Abstract. The Voronoi cell of any atom in a lattice is identical. If atoms are perturbed from their lattice coordinates, then the topologies of the Voronoi cells of the atoms will change. We consider the distribution of Voronoi cell topologies in two-dimensional perturbed systems. These systems can be thought of as simple models of finite-temperature crystals. We give analytical results for the distribution of Voronoi topologies of points in two-dimensional Bravais lattices under infinitesimal perturbations and present a discussion with numerical results for finite perturbations.

Keywords: random/ordered microstructures (theory), exact results, stochastic processes (theory), random graphs, networks
1. Introduction

1.1. Motivation

Statistical topology provides a set of tools for studying the statistics of topological properties of a system, much in the way that statistical mechanics focuses on geometric quantities such as velocities and momenta of large sets of particles. Statistical topology has been applied to many classical systems including glasses [1], polymers [2], radial and cellular networks [3, 4], and ideal gases [5]. In this paper we consider the statistical topology of perturbed lattices. In particular, we consider systems in which atoms are initially located at lattice positions and then perturbed by random displacements; such perturbations might be associated with thermal vibrations of the atoms. In two dimensions, the complete topology of each Voronoi cell is given by its number of sides $n$. 

doi:10.1088/1742-5468/2016/04/043103
What is the distribution of $n$? We provide exact results for each of the Bravais lattices and partial results for the honeycomb structure. Our goal is to provide a more complete understanding of realistic systems by providing an analytic point of comparison.

1.1.1. Statement of problem. We begin by considering a system of atoms arranged on a two-dimensional lattice

$$
\Lambda = \{ n_1 \mathbf{v}_1 + n_2 \mathbf{v}_2 \mid n_1, n_2 \in \mathbb{Z} \},
$$

where $\mathbf{v}_1$ and $\mathbf{v}_2$ are linearly independent. A random perturbation of this lattice is obtained by displacing each atom from its lattice position by a random variable. Properties of such systems are considered in [6, 7]. This system can be considered as an approximation for a two-dimensional crystalline solid whose atoms are displaced from lattice positions by thermal noise and small strains. For a given lattice $\Lambda$, we consider $p(n)$, the probability that a randomly chosen atom will have a Voronoi cell with $n$ edges.

In this paper, we consider perturbations which are chosen from a radially symmetric distribution, such that the probability of a particular displacement from a lattice position depends only on its magnitude. One particular example of such a model is the classical Einstein solid, in which atoms are perturbed from lattice positions using a Gaussian distribution [8]. The Gaussian distribution arises from atomic vibrations within a parabolic potential well, where the width of the distribution is proportional to the square root of the temperature. We begin by obtaining analytic results for infinitesimal perturbations and later consider perturbations of finite amplitude.

1.2. Voronoi tessellations

We consider Voronoi tessellations in $\mathbb{R}^2$ endowed with the standard Euclidean metric $d$. Let $S$ be a discrete, possibly infinite, set of atomic coordinates in $\mathbb{R}^2$. Then,

$$
R_s = \{ x \in \mathbb{R}^2 \mid d(x, s) \leq d(x, s') \text{ for all } s' \in S \}
$$

is the Voronoi cell associated with an atom $s \in S$. In other words, the Voronoi tessellation is the partitioning of the plane into regions such that all points in a region are not closer to any other atom than to its own. Note that a point $x$ can be equidistant to multiple atoms and hence belong to multiple Voronoi cells. Figure 1 shows Voronoi tessellations for a square lattice, a perturbed square lattice, and a set of Poisson-distributed atoms. For our purposes, atoms are said to be in general position if no more than three atoms lie on the boundary of an empty circle, i.e. a circle that has no other atoms within it. If atoms are in general position, then no point belongs to more than three distinct cells, and so no point is adjacent to more than three edges; note that in the square lattice this is not the case. In figures 1(b) and (c), exactly three edges meet at any point. In this case, Euler’s formula for planar graphs [9] requires that the average number of edges per cell be exactly six.

1.3. Classification of two dimensional lattices

Two-dimensional lattices are traditionally classified into five groups, known as the Bravais lattices [10] and illustrated in figure 2. This classification is based on the relative magnitudes and orientations of $\mathbf{v}_1$ and $\mathbf{v}_2$. If $\mathbf{v}_1$ and $\mathbf{v}_2$ are orthogonal, then the
lattice is either square if $\|v_1\| = \|v_2\|$ or rectangular otherwise. If the angle between the vectors is $60^\circ$ or $120^\circ$ and $\|v_1\| = \|v_2\|$, then the lattice is hexagonal. For all other angles, the lattice is either rhombic or oblique, depending on whether $(2v_2 - v_1) \cdot v_1 = 0$.

Each lattice has a Voronoi cell which shares the symmetry group of the lattice itself and which can be repeated to tile the plane. The square and rectangular lattices have four-sided cells, while the hexagonal, rhombic, and oblique lattices each have six-sided cells. The Voronoi tessellation of square and rectangular lattices are unstable in the sense that infinitesimal perturbations of the atom positions will change the topology of the Voronoi structure. That is because in these lattices, vertices in the Voronoi tessellation are equidistant to four atoms. In contrast, the hexagonal, rhombic and oblique tessellations are stable, and no more than three edges meet at any vertex.
1.4. Delaunay triangulation

Every Voronoi tessellation has a dual object called a Delaunay diagram. In a two-dimensional Delaunay diagram, edges connect atoms whose Voronoi cells share an edge. If atoms are in general position (i.e. no four lie on the boundary of an empty circle), then the Delaunay diagram will be a triangulation. Figure 3 illustrates a set of atoms, the Voronoi tessellation of the set, and its dual Delaunay diagram.

The Delaunay diagram is a useful tool for computing properties of its dual Voronoi tessellation. A Delaunay triangulation maximizes the minimum interior angle of each triangle [11]. Simple mechanisms for finding the Delaunay triangulation, and thus the Voronoi tessellation of a set of atoms in general position, are the flip algorithms considered in [11]. These algorithms begin by constructing a triangulation in an arbitrary manner. Next, for each pair of adjacent triangles, the edge separating the pair is flipped and it is checked whether this increases the minimal internal angle in those triangles. If this is indeed the case, then the edge flip is retained; otherwise it is reversed. This ensures that the adjacent triangles are locally Delaunay. This process is repeated until no pair of adjacent triangles is non-Delaunay.

Given the two triangulations for a region bounded by four atoms, picking the one that maximizes the minimum internal angle ensures the Delaunay condition is met locally. A simple corollary to this theorem is that if a pair of opposite angles adds to greater than $180^\circ$ on a quadrilateral, then the Delaunay triangles split this pair. In figure 4, it is clear that (a) has a smaller minimum angle than does (b); it is also clear that two opposite angles add to greater than $180^\circ$ in (a) but not in (b). Therefore, figure 4(b) is Delaunay while figure 4(a) is not.

2. Analytic solutions

2.1. Vertex in a square lattice

Before calculating the distribution of Voronoi topologies $p(n)$ for perturbed lattices, we consider how the Voronoi tessellation near a topologically unstable vertex (i.e. a vertex with more than three edges) resolves under small perturbations of the atomic coordinates. We focus first on the Voronoi tessellation of the square lattice illustrated in
In the unperturbed case, some points in the plane are equidistant to four neighboring atoms and therefore lay on the boundaries of four Voronoi cells. After atom positions are randomly perturbed, each unstable Voronoi vertex will resolve into two topologically-stable vertices. Such a resolution can happen in two distinct ways, as illustrated in figures 5(b) and (c). Given the symmetry of the problem, it seems intuitive that each resolution should occur with equal probability. Here we make this intuition precise.

The Delaunay diagram can be used to study properties of its dual, the Voronoi tessellation. In the unperturbed case illustrated in figure 5(a), all internal angles of the Delaunay diagram are $\pi/2$, including the pair $\alpha = \beta$. After perturbations, the Voronoi cell and Delaunay diagram change such that either (b) $\alpha + \beta < \pi$ and a Voronoi edge forms in one direction, or else (c) $\alpha + \beta > \pi$ and a Voronoi edge forms in the other direction. (a) $\alpha + \beta = \pi$. (b) $\alpha + \beta < \pi$. (c) $\alpha + \beta > \pi$.

As noted above, the Delaunay diagram can be used to study properties of its dual, the Voronoi tessellation. In the unperturbed case illustrated in figure 5(a), all internal angles of the Delaunay diagram are $\pi/2$, including the pair $\alpha, \beta$. After a random perturbation, $\alpha + \beta$ will almost surely either decrease or increase, determining whether the unstable vertex resolves in the manner of figures 5(b) or (c), respectively. Determining the probability of obtaining these configurations thus requires calculating the probability with which $\alpha + \beta$ increases or decreases under a random perturbation.

In the remainder of the paper we denote a spatial configuration of $n$ atoms in $\mathbb{R}^2$ as

$$X = \left\{ \left[ \begin{array}{c} x_1 \\ y_1 \\ \vdots \\ x_n \\ y_n \end{array} \right] \right\} \in \mathbb{R}^{2n},$$

such that the initial, unperturbed configuration of four atoms on a square lattice is
\[ X = \left( \begin{bmatrix} 0 & 1 & 1 & 0 \\ 0 & 0 & 1 & 1 \end{bmatrix} \right) \] (4)

We use \( \phi(X) \) to denote the sum of the angles \( \alpha \) and \( \beta \) illustrated in figure 5(a). We apply a random perturbation to \( \phi \), namely \( \xi \in \mathbb{R}^2 \), where \( \xi \) is chosen from any radially symmetric probability distribution. As \( \phi \) changes smoothly with atomic coordinates, the small perturbation \( \xi \) can be linearized as \( \nabla \phi \cdot \xi \), such that \( \phi(X + \xi) \approx \phi(X) + \nabla \phi \cdot \xi \).

In general, the set of perturbations \( \xi \) that satisfy \( \nabla \phi \cdot \xi = 0 \) can be represented as a hyperplane in \( \mathbb{R}^2 \) passing through the origin. For perturbations \( \xi \) on one side of this hyperplane we have \( \nabla \phi \cdot \xi > 0 \), while for those on the other side we have \( \nabla \phi \cdot \xi < 0 \). For the configuration to resolve as shown in figure 5(b), it must be that \( \phi(X + \xi) > \pi \) or, equivalently, \( \nabla \phi \cdot \xi > 0 \). If \( \nabla \phi \cdot \xi < 0 \), the initial configuration will resolve as shown in figure 5(c). Since the distribution of perturbations is symmetric about the origin, the integral of the probability measure over the region for which \( \nabla \phi \cdot \xi > 0 \) is 0.5; this is also the integral over the region for which \( \nabla \phi \cdot \xi < 0 \). This demonstrates that the topological resolutions illustrated in figures 5(b) and (c) indeed occur with equal probability.

In this example, the probabilities depend on the fractional volume of space on either side of a single hyperplane. We next consider probabilities of more complex configurations, the calculations of which require consideration of fractional volumes associated with multiple intersecting half-spaces.

2.2. Nearby vertices in a square lattice

Another example illustrates the complexities that can arise from correlations between neighboring unstable vertices. Figure 6 illustrates two nearby, unstable vertices in a Voronoi tessellation of a square lattice. The manner in which these resolve is determined by the motions of seven nearby atoms, and how the labeled angles \( \alpha_i \) and \( \beta_i \) change under perturbations.

Before perturbation \( \alpha_i = \beta_i = \pi/2 \) for all \( i \), and the two vertices of the Voronoi tessellation are unstable. After perturbation, if \( \alpha_i + \beta_i > \pi \) a new edge will form between the two atoms, as illustrated in figure 5(c). As shown above, the probability of \( \alpha_i + \beta_i > \pi \) is 0.5. What is the joint probability that both \( \alpha_1 + \beta_1 \) and \( \alpha_2 + \beta_2 \) will increase under a random perturbation?

As before, we denote a configuration of 7 atoms as a vector \( X \in \mathbb{R}^2 \), where \( n = 7 \); we let \( \phi_i(X) = \alpha_i + \beta_i \). Each of \( \nabla \phi_i \cdot \xi = 0 \) determines a hyperplane through the origin in \( \mathbb{R}^2 \), and the two intersecting hyperplanes divide \( \mathbb{R}^2 \) into four regions, each corresponding to one of four possible topologies that result from the infinitesimal perturbations of the atoms, as illustrated in figure 6(b). If we let the initial, unperturbed configuration space of atoms be:

\[ X = \left( \begin{bmatrix} 0 & -1 & 1 & 0 \\ 2 & 1 & 0 & -1 \end{bmatrix} \right) \]

then the gradients \( \nabla \phi_i \) are:

\[ \nabla \phi_1 = \left( \begin{bmatrix} 0 & 1 & 1 & 0 \\ 1 & 0 & -1 & 0 \end{bmatrix} \right) \]

(5)

(6)

doi:10.1088/1742-5468/2016/04/043103
The two hyperplanes defined by these gradient vectors intersect at an angle
\[ \arccos \left( \frac{1}{4} \right) \approx 104.48^\circ \]; this angle does not depend on the choice of coordinates.

Since the perturbations are chosen from a probability distribution that is radially symmetric about the origin, the probabilities of the configuration resolving into each of the four possibilities are equal to the fractional volumes of the four regions separated by the intersecting planes (see figure 7). These fractional volumes can be determined through consideration of the angles at which these two planes intersect.

In the case considered here, the angle between \( \nabla \phi_1 \) and \( \nabla \phi_2 \) is \( \theta = \arccos(-1/4) \approx 104.48^\circ \); this angle does not depend on the choice of coordinates.

Since the perturbations are chosen from a probability distribution that is radially symmetric about the origin, the probabilities of the configuration resolving into each of the four possibilities are equal to the fractional volumes of the four regions separated by the intersecting planes (see figure 7). These fractional volumes can be determined through consideration of the angles at which these two planes intersect.

In the case considered here, the angle between \( \nabla \phi_1 \) and \( \nabla \phi_2 \) is \( \theta = \arccos(-1/4) \), and so the angle subtended by the region where \( \nabla \phi_1 \cdot \xi > 0 \) and \( \nabla \phi_2 \cdot \xi < 0 \) is \( \pi - \theta \). The fractional volume of this region is then \( (\pi - \theta)/2\pi \approx 0.98\% \). The fractional volume of the region where \( \nabla \phi_1 \cdot \xi < 0 \) and \( \nabla \phi_2 \cdot \xi < 0 \) is the same (see figure 7). Hence, the probability of the initial configuration resolving into the configuration shown in figure 6(b) or (d) is \((\pi - \theta)/2\pi\) for each. By the same reasoning, the probability of the initial configuration resolving into the configuration shown in figure 6(c) is \( \theta/2\pi \approx 29.02\% \).

2.3. Square lattice

The above approach allows us to compute \( p(n) \)—the probability that the Voronoi cell of a randomly chosen atom has \( n \) edges—in a perturbed square lattice. Figure 8 illustrates a central atom in a square lattice and its eight nearest neighbors. The resolution of all four unstable vertices in the Voronoi tessellation can be determined by considering how the sums of angles \( \phi_i = \alpha_i + \beta_i \) change under perturbation. Following

\[
\phi_1 = \alpha_1 + \beta_1 \\
\phi_2 = \alpha_2 + \beta_2
\]

\[
\nabla \phi_1 \cdot \xi > 0 \\
\nabla \phi_2 \cdot \xi > 0
\]

\[
\nabla \phi_1 \cdot \xi > 0 \\
\nabla \phi_2 \cdot \xi < 0
\]

\[
\nabla \phi_1 \cdot \xi < 0 \\
\nabla \phi_2 \cdot \xi < 0
\]

Figure 6. (a) Prior to perturbation, all internal angles of the Delaunay diagram are \( \pi/2 \), and the two vertices of the Voronoi tessellation are unstable. After perturbation, each vertex will resolve in a manner that depends on \( \alpha_i + \beta_i \). (b)–(d) show how the topology of the Delaunay diagram resolves depending on the signs of \( \nabla \phi_1 \cdot \xi \) and \( \nabla \phi_2 \cdot \xi \). Flipping the signs in (c) leads to the remaining resolution, which is identical to (c) up to rotation by \( 180^\circ \). (a) \( \phi_1 = \alpha_1 + \beta_1, \phi_2 = \alpha_1 + \beta_2 \). (b) \( \nabla \phi_1 \cdot \xi > 0, \nabla \phi_2 \cdot \xi > 0 \). (c) \( \nabla \phi_1 \cdot \xi > 0, \nabla \phi_2 \cdot \xi < 0 \). (d) \( \nabla \phi_1 \cdot \xi < 0, \nabla \phi_2 \cdot \xi < 0 \).

and

\[
\nabla \phi_2 = \begin{bmatrix} 0 & 0 \\ 0 & 1 \\ -1 & 0 \\ -1 & -1 \end{bmatrix}
\]

The two hyperplanes defined by these gradient vectors intersect at an angle
\[ \theta = \frac{\pi}{2} \approx 90^\circ \]; this angle does not depend on the choice of coordinates.

Since the perturbations are chosen from a probability distribution that is radially symmetric about the origin, the probabilities of the configuration resolving into each of the four possibilities are equal to the fractional volumes of the four regions separated by the intersecting planes (see figure 8). These fractional volumes can be determined through consideration of the angles at which these two planes intersect.

In the case considered here, the angle between \( \nabla \phi_1 \) and \( \nabla \phi_2 \) is \( \theta = \frac{\pi}{2} \), and so the angle subtended by the region where \( \nabla \phi_1 \cdot \xi > 0 \) and \( \nabla \phi_2 \cdot \xi < 0 \) is \( \pi - \theta \). The fractional volume of this region is then \( (\pi - \theta)/2\pi \approx 0.98\% \). The fractional volume of the region where \( \nabla \phi_1 \cdot \xi < 0 \) and \( \nabla \phi_2 \cdot \xi < 0 \) is the same (see figure 8). Hence, the probability of the initial configuration resolving into the configuration shown in figure 6(b) or (d) is \((\pi - \theta)/2\pi\) for each. By the same reasoning, the probability of the initial configuration resolving into the configuration shown in figure 6(c) is \( \theta/2\pi \approx 29.02\% \).

2.3. Square lattice

The above approach allows us to compute \( p(n) \)—the probability that the Voronoi cell of a randomly chosen atom has \( n \) edges—in a perturbed square lattice. Figure 8 illustrates a central atom in a square lattice and its eight nearest neighbors. The resolution of all four unstable vertices in the Voronoi tessellation can be determined by considering how the sums of angles \( \phi_i = \alpha_i + \beta_i \) change under perturbation. Following
the approach above, the resolution of the entire configuration under a perturbation $\xi$ is determined by the signs of $\nabla \phi_i \cdot \xi$.

The resolution of each unstable vertex can be described by a pair of half-spaces in $\mathbb{R}^{2n}$ with $n = 9$; in one half space $\nabla \phi_i \cdot \xi > 0$, and in the other $\nabla \phi_i \cdot \xi < 0$. The way in which all four unstable vertices resolve is then described by the intersection of four of these half-spaces—one for each pair. Under a random perturbation $\xi$, the Voronoi cell of a central atom gains one edge for each $i$ for which $\nabla \phi_i \cdot \xi > 0$. The probability that the Voronoi cell of an atom in a square lattice resolves with 8 edges is the fractional size of the region in $\mathbb{R}^8$ in which $\nabla \phi_i \cdot \xi > 0$ for all $i$. The probability that a Voronoi cell resolves with 4 edges is equal to the fractional size of the region in $\mathbb{R}^8$ in which $\nabla \phi_i \cdot \xi < 0$ for all $i$.

If the initial, unperturbed configuration of the atoms is
\[ X = \begin{pmatrix} -1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ 1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ -1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ -1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \end{pmatrix} \]  \tag{8}

the gradients \( \nabla \phi_i \) are

\[ \nabla \phi_1 = \begin{pmatrix} -1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ 1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ -1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ -1 & 0 & 1 & -1 & 0 & 1 & -1 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \end{pmatrix} \]  \tag{9}

\[ \nabla \phi_2 = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \end{pmatrix} \]  \tag{10}

\[ \nabla \phi_3 = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 1 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 & 1 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 & 1 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 & 1 & 0 & -1 & 0 \end{pmatrix} \]  \tag{11}

\[ \nabla \phi_4 = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 1 & 0 & 0 \end{pmatrix} \]  \tag{12}

By consideration of their dot products, it is straightforward to see that \( \nabla \phi_1 \) is orthogonal to \( \nabla \phi_2 \) and \( \nabla \phi_4 \), though not to \( \nabla \phi_3 \). In fact, each \( \nabla \phi_i \) is orthogonal to the \( \nabla \phi_i \) associated with its two nearest neighbors. For any two orthogonal vectors \( w_1, w_2 \) and a random perturbation \( \xi \) chosen from a radially symmetric distribution, \( P[w_1 \cdot \xi > 0, w_2 \cdot \xi > 0] = P[w_1 \cdot \xi > 0] P[w_2 \cdot \xi > 0] \). This follows from the fact that a radially symmetric distribution is invariant under rotation. Therefore, we can compute the joint probability \( P[\nabla \phi_1 \cdot \xi > 0, \nabla \phi_3 \cdot \xi > 0] \) independently of the joint probability \( P[\nabla \phi_2 \cdot \xi > 0, \nabla \phi_4 \cdot \xi > 0] \), and so the probability of a Voronoi cell resolving with 8 edges is given by:

\[ p(8) = P[\nabla \phi_1 \cdot \xi > 0] P[\nabla \phi_2 \cdot \xi > 0, \nabla \phi_3 \cdot \xi > 0] P[\nabla \phi_2 \cdot \xi > 0, \nabla \phi_3 \cdot \xi > 0]. \]  \tag{13}

Since we have shown (section 2.2) that \( P[\nabla \phi_1 \cdot \xi > 0, \nabla \phi_3 \cdot \xi > 0] = P[\nabla \phi_2 \cdot \xi > 0, \nabla \phi_4 \cdot \xi > 0] = (\pi - \theta)/2\pi \), the joint probability of \( \nabla \phi_i \cdot \xi > 0 \) for all \( i \) is \( [(\pi - \theta)/2\pi]^2 \approx 4.40\% \). Because the probability distribution of the perturbations is radially symmetric, this is also the probability \( p(4) \). This is consistent with numerical results reported below in section 3.

We determine \( p(5) \) and \( p(7) \) in a similar manner. For the Voronoi cell to resolve with five edges, one vertex must resolve to gain an edge and all remaining vertices must resolve so that no other edge is gained. This can occur in four ways, one for each vertex that can gain an edge; the sum of the probabilities of these four events is:

\[ p(5) = P[\nabla \phi_1 \cdot \xi < 0, \nabla \phi_3 \cdot \xi < 0, \nabla \phi_3 \cdot \xi < 0, \nabla \phi_4 \cdot \xi < 0] \]
\[ + P[\nabla \phi_1 \cdot \xi > 0, \nabla \phi_3 \cdot \xi < 0, \nabla \phi_3 \cdot \xi < 0, \nabla \phi_4 \cdot \xi < 0] \]
\[ + P[\nabla \phi_1 \cdot \xi < 0, \nabla \phi_3 \cdot \xi > 0, \nabla \phi_3 \cdot \xi < 0, \nabla \phi_4 \cdot \xi < 0] \]
\[ + P[\nabla \phi_1 \cdot \xi < 0, \nabla \phi_3 \cdot \xi < 0, \nabla \phi_3 \cdot \xi < 0, \nabla \phi_4 \cdot \xi < 0] \]  \tag{14}
\[
\frac{\theta(\pi - \theta)}{\pi^2} \approx 24.35\%.
\]

Noting that the probability of two opposite sides both gaining an edge is the same as both not gaining an edge, clearly \( p(5) = p(7) \). We can calculate \( p(6) \) by noting that we have already determined every other case (since it must resolve so that the central atom has 4 to 8 edges); i.e. \( p(6) = 1 - p(4) - p(5) - p(7) - p(8) = \frac{1}{2} - \frac{4}{\pi} + \frac{3}{2}\left(\frac{\pi}{2}\right)^2 \approx 42.49\% \).

2.4. Rectangular lattice

Our approach for the square lattice naturally leads to results for rectangular lattices. We can describe the aspect ratio of a rectangular lattice as \( y = \|\mathbf{v}_1\|/\|\mathbf{v}_2\| \), where \( \mathbf{v}_1 \) and \( \mathbf{v}_2 \) are basis vectors for the lattice as described in section 1.3. Following the conventions and definitions for the square lattice, if the initial positions of the atoms are given by the configuration

\[
X = \left( \begin{bmatrix} -1 & \ldots & 0 \\ y & \ldots & 1 \end{bmatrix}, \begin{bmatrix} 0 & \ldots & 1 \\ 0 & \ldots & -1 \end{bmatrix} \right)
\]

then the gradients \( \nabla \phi_i \) are

\[
\nabla \phi_1 = \left( \begin{bmatrix} 1/y & \ldots & 0 \\ -1 & \ldots & 1 \end{bmatrix}, \begin{bmatrix} 0 & \ldots & 1 \\ 0 & \ldots & -1 \end{bmatrix} \right)
\]

\[
\nabla \phi_2 = \left( \begin{bmatrix} 0 & \ldots & -1/y \\ 0 & \ldots & -1 \end{bmatrix}, \begin{bmatrix} 0 & \ldots & 1/y \end{bmatrix} \right)
\]

\[
\nabla \phi_3 = \left( \begin{bmatrix} 0 & \ldots & 0 \\ 0 & \ldots & 1 \end{bmatrix}, \begin{bmatrix} 0 & \ldots & -1/y \end{bmatrix} \right)
\]

\[
\nabla \phi_4 = \left( \begin{bmatrix} 0 & \ldots & 0 \\ 0 & \ldots & -1 \end{bmatrix}, \begin{bmatrix} 0 & \ldots & 1/y \end{bmatrix} \right)
\]

Under a random perturbation \( \xi \), the rectangular Voronoi cell of a central atom will gain an edge for each \( i \) for which \( \nabla \phi_i \cdot \xi > 0 \), in a manner similar to that illustrated in figure 5. Determining the distribution of Voronoi topologies \( p(n) \) of a perturbed rectangular lattice requires computing the fractional size of the region in which \( \nabla \phi_i \cdot \xi \) have particular signs. In the general case, this fractional volume is equivalent to the solid angle subtended by the region, divided by the surface area of the unit \( n \)-sphere.

No closed-form expression exists to calculate the solid angle bounded by \( n \) linearly-independent vectors when \( n > 3 \) [12]. Nevertheless, several approaches are available to numerically compute this solid angle [13]. In this paper, we compute the probabilities \( p(n) \) for each \( n \) by numerically integrating the Gaussian function over the intersection of the four half spaces. Although any radially symmetric probability function could have been used in the integration, the Gaussian was selected due to its well-understood
Figure 9. Numerical integration results for an infinitesimal perturbation as a function of rectangular lattice aspect ratio, \( y \). When the aspect ratio is one, the result is the square lattice. Large circles indicate \( p(n) \) for aspect ratio 1, and also for asymptotic values as the aspect ratio approaches infinity.

It is interesting to inquire how \( p(n) \) behaves as \( y \) approaches 0 or \( \infty \). Limit analysis of the integral equations, combined with numerical integration, shows that

\[
p(4) = 5.46\%, \quad p(5) = 20.09\%, \quad p(6) = 48.90\%, \quad p(7) = 20.09\%, \quad p(8) = 5.46\%
\]

as \( y \to \infty \) or \( y \to 0 \) (see figure 9).

2.5. Vertex in a honeycomb structure

We next focus on the honeycomb structure shown in figure 2(f); we note that this is not a Bravais lattice, since there are atoms not equivalent by translation. In the center of the hexagonal arrangement of atoms is an unstable Voronoi vertex which is adjacent to six edges. With probability one, the original unstable vertex resolves into three stable vertices, each of which is adjacent to three edges. Such a resolution can occur in one of three ways, as illustrated in figures 10(a)–(c).

To compute the probabilities of each of these resolutions, we consider the initial configuration of atoms

\[
X = \left[ \begin{array}{c}
\frac{1}{2} \\
\frac{\sqrt{3}}{2}
\end{array} \right] \left[ \begin{array}{c}
-\frac{1}{2} \\
-\frac{\sqrt{3}}{2}
\end{array} \right] \left[ \begin{array}{c}
-1 \\
0
\end{array} \right] \left[ \begin{array}{c}
-\frac{1}{2} \\
-\frac{\sqrt{3}}{2}
\end{array} \right] \left[ \begin{array}{c}
\frac{1}{2} \\
-\frac{\sqrt{3}}{2}
\end{array} \right] \left[ \begin{array}{c}
1
\end{array} \right]
\]

(21)

and the corresponding gradients \( \nabla \phi_i \)

\[
\nabla \phi_1 = \left[ \begin{array}{c}
\frac{\sqrt{3}}{4} \\
\frac{3}{4}
\end{array} \right] \left[ \begin{array}{c}
0 \\
0
\end{array} \right] \left[ \begin{array}{c}
\frac{\sqrt{3}}{4} \\
\frac{3}{4}
\end{array} \right] \left[ \begin{array}{c}
\frac{\sqrt{3}}{2} \\
\frac{3}{2}
\end{array} \right] \left[ \begin{array}{c}
-\sqrt{3}
\end{array} \right]
\]

(22)

\[
\nabla \phi_2 = \left[ \begin{array}{c}
1 \\
2\sqrt{3}
\end{array} \right] \left[ \begin{array}{c}
0 \\
0
\end{array} \right] \left[ \begin{array}{c}
2/\sqrt{3} \\
0
\end{array} \right] \left[ \begin{array}{c}
-\sqrt{3}/2 \\
-3/2
\end{array} \right] \left[ \begin{array}{c}
-1/\sqrt{3} \\
1
\end{array} \right] \left[ \begin{array}{c}
0
\end{array} \right]
\]

(23)
where $\phi = \alpha + \beta$, for a resolution of the initial configuration as shown in figure 2(f).

Each of the resolutions shown in figures 10(a)–(c) has a triplet of gradient vectors associated with it that determine the probability of the initial configuration resolving as shown. The Voronoi topology resulting from a perturbation can be determined through examination of the Delaunay diagrams. Each topological resolution corresponds to a distinct Delaunay triangulation, the topology of which depends on satisfying a set of inequalities in terms of internal angles formed by the six neighboring atoms.

For example, in order for the unstable vertex to resolve in the manner illustrated in figure 10(a), it is necessary that $\alpha + \beta > \pi$ for all $i$; this occurs when $\phi \cdot \nabla > 0$ for all $i$. The probability of obtaining this resolution is equal to the fractional region of space of three intersecting half spaces, each described by $\phi \cdot \nabla > 0$. We can compute this fractional region by considering $\Omega$, the solid angle subtended by this region of space. For the configuration described by the Voronoi tessellation in figure 10(a) and the Delaunay diagram in figure 10(d), we compute $\Omega = 2 \tan^{-1}(\sqrt{6} - 2)$. We then have:

$$P[\nabla \phi > 0] = \frac{\Omega}{4\pi} = \frac{2 \tan^{-1}(\sqrt{6} - 2)}{4\pi} \approx 0.067,$$

(25)

since the fractional volume of a three-dimensional wedge is its solid angle divided by the surface area of a three-dimensional sphere. Accounting for symmetries, figure 10(a) can be obtained in six equivalent ways (obtained by $60^\circ$ rotations), such that the probability of obtaining this topology is $6 \times 0.067 \approx 0.4034\%$. Similar calculations show that the probability of obtaining the topology in figure 10(b) is $\frac{3}{\pi} \tan^{-1}\left(\frac{2\sqrt{6} - 3}{5}\right) \approx 34.66\%$ and in figure 10(c) is $1/4$. 

Figure 10. (a)–(c) Resolutions of unstable vertex in the honeycomb structure; (d)–(f) corresponding Delaunay diagrams.
2.6. Hexagonal, oblique, and rhombic lattices

For infinitesimal perturbations, the hexagonal, oblique and rhombic lattices are all stable, since for any empty quadrilateral, opposite angles never sum to \( \pi \); the Delaunay condition (section 1.4) shows that these are all stable triangulations. With probability 1, an infinitesimal perturbation cannot change this; in the limit that the magnitude of the random perturbation goes to zero, there will be a point at which the linearization \( |\nabla \cdot \xi| < \gamma \) where \( \gamma \) is the difference from the original sum of opposite angles and \( \pi \). Therefore, in this limit, these structures are stable. As an example, consider the hexagonal lattice from figure 2(f), where any empty quadrilateral has a pair of opposite angles that sum to \( 2\pi/3 \) and thus are stable for all small perturbations.

Table 1 summarizes the analytic results from section 1, as well as the known Poisson–Voronoi distribution.

| Lattice          | p(3)   | p(4)   | p(5)   | p(6)   | p(7)   | p(8)   | p(9)   |
|------------------|--------|--------|--------|--------|--------|--------|--------|
| Square (exact)   | 0      | 0.2435 | 0.4249 | 0.2435 | 0.440  | 0      |
| Square (approx.) | 0.0440 | 0.2435 | 0.4249 | 0.2435 | 0.440  | 0      |
| Rectangular (3)  | 0.0499 | 0.2198 | 0.4605 | 0.2197 | 0.499  | 0      |
| Rectangular (10) | 0.0539 | 0.2033 | 0.4856 | 0.2033 | 0.539  | 0      |
| Rectangular (∞)  | 0.0546 | 0.2009 | 0.4888 | 0.2009 | 0.546  | 0      |
| Hexagonal        | 0      | 0      | 1      | 0      | 0      | 0      |
| Rhombic          | 0      | 0      | 1      | 0      | 0      | 0      |
| Oblique          | 0      | 0      | 1      | 0      | 0      | 0      |
| Poisson          | 0.0112 | 0.1068 | 0.2595 | 0.2947 | 0.1987 | 0.0897 | 0.0295 |

Note: As explained in section 2.2, \( \theta = \arccos(-1/4) \).

3. Simulations

While we reported above on the effects of infinitesimal perturbations on the topological distributions of Voronoi cells, we now examine the impact of the perturbation amplitude on the same distributions. For this purpose, we displace each atom \( i \) from its initial lattice position \( x_i \) by a two-dimensional independent random variable \( \Delta x_i \), chosen from a Gaussian distribution centered at the origin, and described by the probability density function

\[
f(\Delta x_i) = \frac{1}{2\pi\sigma^2} e^{-|\Delta x_i|^2/2\sigma^2};
\]

the new position of each atom after the perturbation is \( x_i' = x_i + \Delta x_i \).

We consider how the distribution of Voronoi topologies \( p(n) \) in the perturbed system varies with the width of the Gaussian \( \sigma \). When \( \sigma \) is small, the results obtained in section 1 should be closely reproduced. When \( \sigma \) is large, \( p(n) \) approaches the Poisson–Voronoi
distribution, as the distribution of atoms within any region approaches a Poisson distribution. The Poisson–Voronoi distribution is known analytically in integral form \[14\] and has been evaluated numerically and through simulations \[15\]. Some geometric and topological features of random perturbations of the square, hexagonal, and honeycomb structures have been studied numerically in \[16\].

3.1. Simulation design

All simulations were designed in Matlab using the standard Voronoi package and random number generator. A grid of atoms for a lattice structure was initialized and each atom was displaced from its lattice position by choosing random displacements in \(x\) and \(y\) according to a Gaussian distribution with mean \(\mu = 0\) and standard deviation \(\sigma\). The standard deviation \(\sigma\) was measured in units of the square root of the unit cell area: \(\sqrt{||v_1 \times v_2||}\).

Each simulation considered a system of at least one million atoms; periodic boundary conditions were used to minimize finite-size effects. The simulations recorded the fraction of Voronoi cells with a particular topology.

3.2. Square lattice

Figure 11 shows the distribution of Voronoi topologies \(p(n)\) of a perturbed square lattice for \(0 < \sigma \leq 1\), obtained through numerical simulation as described above. Simulation data for small \(\sigma\) are consistent with the analytical results reported in section 2.3. The symmetry of the analytic result is quickly broken as \(\sigma\) increases. Whereas \(p(5) = p(7)\) and \(p(4) = p(8)\) in the limit as \(\sigma\) approaches 0, for finite \(\sigma\), this is not the case. Moreover, where \(p(n) = 0\) for \(n = 3\) and \(n > 8\) in the small \(\sigma\) limit, these numbers become positive for finite \(\sigma\). As \(\sigma\) increases, \(p(n)\) evolves monotonically toward the Poisson–Voronoi distribution except for \(p(4)\) which has an inflection point near \(\sigma = 0.3\). The fraction of Voronoi cells with \(n \geq 8\) edges increases rapidly between \(\sigma = 0.2\) and \(\sigma = 0.4\), and then levels off by \(\sigma = 0.5\). By \(\sigma = 1\), it is difficult to distinguish the distribution \(p(n)\) from the Poisson–Voronoi distribution.

3.3. Rectangular lattice

Figure 12 shows the distribution of Voronoi topologies \(p(n)\) of rectangular lattices of aspect ratio three and ten respectively for \(0 < \sigma \leq 1\). For low \(\sigma\), the numerical results are consistent with the analytic results obtained for infinitesimal perturbations. Qualitatively, the Voronoi cells with 4, 6, and 8 edges are favored more heavily than they are in the square lattice. Voronoi cells with 5 and 7 edges become more frequent until an abrupt change around \(\sigma = 0.2\) for aspect ratio three and \(\sigma = 0.1\) for aspect ratio ten. While \(p(n)\) is monotonic for most \(n\) in the square lattice, \(p(n)\) is not monotonic for most \(n\) in the rectangle lattice at aspect ratio ten. As the aspect ratio increases, the rectangle lattices show more variation of \(p(n)\) with changing \(\sigma\). For example, there is a sharp peak of five-sided Voronoi cells near \(\sigma = 0.05\). Furthermore, notice that for \(\sigma\) between roughly 0.25 and 0.50, there are more five-sided Voronoi cells than any other. This might be contrasted with the cases of the square, hexagonal, and honeycomb...
Statistical topology of perturbed two-dimensional lattices

The convergence to the Poisson–Voronoi distribution is also slower as the aspect ratio increases.

3.4. Hexagonal lattice

Figure 13 shows the distribution of Voronoi topologies \( p(n) \) for the hexagonal lattice, obtained through numerical simulation, as a function of \( \sigma \). The distribution \( p(n) \) is relatively insensitive to changes of \( \sigma \) at both low and high \( \sigma \) limits. The switch between these limits is abrupt; it occurs largely within the limits \( 0.1 < \sigma < 0.3 \). As a result, the distribution \( p(n) \) varies monotonically for most \( n \) between the small and large \( \sigma \) limits.

3.5. Honeycomb structure

Figure 14 shows the distribution of Voronoi topologies of the honeycomb structure for \( 0 < \sigma \leq 1 \) as obtained through numerical simulation. The variation of \( p(n) \) with \( \sigma \) is ‘gentle’ relative to the lattices discussed above. This might result from the similarity between the low- and high-\( \sigma \) frequencies \( p(n) \). In the honeycomb structure, even as \( \sigma \)
approaches 0 there is a large diversity of topological types, with non-zero frequencies for $n = 3$ and $9 \leq n \leq 12$; in contrast, for other lattices these frequencies are all zero as $\sigma$ approaches 0.

4. Conclusions

In this paper, we explored how perturbations affect the distribution of Voronoi cell topologies for the two-dimensional Bravais lattices and the honeycomb structure. Such results are important for describing crystalline materials at finite temperature. Of particular interest are those structures for which the zero-temperature Voronoi topology distributions are unstable with respect to infinitesimal perturbations. In applications to crystalline materials, it is the small-perturbation limit that is of most interest at temperatures well below the melting point. Even approaching the melting temperature, $\sigma$ rarely exceeds 0.05–0.15 [17].

Figure 13. Frequencies of $n$-sided Voronoi cells for $0 < \sigma \leq 1$ for the hexagonal lattice; large circles indicate asymptotic values.

Figure 14. Frequencies of $n$-sided Voronoi cells for $0 < \sigma \leq 1$ for the honeycomb structure; large circles indicate asymptotic values.
Voronoi cell descriptions of the local structure have been growing in importance for material characterization, both for atomic-scale simulations and in experiment. Although here we focus on two-dimensional lattices, the same methods can be applied in three dimensions, where most materials and structures live. Indeed a similar approach has been used in recent work [18] to characterize disorder in three-dimensional atomic systems. A combinatorial approach for calculating distributions of topological types in three-dimensional perturbed lattices is suggested in [19]. That approach, however, does not account for pairwise correlation between distinct topological resolutions, instead treating each as an independent event. This simplification leads to large differences between predicted and observed data in that work. Some geometric and topological features of random perturbations of face-centered cubic crystals have been studied numerically in [20]. When perturbations become large relative to the lattice constant, statistics of such systems approach those of an ideal gas; geometric and topological features of the ideal gas have been considered at length in [5].

While other papers have focused on topological distributions for Poisson distributed points, our paper considers the topology in systems which have much more order, but are not strictly periodic. The most significant achievement in this paper is the summary of exact results for the distribution of Voronoi topologies of all two-dimensional Bravais lattices under random infinitesimal perturbations chosen from any radially symmetric distribution (see table 1). Because we present numerical data for finite perturbations, these results also show how the topology transitions from that of a perfect lattice to a perfectly disordered (Poisson) system. While the present study focuses on uncorrelated perturbations in the Einstein-sense, it would be of interest to see how accurate this description is for realistic atomic systems in which vibrations are correlated, as described by the phonon spectra.

References

[1] Rivier N 1983 Helv. Phys. Acta 56 307
[2] Orlandini E and Whittington S G 2007 Rev. Mod. Phys. 79 611
[3] Seong R K, Salaﬁa C M and Vvedensky D D 2012 Phil. Mag. 92 230
[4] Mason J K, Lazar E A, MacPherson R D and Srolovitz D J 2012 Phys. Rev. E 85 051128
[5] Lazar E A, Mason J K, MacPherson R D and Srolovitz D J 2013 Phys. Rev. E 88 063309
[6] Holroyd A E and Soo T 2013 Electron. J. Probab. 18 1
[7] Peres Y and Sly A 2014 arXiv:1409.4490
[8] Kittel C and McEuen P 1976 Introduction to Solid State Physics vol 8 (New York: Wiley)
[9] West D B et al 2001 Introduction to Graph Theory vol 2 (Upper Saddle River, NJ: Prentice Hall)
[10] Bravais A and de Beaumont L É 1866 Études Cristallographiques: Mémoire sur les Systèmes Formés par des Points Distribués Régulièrement sur un Plan ou dans l’Espace (Paris: Gauthiers-Villars)
[11] De Berg M, Van Kreveld M, Overmars M and Schwarzkopf O C 2000 Computational Geometry (New York: Springer)
[12] Murakami J and Yano M 2005 Commun. Anal. Geom. 13 379
[13] Ribando J M 2006 Discrete Comput. Geom. 36 479
[14] Calka P 2003 Adv. Appl. Probab. 35 863
[15] Brakke K 1986 200000000 Random Voronoi Polygons
[16] Lucarini V 2008 J. Stat. Phys. 130 1047
[17] Lawson A C 2009 Phil. Mag. 89 1757
[18] Lazar E A, Han J and Srolovitz D J 2015 Proc. Natl Acad. Sci. 112 E5760
[19] Troadec J, Gervois A and Oger L 1998 Europhys. Lett. 42 167
[20] Lucarini V 2009 J. Stat. Phys. 134 185

doi:10.1088/1742-5468/2016/04/043103