Flexible Crystal Frameworks

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Abstract

Building upon and complementing recent results on the rigidity theory of periodic bar-and-joint frameworks, this paper studies tetrahedral structures modeled on specific crystalline materials: quartz, cristobalite and tridymite. The general theory predicts at least three infinitesimal degrees-of-freedom. Here, we investigate the actual deformations of these structures. We show that quartz and cristobalite have smooth three-dimensional configuration spaces, but ideal high tridymite is a singular configuration with a six-dimensional tangent space. The topology around this singularity is explicitly described.

1 Introduction

Motivated by questions arising in mathematical crystallography and computational materials science, we present in this paper specific geometric applications of the general theory of rigidity and flexibility for periodic frameworks developed in our recent papers [1, 4, 5, 3].

Molecules as mechanical frameworks. At a certain approximation level, many molecules can be modeled as mechanical frameworks, with rigid, fixed-length bonds between particular pairs of atoms and fixed-angles between particular adjacent bonds. This opens the possibility of using techniques from rigidity theory in molecular flexibility analysis. Considerations related to *framework flexibility* appear already in the early 20th century structural investigations based on X-ray crystallography, see e.g. [6, 8, 16]. Geometric models of deforming frameworks have been used in studying *displacive phase transitions* in materials [7].

Rigidity and flexibility of crystalline materials. Most of the molecular flexibility studies rely on computationally intensive physics-based simulations or simplified, kinematics-based methods [20, 10]. For large molecules, and especially for crystalline materials, these approaches are not only prohibitively expensive but also numerically imprecise. Faster approaches for degree-of-freedom counting and rigid component calculations are known for mechanical frameworks characterized by theorems of Maxwell-Laman type. For the infinite, periodic structures relevant to crystallography, an adequate *generic* rigidity theoretical formulation has been proposed only recently [1], leading to a combinatorial treatment [4, 5] and efficient algorithms.

Generic and non-generic frameworks. Maxwell-Laman theorems are rare and difficult to obtain (see [9, 17] and the references given there), yet they are the starting point of any research on computationally tractable rigidity and flexibility studies of mechanical structures. They provide generic, *combinatorial characterizations* in graph-theoretical terms, for those structures which are minimally rigid for *almost all possible geometric realizations.* For a measure zero set of non-generic situations, such a theorem will not hold. The theory also predicts flexibility and counts infinitesimal degrees of freedom in generic situations. While deciding rigidity and flexibility for generic frameworks is a tractable problem in most of the situations where Maxwell-Laman theorems have been found (see [14]), the non-generic cases remain elusive.

Results. In this paper we apply our theory of periodic bar-and-joint frameworks to tetrahedral crystal structures modeled on quartz, cristobalite and tridymite. We obtain topological descriptions of their configuration spaces.

2 Generic rigidity for Periodic frameworks

To put in perspective the present results, we remind the reader the classical combinatorial characterization of rigidity, in terms of graph sparsity. Then, we give a brief overview of our recent theorems, characterizing generic periodic rigidity in arbitrary dimensions and the flexibility of frameworks made from vertex-sharing simplices, as well as the algorithmic implications.

Maxwell-Laman sparsity conditions. The theory of finite frameworks goes back almost 150 years to Maxwell [15], who identified a *sparsity condition* to be necessary for minimal rigidity of bar-and-joint frameworks: in dimension d, for any subset of $d \leq n' \leq |V|$ vertices, the underlying graph (with a node for each joint and an edge for each bar) should span at most $dn' - \binom{d+1}{2}$ edges, with equality for the whole set of $n = |V|$ vertices. The sufficiency of this condition for generic frameworks in dimension two was proven over 100 years later (Laman's

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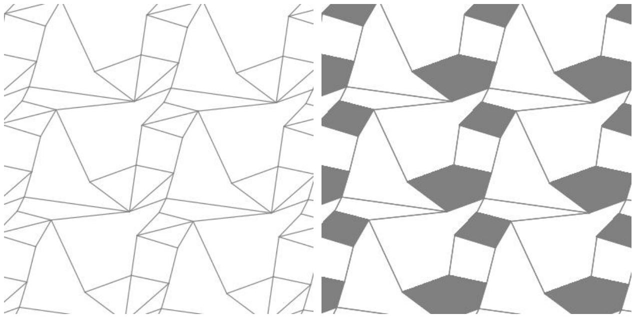

Figure 1: Left: A 2D periodic bar-and-joint framework containing smaller rigid components. Right: the same framework, viewed as a body-and-bar framework; the rigid components form the bodies, and the remaining bars connect distinct bodies.

theorem [12]), and is known to fail in higher dimensions. The problem of completing the combinatorial characterization for bar-and-joint frameworks in arbitrary dimensions remains an elusive open question. However, some restricted classes of finite frameworks have been shown to have similar Maxwell-Laman counting characterizations: body-and-bar and body-and-hinge frameworks [18, 19], and panel-and-hinge frameworks [11].

Maxwell-Laman sparsity for periodic frameworks. The connection pattern of a periodic bar-and-joint framework determines an infinite graph $G = (V, E)$. Periodicity, or more precisely d-periodicity (where d represents the dimension of the ambient space in which the graph is realized geometrically) requires a free Abelian automorphism subgroup $\Gamma \subset Aut(G)$ of rank d. We work under the assumption that the quotient graph G/Γ has a finite number n of vertex orbits and a finite number m of edge orbits. The problem of characterizing periodic frameworks is substantially different from the finite case, and the generic periodic bar-and-joint frameworks have been characterized in all dimensions by a Maxwell-sparsity condition *on the quotient graph*. However, a quotient graph may correspond to several periodic frameworks, called "liftings". The following result gives, therefore, a necessary condition for rigidity which is also sufficient in almost all the situations (i.e. except for a measure-zero set of possibilities).

Theorem 1 *([4]) Let (G, Γ) be a d-periodic graph. Let n and m denote the number of vertices, respectively edges of the graph G modulo the periodicity group Γ.*

If (G, Γ) is minimally rigid, then $m = dn + \binom{d}{2}$ and the quotient graph G/Γ contains a subgraph with $dn - d$ edges on the n vertices, which is $(dn - d)$-sparse.

Conversely, if $m = dn + \binom{d}{2}$ and the quotient graph G/Γ contains a subgraph with $dn - d$ edges on the n vertices, which is $(dn - d)$-sparse, then a generic lifting of the edges yields a minimally rigid d-periodic graph, that is, a generic quotient equivalent of (G, Γ) is minimally rigid.

As a consequence, there are efficient (pebble game) algorithms for deciding generic periodic bar-and-joint rigidity [13].

As we said, a quotient graph may correspond to several periodic frameworks. Distinguishing among them the truly rigid ones remains a difficult problem. Sometimes, substructures in the infinite graph can be identified from the outset as being rigid; see Fig. 1 for an example in 2D. The quotient graph loses this information. We overcome this problem if we work with more specific types of frameworks, such as periodic body-and-bar, body-and-hinge, body-and-pin, etc. They all appear as special cases where *additional algebraic dependencies* are present. Such cases are not guaranteed to be generic *a priori*, and even getting a necessary sparsity condition (something that was trivial in the finite case) is not easy. A recent result along these lines is [5], which covers many situations occurring in molecular frameworks (body-and-bar, body-and-hinge, mixed plate-and-bar), but not the vertex-sharing polyhedra of this paper. Indeed, characterizing generic body-and-pin structures remains an open question, in both the finite and periodic case.

We have further proven that:

Theorem 2 *([1]) A periodic framework in R^d consisting in vertex-sharing simplices has at least $\binom{d}{2}$ infinitesimal degrees-of-freedom (flexes). However, rigid examples can be constructed.*

For the structures studied in this paper, all of which are vertex-sharing tetrahedra, this theorem implies the existence of at least 3 infinitesimal flexes. However, the existence of infinitesimal flexes does not imply a configuration space of dimension three; in fact, in [1] we construct an explicit rigid example.

With these preliminaries in mind, one can see that our results (presented next) confirm the predictions of the generic theory, but do not follow directly from these theorems; indeed, they require different proof techniques, analyzing the entire configuration space (via appropriate parametrizations), rather than just the infinitesimal behavior.

3 The quartz framework

The ideal quartz structure considered here is built from congruent regular tetrahedra. Quartz is made from two types of atoms, oxygen and silicon. Oxygen atoms correspond to the vertices of the tetrahedra. Each oxygen is shared by two tetrahedra and allows for their relative rotations. Silicon atoms are placed at the centers of the tetrahedra, and are rigidly attached to the oxygens. We aim at examining all the geometric configurations of the periodic framework, without concern for self-collision or any other prohibition of a physical nature.

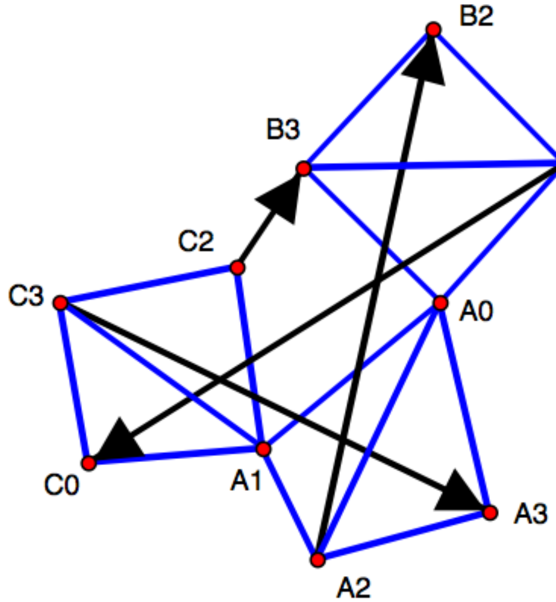

Figure 2: A fragment of the tetrahedral framework of quartz. The periodicity lattice is generated by the four black vectors, which must maintain a zero sum under deformation. The full (infinite) framework is obtained by translating the depicted tetrahedra with all periods (black arrows).

We rely on the notation described in Figure 2. Equivalence under Euclidean motions is eliminated by assuming the tetrahedron marked $A_0A_1A_2A_3$ as fixed. Since all edges maintain their length, the positions of the two tetrahedra which share the vertices A_0 and A_1 are completely described by two orthogonal transformations R_0, respectively R_1 as follows: R_0 fixes A_0 and takes A_i to B_i, $i \neq 0$, while R_1 fixes A_1 and takes A_j to C_j, $j \neq 1$. Figure 2, by depicting only the 'visible' edges, implies that both R_0 and R_1 are orientation reversing, that is, as orthogonal matrices $-R_0, -R_1 \in SO(3)$.

If we denote the edge vectors $A_i - A_0$ by $e_i, i = 1, 2, 3$, we have:

$$B_3 - C_2 = R_0e_3 - (e_1 + R_1(e_2 - e_1))$$

$$A_3 - C_3 = e_3 - (e_1 + R_1(e_3 - e_1))$$

$$B_2 - A_2 = R_0e_2 - e_2$$

$$C_0 - B_1 = e_1 - R_1e_1 - R_0e_1$$

It follows that the dependency condition of a zero sum for these four generators of the periodicity lattice takes the form

$$R_1(e_1 - e_2 - e_3) - R_0(e_1 - e_2 - e_3) = e_1 + e_2 - e_3 \quad (1)$$

Under our regularity assumptions, the three vectors $R_1(e_1 - e_2 - e_3)$, $R_0(e_1 - e_2 - e_3)$ and $(e_1 + e_2 - e_3)$ have the same length and form an equilateral triangle. This restricts $R_0(e_1 - e_2 - e_3)$ to the circle on the sphere of radius $||e_1 - e_2 - e_3||$ (which corresponde with an angle of $2\pi/3$ with $e_1 + e_2 - e_3$). Thus, $-R_0 \in SO(3)$ is constrained to a surface, which is differentiably a two-torus $(S^1)^2$.

For each choice of $-R_0$ on this torus, $R_1(e_1 - e_2 - e_3)$ is determined by (1), hence $-R_1$ is restricted to a circle S^1 in $SO(3)$. We summarize these calculations as:

Theorem 3 *The deformation space of the ideal quartz framework is given by a three dimensional torus $(S^1)^3$ minus the degenerate cases when the span of the four vectors is less than three dimensional.*

4 The cristobalite framework

The 'ideal β cristobalite' structure is illustrated in Figures 3 and 4. The periodicity group of the framework is given by all the translational symmetries of the ideal crystal framework. As a result, there are $n = 4$ orbits of vertices and $m = 12$ orbits of edges.

Adopting the notations of Figure 3, we may assume the tetrahedron $Os_1s_2s_3$ as fixed and parametrize the possible positions of the other tetrahedon by a rotation around the origin O. We obtain that:

Theorem 4 *The deformation space of the ideal high cristobalite framework is naturally parametrized by the open set in $SO(3)$ where the depicted generators remain linearly independent.*

5 The tridymite framework

The tetrahedral framework (G, Γ) of tridymite is depicted in Figure 5. We consider the ideal case made of regular tetrahedra. The quotient graph has $|V/\Gamma| = 8$ and $|E/\Gamma| = 24$. All deformations can be described by three orthogonal transformations (matrices) R_0, R_1, R_2 acting with centers at $O, O1$ and respectively $O2$. With O as the origin and the tetrahedron $OD_1E_1O_1$ assumed fixed, we denote:

$$O_1 = f_0, \quad D_1 = f_1 \ \text{ and } \ E_1 = f_2$$

Then, our orthogonal transformations are determined by the following relations:

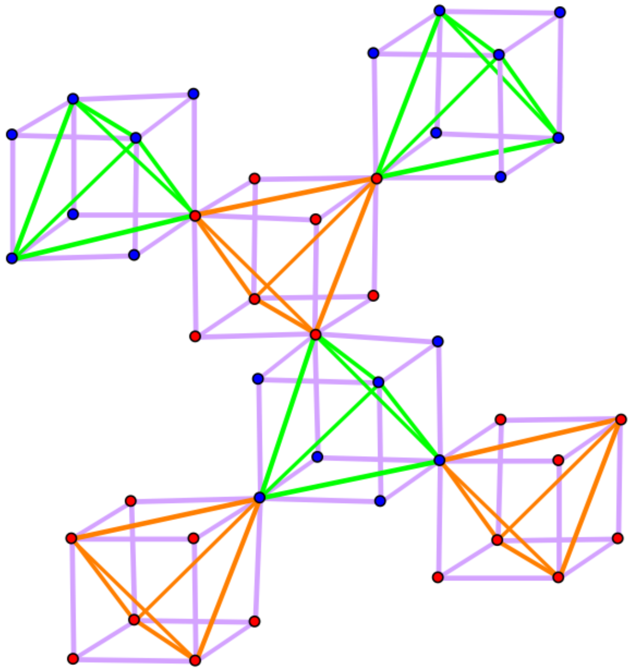

Figure 3: The ideal cristobalite framework (aristotype). The framework is made of vertex sharing regular tetrahedra. Cubes are traced only for suggestive purposes regarding symmetry and periodicity. See also Figure 4.

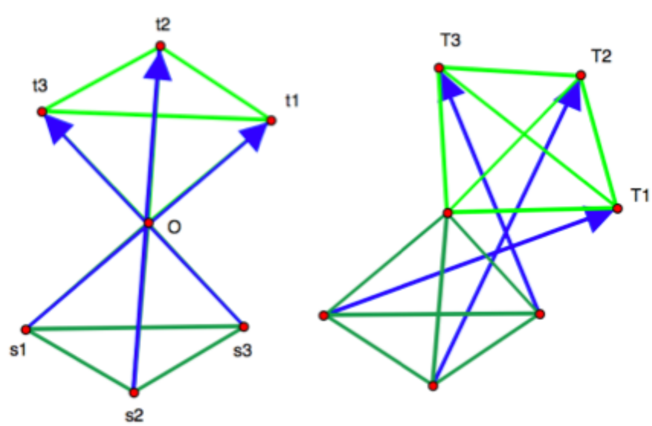

Figure 4: Deforming the ideal cristobalite framework. The periodicity lattice is generated by the three vectors $\gamma_i = t_i - s_i$ which vary as the framework deforms.

$$O_2 = R_0 f_0, \quad D_2 = R_0 f_1 \quad \text{and} \quad E_2 = R_0 f_2$$

$$A_1 = f_0 + R_1(f_1 - f_0)$$

$$B_1 = f_0 + R_1(f_2 - f_0)$$

$$C_1 = f_0 - R_1 f_0$$

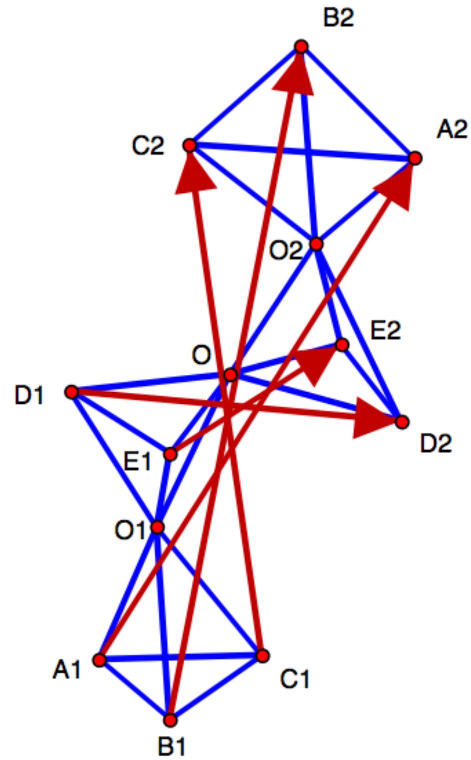

Figure 5: The tetrahedral framework of tridymite. The periodicity lattice is generated by the marked vectors, subject to the relations $(C_2 - C_1) + (D_2 - D_1) = (A_2 - A_1)$ and $(C_2 - C_1) + (E_2 - E_1) = (B_2 - B_1)$.

and

$$A_2 = R_0 f_0 + R_2 R_0 (f_1 - f_0)$$

$$B_2 = R_0 f_0 + R_2 R_0 (f_2 - f_0)$$

$$C_2 = R_0 f_0 - R_2 R_0 f_0$$

As a result, the two linear dependence relations between the six depicted periods take the form:

$$(I - R_0 - R_1 + R_2 R_0) f_i = 0, \quad i = 1, 2 \qquad (2)$$

where I denotes the identity. We note that the ideal high tridymite structure (the *aristotype*) corresponds to $R_0 = -I$ and $R_1 = R_2$ the reflection in the plane $span(f_1, f_2)$.

We now describe the deformation space in a neighborhood of this high tridymite structure. We put $-R_0 = Q$, $R_1 = Q_1$ and $-R_2 R_0 = Q_2$, so that (2) becomes:

$$I + Q = Q_1 + Q_2 \quad \text{on} \quad span(f_1, f_2) \qquad (3)$$

with $Q, -Q_1, -Q_2 \in SO(3)$. Since the orthogonal transformations Q, Q_1, Q_2 are completely determined by their values on two vectors e_1, e_2 of a Cartesian frame with $span(e_1, e_2) = span(f_1, f_2)$, we have to solve the system:

$$e_i + Qe_i = Q_1 e_i + Q_2 e_i \quad i = 1, 2 \qquad (4)$$

where we assume $Q \in SO(3)$ given in a neighborhood of the identity transformation, and look for solutions Q_1, Q_2.

This system may be interpreted in terms of *spherical four-bar mechanisms* in the following way. All the vectors implicated in (4) are unit vectors and can be depicted as points on the unit sphere S^2. For a given Q, we mark by M_i the midpoint of the spherical geodesic segment $[e_i, Qe_i]$ and trace the circle with center M_i and diameter $[e_i, Qe_i]$. This is illustrated in Figure 6.

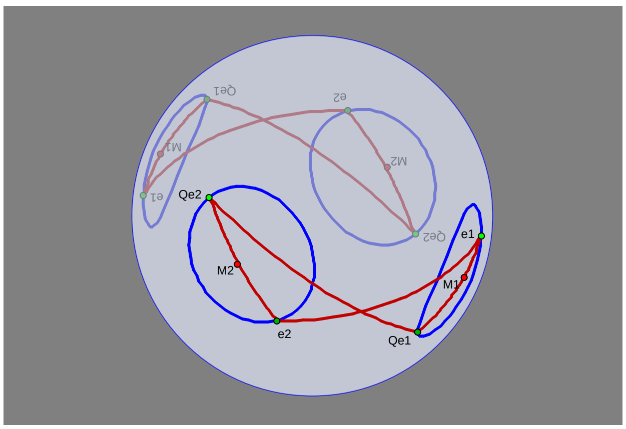

Figure 6: The spherical four-bar mechanism associated to the system (4).

It is an elementary observation that any solution $Q_1 e_i$ and $Q_2 e_i$ determines diameters of the corresponding circles for $i = 1, 2$, with the two geodesic arcs $[Q_k e_1, Q_k e_2]$, like $[e_1, e_2]$ and $[Qe_1, Qe_2]$, of length $\pi/2$. Thus, the two spherical quadrilaterals with vertices at e_1, Qe_1, Qe_2, e_2 and respectively $Q_1 e_1$, $Q_2 e_1$, $Q_2 e_2$, $Q_1 e_2$ are two configurations of the same four-bar mechanism and moreover, the distance between the midpoints of the opposite edges represented by diameters is the same.

It follows from the theory of the spherical four-bar mechanism that, for a generic Q near the identity of $SO(3)$, the abstract configuration space is made of two loops which correspond by reflecting the corresponding realizations. Each loop component has two configurations with the prescibed distance $[M_1 M_2]$. Thus, there are four configurations with the prescribed distance.

We observe that if we replace Q_1 by Q_2 and Q_2 by Q_1 in the labeling of the vertices of a realization, the orientation is reversed, hence the configuration belongs to the other component. Thus, the two obvious solutions of (4), namely:

$$Q_1 e_i = e_i, \; Q_2 e_i = Qe_i$$

and

$$Q_1 e_i = Qe_i, \; Q_2 e_i = e_i, \quad i = 1, 2$$

correspond to configurations belonging to different loop components, as do the remaining two, which are also paired by relabeling. This discussion shows that all four solutions are obtained from the quadrilateral e_1, Qe_1, Qe_2, e_2 and its reflection in the geodesic $[M_1, M_2]$, by the two relabelings with Q_1 and Q_2 possible in each case.

In Figure 7 we have depicted the quadrilateral e_1, Qe_1, Qe_2, e_2 as $A_1 B_1 B_2 A_2$, with reflection in $[M_1 M_2]$ marked as rA_1, rB_1, rB_2, rA_2. Then,, the solutions $(Q_1 e_1, Q_1 e_2, Q_2 e_1, Q_2 e_2)$ of the system (4) are the following four solutions: (A_1, A_2, B_1, B_2), (B_1, B_2, A_1, A_2), (rA_1, rA_2, rB_1, rB_2) and (rB_1, rB_2, rA_1, rA_2).

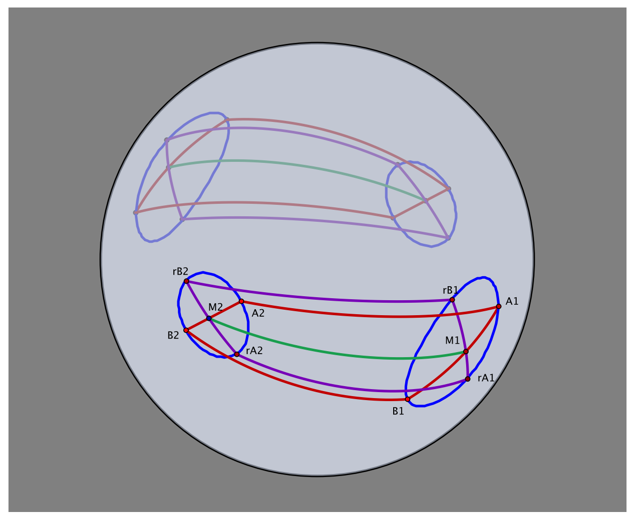

Figure 7: Spherical four-bar mechanism and reflection in $[M_1, M_2]$.

We summarize this result as:

Theorem 5 *The deformation space of the tridymite framework is singular in a neighbourhood of the aristotype and can be represented as a ramified covering with four sheets of a three-dimensional domain. There is a natural $Z_2 \times Z_2$ action on this covering which fixes the aristotype framework.*

Indeed, the two involutions, inverting the labeling and reflecting in $[M_1, M_2]$, commute and give a $Z_2 \times Z_2$. action on the covering. The dimension of the tangent space at the aristotype framework is computed from the linear version of (4) and is six.

6 Conclusions

Periodic (crystalline) materials, either occurring in nature or man-made, have been studied with experimental tools (such as X-ray crystallography) for over 100 years, and yet important phenomena related to their flexibility properties remain largely uncharted. In this paper, we studied the flexibility of three important families of periodic structures. These frameworks are flexible, and descriptions of their configuration spaces were explicitly given. A version of this paper has been posted on the arxiv [2].

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