_\mu = G\mu F^{-1}$, $\pi_1 \in A(n_1-1)$, $L_1 \in \mathbb{Z}^{3,n_1-1}$, $\pi_k \in A(n_k)$ and $L_k \in \mathbb{Z}^{3,n_k}$ for $k = 2,\dots,K$. Similarly we define the minimization problem $\mathcal{I}^j_{r,p,q,\boldsymbol{\alpha}}(\mathcal{E},\Sigma)$ for a skeletal lattice generated by the j-th species. The total minimization problem between essential multi-atomic lattices then reads

$$\mathcal{I}_{r,p,q,\boldsymbol{\alpha}}(\mathcal{E},\Sigma) = \min_{j=1,\dots,K} \mathcal{I}^j_{r,p,q,\boldsymbol{\alpha}}(\mathcal{E},\Sigma). \tag{16}$$

Next, we consider the more general case of transformations between $\{(k,n_k)\}_{k=1}^K$ and $\{(k,m_k)\}_{k=1}^K$ multi-atomic lattices. Since no atoms can be created or destroyed during transformation, the ratio of atoms of each species is constant, i.e. $\frac{n_k}{m_k} = \frac{n_j}{m_j}$ for all $j,k = 1,\dots,K$. Setting $c_k = \mathrm{lcm}(n_k,m_k)$ the resulting transformation is between multi-atomic $\{(k,c_k)\}_{k=1}^K$ lattices. For each atomic species, the procedure of finding the lattice vectors of the c_k-lattice is the same as in the case of monoatomic n-lattices and the total minimization problem can be obtained by combining (16) with (8).

C.2. Notes on FORTRAN implementation

TODO

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