# Generating derivative superstructures for systems with high configurational freedom 

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#### Abstract

Modeling alloys requires the exploration of all possible configurations of atoms. Additionally, modeling the thermal properties of materials requires knowledge of the possible ways of displacing the atoms. One solution to finding all symmetrically unique configurations and displacements is to generate the complete list of possible configurations and remove those that are symmetrically equivalent. This approach, however, suffers from a combinatorial explosion when the supercell size is large, when there are more than two atom types, or when there are many displaced atoms. This problem persists even when there are only a relatively small number of unique arrangements that survive the elimination process. Here, we extend an existing algorithm to include the extra configurational degrees of freedom from the inclusion of displacement directions. The algorithm uses group theory and a tree-like data structure to eliminate large classes of configurations, avoiding the typical combinatoric explosion. With this approach we can now enumerate previously inaccessible cases, including atomic displacements.


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## 1. Introduction

In computational material science, one frequently needs to list the "derivative superstructures" [1] of a given lattice. A derivative superstructure is a structure with lattice vectors that are multiples of a "parent lattice" and have atomic basis vectors constructed from the lattice points of the parent lattice. For example, many phases in metal alloys are merely "superstructures" of fcc, bcc, or hcp lattices ( $\mathrm{L} 1_{0}, \mathrm{L1}_{2}, \mathrm{~B} 2, \mathrm{D} 0_{19}$, etc.). When modeling alloys, it is necessary to explore all possible configurations and concentrations of atoms within these superstructures. When determining if a material is thermodynamically stable, the energies of the unique arrangements are compared to determine which has the lowest energy.

Derivative superstructures are found using combinatoric searches [2-8], comparing every possible combination of atoms to determine which are unique. However, these searches can be computationally expensive for systems with high configurational freedom and are sometimes impractical due to the combinatoric explosion of possible arrangements.

Other problems impaired by the inefficiency of current enumeration methods include modeling materials that have disorder in

[^0]their structures, such as site-disordered solids [9] or that include atomic displacements as a degree of freedom [10-12]. There are numerous techniques available for modeling these systems including cluster expansion (CE) [13] and a recently developed "small set of ordered structures" (SSOS) method [14]. However, the accuracy of these methods is still linked to the number of unique configurations being modeled. In other words, if the model is trained on a small set of configurations then it will not be able to make accurate predictions. Increasing the number of configurations used to train the models can improve their predictive power. Increasing the number of structures being used requires a more efficient enumeration technique than those currently available.

Leveraging the basic concepts of the algorithm presented in Ref. [6], we altered the algorithm to have more favorable scaling in multinary cases. The basic idea is to imagine the enumeration as a tree search and employ two new ideas: (1) "partial colorings" and (2) stabilizer subgroups. Section 3 illustrates the algorithm with a concrete example.

The concept of partial colorings is to skip entire branches of the tree that are symmetrically equivalent to previously visited branches. A partial coloring is an intermediate level in the tree (see Fig. 1) where configurations are not yet completely specified. It frequently happens that symmetric redundancy can be identified at an early, "partially colored" stage, avoiding the need to descend further down the tree.


Fig. 1. The empty lattice and 8 of the 36 configurations with only red atoms are shown for the example discussed in Section 3 . Above each partial coloring is a vector that indicates its location in the tree, i.e. $\left(x_{r}, x_{y}, x_{p}\right)$, where the $x_{i}$ s are integers that indicate which arrangement of that color is on the lattice and a $\bullet$ means that no atoms of that color have been placed yet. Below each configuration is either the label of a symmetrically equivalent configuration, along with the group operation that makes them equivalent, or the letters A and B. A and B are the branches that are built from the 1-partial colorings that are unique and are displayed in Fig. 2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Stabilizer subgroups [15] further increase the efficiency of the new algorithm. The stabilizer subgroup at each stage is the set of symmetries which leave the current partial coloring unchanged. As we add more colors, and eliminate symmetrically equivalent colorings, we need not consider colorings which would be equivalent by non-stabilizer symmetries, since those colorings have already been implicitly eliminated. Note that the stabilizer subgroup will get smaller as we proceed down the search tree, thus simplifying and speeding our search.

## 2. Supercell selection and the symmetry group

The first step in enumerating derivative superstructures is the enumeration of unique supercells. This step was solved in Ref. [8], but due to its importance to the algorithm we provide a brief overview.

The supercells, of size $n$, are found by constructing all Hermite Normal Form (HNF) matrices whose determinant is $n$. An HNF matrix is an integer matrix with the following form and relations:

$$
\left(\begin{array}{lll}
a & 0 & 0  \tag{1}\\
b & c & 0 \\
d & e & f
\end{array}\right), 0 \leqslant b<c, 0 \leqslant d<f, e<f
$$

where $a c f=n$. The HNFs determine all possible the supercells for the system. For example, consider a 9 -atom cell, then $n=9$ and $a$, $c, f$ are limited to permutations of $(1,3,3)$ and $(1,1,9)$. Then following the rules for the values of $b, d$, and $e$, every HNF for this system can be constructed. These HNFs represent all the possible supercells of size $n$ of the selected lattice. Some of these are equivalent by symmetry, so the symmetry group of the parent lattice is used to eliminate any duplicates.

Next, we convert the symmetries of the lattice to a list of permutations of atomic sites. There is a one-to-one mapping between the symmetries of the lattice and atomic site permutations, i.e., the groups are isomorphic. The mapping from the symmetry operations to the permutation group is accomplished using the quotient group $G=L / L^{\prime}$, where $L$ is the lattice, constructed from the unit cell, and $L^{\prime}$ is the superlattice, constructed from the supercell. The
quotient group $G$ is found directly from the Smith Normal Form (SNF) matrices, which can be constructed from the HNFs via a standard algorithm using integer row and column operations. Thus $S=U H V$ where $U$ and $V$ are integer matrices with determinant $\pm 1$ and $S$ is the diagonal SNF matrix, where each positive integer diagonal entry divides the next one down. The group, $G$, is then $G=Z_{s_{1}} \oplus Z_{s_{2}} \oplus Z_{s_{3}}$, where $s_{i}$ is $i$ th diagonal of the SNF and $Z_{s_{i}}$ represents the cyclic group of order $n$.

Once the supercells have been found and their symmetry groups have been converted to the isomorphic permutation group, the algorithm can begin finding the unique arrangements of atoms within each supercell in a tree search framework. This is accomplished by treating each supercell with its symmetry group as a separate enumeration problem. The results of the enumeration across all supercells are then combined to produce the full enumeration.

## 3. Tree search

Once a supercell has been selected, the remainder of the enumeration algorithm resembles a tree search. It is often possible to skip the descendents of a node because we know all its "leaves" will represent duplicate structures. These nodes represent incomplete configurations, or partial colorings (see Figs. 1 and 2). The partial colorings are identified using a "location vector" - a list of indices that specify the node in the tree. Once a partial coloring is constructed, the stabilizer subgroup for that partial coloring is found. The stabilizer subgroup allows for the comparison of branches within the tree in a manner that minimizes the number of group operations used. These tools (partial colorings and the stabilizer subgroup) are used to "prune" branches of the tree as they are being constructed, eliminating large classes of arrangements at once (Fig. 3).

We will use a 2D example of a 9-atom cell to illustrate the algorithm. The lattice will be populated with the following atomic species; 2 red atoms, 3 yellow atoms, and 4 purple atoms. A subset of the possible arrangements of this system is shown in Fig. 2. The concepts illustrated with this 2D example are equally applicable in 3D.

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