Icosahedral order in glass: Acoustic properties

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An icosahedral crystal in the non-Euclidean three-dimensional space S^3 models domains of icosahedral order in metallic glass. Crystal symmetries allow explicit construction and classification of vibrational eigenstates. We evaluate phonon frequencies numerically. Long-wavelength, low-frequency vibrations follow the predictions of continuum elastic theory in S^3, and of a hypothetical perfect icosahedral crystal in $\mathbb{R}^3$. Isotropy of elastic moduli distinguishes icosahedral order from face-centered-cubic order.

I. INTRODUCTION

Polytope 120 (a close-packed, icosahedral crystal consisting of 120 atoms in S^3) models local order in metallic glass.[1–3] Structural and electronic properties of the polytope approximate corresponding physical properties of real glass in flat space.[4–7] This paper addresses the acoustic and vibrational properties of Polytope 120. Isotropy of the elastic moduli distinguishes close-packed icosahedral order from face-centered-cubic order.[8] Isotropy of elastic moduli is shown to arise from the high symmetry of the icosahedral point group. Soft modes are suppressed because the polytope contains only tetrahedral cells, which resist distortions more effectively than octahedra.[9]

Continuum elastic theory describes long-wavelength, low-frequency vibrations of crystals. Section II of this paper develops continuum elastic theory in the three-dimensional curved space S^3. We generalize elastic free energy and strain tensors to non-Euclidean space and derive the spectrum of phonons on S^3.

Section III of this paper addresses the spectrum of phonons on the polytope. We assume atoms interact with their neighbors through the potential $\Phi(s)=4/s^{12}$, where s is the Euclidean distance between atoms. Symmetries of Polytope 120 allow explicit construction of vibrational wave functions transforming under irreducible representations of the symmetry group. We evaluate the frequencies of these vibrational modes. Finally, we discuss the relationship between continuum elastic theory and the polytope vibrations. Elastic constants are calculated and compared with a hypothetical perfect icosahedral crystal in $\mathbb{R}^3$.

The remainder of this Introduction develops the mathematical tools required to describe and manipulate vector fields in S^3. We use quaternions to label points in S^3. Quaternion multiplication provides a convenient set of local coordinate systems and simple expressions for covariant derivatives. Vector fields in these coordinate systems may be expanded in terms of vector spherical harmonics. We investigate properties of these harmonics.

A. Quaternions

Previous studies of Polytope 120 exploited the isomorphism between S^3 and the group SU(2) (Refs. 6, 7, and 10). In this paper we exploit an additional isomorphism between the vector space R^4 and the ring of quaternions.[11] Consider

$$\mathbf{u}=u^{\alpha}\hat{\mathbf{e}}_{\alpha}\in\mathbb{R}^4\,, \tag{1.1a}$$

$$\mathbf{v}=v^{\alpha}\hat{\mathbf{e}}_{\alpha}\in\mathbb{R}^4\,. \tag{1.1b}$$

The quaternion product of $\mathbf{u}$ and $\mathbf{v}$ is

$$\mathbf{uv}=(u^0v^0-u^jv_j)\hat{\mathbf{e}}_0+(u^0v^j+v^0u^j)\hat{\mathbf{e}}_j+\varepsilon^i{}_{jk}u^jv^k\hat{\mathbf{e}}_i\,, \tag{1.2}$$

where $i,j,k\in\{1,2,3\}$. The inverse of $\mathbf{u}$ is

$$\mathbf{u}^{-1}=u^0\hat{\mathbf{e}}_0-u^i\hat{\mathbf{e}}_i\,. \tag{1.3}$$

Quaternions may be added together by

$$\mathbf{u}+\mathbf{v}=(u^{\alpha}+v^{\alpha})\hat{\mathbf{e}}_{\alpha}\,. \tag{1.4}$$

We also define the dot product of two quaternions

$$\mathbf{u}\cdot\mathbf{v}=u^{\alpha}v_{\alpha}\,. \tag{1.5}$$

The group of unit quaternions,

$$Q=\{u^{\alpha}\hat{\mathbf{e}}_{\alpha}:\ u^{\alpha}u_{\alpha}=1\}\,, \tag{1.6}$$

is isomorphic to SU(2) through the mapping

$$\mathbf{u}=u^{\alpha}\hat{\mathbf{e}}_{\alpha}\in Q\leftrightarrow u=\begin{bmatrix}u^0+iu^3 & iu^1+u^2\\ iu^1-u^2 & u^0-iu^3\end{bmatrix}\in\mathrm{SU}(2)\,. \tag{1.7}$$

Thus the icosahedral symmetry group $Y'\subset\mathrm{SU}(2)$ can be embedded in Q. Because of the isomorphism between Y' and Polytope 120, the four components of each quaternion in Y^1 are the four Cartesian coordinates of each atom in the polytope.[7,11] Furthermore, Q is isomorphic to the unit sphere in four dimensions, S^3. Thus the symmetry group[12] of S^3,

$$\mathrm{SO}(4)=[\mathrm{SU}(2)\times\mathrm{SU}(2)]/Z_2\,. \tag{1.8}$$

We refer to the set $SU(2)=\{(l,r)\in SO(4):\ l=r\}$ as the "diagonal subgroup" of SO(4). Irreducible representations of SO(4) are denoted by integers (M,N), where $M/2$ and $N/2$ label corresponding irreducible representations of SU(2). Irreducible representations of the form (M,M) are "diagonal irreducible representations" and describe scalar fields on S^3. Note that the SU(2) matrix elements may be associated with the four vectors

$$\hat{e}_{1/2\,1/2}=\hat{e}_0+i\hat{e}_3,\quad \hat{e}_{1/2\,-1/2}=i\hat{e}_1+\hat{e}_2\ ,\quad \hat{e}_{-1/2\,1/2}=i\hat{e}_1-\hat{e}_2,\quad \hat{e}_{-1/2\,1/2}=\hat{e}_0-i\hat{e}_3\ , \tag{1.9}$$

which form a basis for the (1,1) irreducible representation of SO(4).

Two inequivalent, orthonormal, local coordinate systems for $\mathbb{R}^4$ at $\mathbf{u}\in Q$ are furnished by

$$\hat{e}^R_\alpha(\mathbf{u})=\hat{e}_\alpha\mathbf{u}\ , \tag{1.10a}$$

$$\hat{e}^L_\alpha(\mathbf{u})=\mathbf{u}\hat{e}_\alpha\ . \tag{1.10b}$$

We will always work in the coordinate system $\hat{e}_\alpha(\mathbf{u})=\hat{e}^R_\alpha(\mathbf{u})$. The outward normal to S^3 at $\mathbf{u}$ is $\hat{e}_0(\mathbf{u})=\mathbf{u}$. The remaining components $\hat{e}_\alpha(\mathbf{u})$ $(\alpha=1,2,3)$ form a right-handed local coordinate system for S^3.

Misner, Thorne, and Wheeler[13] discuss the differential geometry of SO(3). They find that the connection coefficients of S^3 are given by

$$\Gamma^a_{bc}=\varepsilon^a_{bc}\ , \tag{1.11a}$$

the commutation coefficients are

$$C^c_{ab}=-2\varepsilon^a_{bc}\ , \tag{1.11b}$$

and the Riemann tensor is

$$R^a_{bcd}=\delta^a_c\delta_{bd}-\delta^a_d\delta_{bc}\ . \tag{1.11c}$$

The factors of 2 by which Eqs. (1.11) differ from Misner, Thorne, and Wheeler arise because they discuss the rotation group SO(3), not the sphere S^3. The covariant derivative of a vector field ψ^a is

$$\psi^a{}_{;b}=\psi^a{}_{,b}+\varepsilon^a{}_{bc}\psi^c\ . \tag{1.12}$$

B. Vector hyperspherical harmonics

Vector hyperspherical harmonics[14,15] provide a complete set of basis functions in which to express vector fields on S^3. The harmonics may be defined by their transformation properties under SO(4). In this section we derive expressions for vector hyperspherical harmonics and analyze the differential equation they obey.

The most general vector field on S^3 may be written

$$\boldsymbol{\psi}(\mathbf{u})=\psi^\alpha(\mathbf{u})\hat{e}_\alpha\ , \tag{1.13}$$

where $\psi^\alpha(\mathbf{u})$ are scalar fields. Expanding the scalar fields in hyperspherical harmonics, substituting the (1,1) irreducible representation (1.9) of SO(4) for the usual $\mathbb{R}^4$ basis $\hat{e}_\alpha$, and exploiting the relationship (1.8) between SU(2) and SO(4), we find that a complete set of vector fields on S^3 is given by[16]

$$\mathbf{Y}_{MM_1M_2,m_1m_2}(\mathbf{u})=\sum_{\mu,\nu,a,b}\begin{bmatrix}M/2 & M_1/2 & \frac{1}{2}\\ \mu & m_1 & a\end{bmatrix}\begin{bmatrix}M/2 & M_2/2 & \frac{1}{2}\\ \nu & m_2 & b\end{bmatrix}Y_{M,\mu\nu}(\mathbf{u})\hat{e}_{ab}\ . \tag{1.14}$$

Equation (1.14) multiplies the $(M+1)^2$ dimensional scalar hyperspherical harmonic (M,M) by the four-dimensional vector representation (1,1), yielding two diagonal representations $(M\pm1,M\pm1)$ of dimension $[M+(1\pm1)]^2$ and two off-diagonal representations $(M\pm1,M\mp1)$ of dimension $M(M+2)$. One can easily show that the off-diagonal vector fields are tangent to S^3. The $2M^2+4M+4$ diagonals are not, in general, tangent to S^3. In fact, one can show that M^2 linear combinations form vector fields normal to S^3 transforming like the $M-1$ hyperspherical harmonic. The other $(M+2)^2$ linear combinations form tangential vector fields.

Interesting special cases of Eq. (1.14) include

$$\mathbf{Y}_{120,\pm10}(\mathbf{u})=\frac{1}{\sqrt{6i}}[i\hat{e}^L_1(\mathbf{u})\pm\hat{e}^L_2(\mathbf{u})]\ ,\qquad \mathbf{Y}_{120,00}(\mathbf{u})=\frac{1}{\sqrt{3i}}\hat{e}^L_3(\mathbf{u}) \tag{1.15a}$$

and

$$\mathbf{Y}_{102,0\pm1}(\mathbf{u})=\frac{1}{\sqrt{6i}}[i\hat{e}^R_1(\mathbf{e})\pm\hat{e}^R_2(\mathbf{u})]\ ,\qquad Y_{102,00}(\mathbf{u})=\frac{1}{\sqrt{3i}}\hat{e}^R_3(\mathbf{u})\ , \tag{1.15b}$$

which we recognize as left and right screws. Also, we find $\mathbf{Y}_{011,mn}(\mathbf{u})$ as constant vector fields in $\mathbb{R}^4$ pointing in the direction $\hat{e}_{-m\,-n}$, and $\mathbf{Y}_{100,00}(\mathbf{u})=\mathbf{u}$ as the "breathing mode".

The split into normal and tangential components is most easily seen in the local coordinate system $\hat{e}_\alpha(\mathbf{u})$:

$$\boldsymbol{\psi}^N_{M,m_1m_2}(\mathbf{u})=Y_{M,m_1m_2}(\mathbf{u})\hat{e}_0(\mathbf{u}) \tag{1.16}$$

are normal vector fields transforming like the Mth hyperspherical harmonic. The remaining diagonal vector fields are tangent to S^3 and may be expressed

$$\boldsymbol{\psi}^0_{M,m_1m_2}(\mathbf{u})=\nabla Y_{M,m_1m_2}(\mathbf{u})=Y_{M,m_1m_2;\alpha}\hat{e}_\alpha(\mathbf{u})\ . \tag{1.17}$$

We call the vector fields (1.17) "longitudinal phonons" because in the large-M (short-wavelength) limit (1.17) is the usual definition of a longitudinal mode as the gradient of a scalar field.

Off-diagonal vector fields can also be easily expressed in terms of local coordinate systems. Define

$$\psi^{\tau\sigma}_{M,m_1m_2}(\mathbf{u})=\boldsymbol{\psi}^{\tau}_{M,m_1m_2}(\mathbf{u})\cdot\hat{\mathbf{e}}_{\sigma}(\mathbf{u}) , \tag{1.18}$$

where $\tau=-,0,+$ and $\sigma=-,+,3$. Longitudinal phonons correspond to $\tau=0$, $\tau=\pm$ denote the $(M\pm1,M\mp1)$ irreducible representation of SO(4), and

$$\hat{\mathbf{e}}_{\pm}(\mathbf{u})=i\hat{\mathbf{e}}_1(\mathbf{u})\pm\hat{\mathbf{e}}_2(\mathbf{u}) . \tag{1.19}$$

Sen[15] demonstrates

$$\psi^{\tau,3}_{M,m_1m_2}(\mathbf{u})=\cos\alpha(M,m_1)Y_{M-\tau,m_1m_2}(\mathbf{u}) ,$$

$$\psi^{\tau,+}_{M,m_1m_2}(\mathbf{u})=\sqrt{2}\sin\alpha(M,m_1)\cos\beta^{\tau}(M,m_1)\times Y_{M-\tau,m_1+1,m_2}(\mathbf{u}) , \tag{1.20}$$

$$\psi^{\tau,-}_{M,m_1m_2}(\mathbf{u})=\sqrt{2}\sin\alpha(M,m_1)\sin\beta^{\tau}(M,m_1)\times Y_{M-\tau,m_1-1,m_2}(\mathbf{u}) ,$$

where

$$\alpha(M,m_1)=\cos^{-1}\{[(M+2m_1+1)(M-2m_1+1)/2M(M+1)]^{1/2}\} ,$$
$$\beta^{+}(M,m_1)=-\tan^{-1}(\{[(M-2m_1)^2-1]/[(M+2m_1)^2-1]\}^{1/2}) , \tag{1.21}$$
$$\beta^{-}(M,m_1)=\pi-\tan^{-1}(\{[(M+2m_1+2)^2-1]/[(M-2m_1+2)^2-1]\}^{1/2}) .$$

Transverse vector fields satisfy[14]

$$\nabla\times\boldsymbol{\psi}^{\tau}_{M,m_1m_2}=-\tau(M+1)\boldsymbol{\psi}^{\tau}_{M,m_1m_2} , \tag{1.22}$$

whereas longitudinal vector fields have zero curl. Similarly,[12]

$$\nabla\cdot\boldsymbol{\psi}^{0}_{M,m_1m_2}=\nabla^2 Y_{M,m_1m_2}=-M(M+2)Y_{M,m_1m_2} , \tag{1.23}$$

whereas transverse vector fields have zero divergence. Transverse phonons in $\mathbf{R}^3$ obey a differential equation similar to (1.22), e.g.,

$$\nabla\times(i\hat{\mathbf{e}}_x+\hat{\mathbf{e}}_y)e^{ik\hat{\mathbf{e}}_z\cdot\mathbf{R}}=-k(i\hat{\mathbf{e}}_x+\hat{\mathbf{e}}_y)e^{ik\hat{\mathbf{e}}_z\cdot\mathbf{R}} . \tag{1.24}$$

II. CONTINUUM ELASTIC THEORY

This section adapts continuum elastic theory to non-Euclidean space. The strain tensor in curved space resembles the strain tensor in flat space with covariant derivatives replacing ordinary derivatives. The free-energy density remains a quadratic form in the strain tensor. We show that the elastic moduli of an icosahedral system are isotropic,[8] distinguishing icosahedral symmetry from any flat-space crystalline symmetry. Useful identifies among vector differential operators, and integration by parts, are used to calculate the phonon spectrum of S^3.

Given a displacement field $\boldsymbol{\psi}=\psi^a\hat{\mathbf{e}}_a$ in flat space one defines the symmetric strain tensor[17]

$$u_{ab}=\frac{1}{2}\left[\frac{\partial\psi^a}{\partial x_b}+\frac{\partial\psi^b}{\partial x_a}\right] . \tag{2.1}$$

After deformation, two points initially separated by $dR=(dx_a\,dx_a)^{1/2}$ become separated by dR', where

$$(dR')^2=dR^2+2u_{ab}\,dx_a\,dx_b . \tag{2.2}$$

Now imagine deformations which occur within a non-Euclidean space. To compensate for the rotation of intrinsic coordinate systems as viewed from a higher dimensional embedding space, one simply substitutes covariant derivatives for ordinary derivatives,[13]

$$u^a_b=\tfrac{1}{2}(\psi^a{}_{;b}+\psi_b{}^{;a}) . \tag{2.3}$$

Note that in the coordinate system $[\hat{\mathbf{e}}_\alpha(u)]$, the connection coefficients (1.9) cancel in Eq. (2.3). Thus the definitions (2.3) and (2.1) are equivalent in any coordinate system in which

$$\Gamma^a{}_{bc}+\Gamma^c{}_{ba}=0 . \tag{2.4}$$

Equation (2.4) holds, in particular, in orthonormal coordinate systems in spaces of constant metric.

Were we to find the strain tensor in the coordinate system of our flat, embedding space, additional terms would have to be included in the strain tensor to relate this system to our intrinsic coordinates. These terms are required to compensate for the fact that a curved surface is not always "horizontal" in the flat, embedding space. Sachdev and Nelson[18] show that these terms are

$$A_{ab}=\frac{1}{2}\frac{\partial x_l}{\partial x_a}\frac{\partial x_l}{\partial x_b} , \tag{2.5}$$

where $[x_a]$ forms an orthonormal coordinate system of the non-Euclidean space at some point, and $[x_l]$ includes the remaining orthogonal coordinates in the flat, embedding space.

Following Landau and Lifschitz[17] we write the free-energy density

$$f=\tfrac{1}{2}\lambda_{abcd}u^a{}_b u^c{}_d . \tag{2.6}$$

Symmetries of the polytope constrain the number of independent components of the elastic tensor λ_{abcd}. In fact, icosahedral symmetry allows only two independent elastic constants. Thus the continuum elastic theory of the polytope is identical to the continuum elastic theory of an isotropic medium.

Each index of λ_{abcd} runs over the three spatial components of our coordinate system $[e_a(\mathbf{u})]$. Thus, under rotations of the polytope leaving a site invariant, an index of λ_{abcd} transforms like an $l=1$ spherical harmonic under the icosahedral symmetry group. In order to construct an invariant free-energy density, we must find combinations of the indices which transform under the unit representation of Y'. By addition of angular momentum we can construct the following irreducible representations:

$$D^1 \otimes D^1 \otimes D^1 \otimes D^1 = 3D^0 \oplus 6D^1 \oplus 6D^2 \oplus 3D^3 \oplus D^4 , \quad (2.7)$$

where each D^1 on the left-hand side of (2.7) corresponds to one of the indices of λ_{abcd}.

Spherical harmonics Y_l contain the unit representation of Y' when $l=0,6,10,12,\ldots$.[6] Thus only the three combinations of indices yielding D^0 transform as the unit representation of Y^1. These are

$$\lambda_{aacc},\ \lambda_{abab},\ \lambda_{abba} . \quad (2.8)$$

However, the last two are equivalent because of the symmetry of the strain tensor

$$u^a{}_b = u^b{}_a . \quad (2.9)$$

Thus the most general elastic tensor takes the form

$$\lambda_{abcd} = \lambda \delta_{ab} \delta_{cd} + 2\mu \delta_{ac} \delta_{bd} . \quad (2.10)$$

Cubic systems possess an additional invariant, λ_{aaaa}, because the $l=4$ spherical harmonic contains the unit representation of the cubic-symmetry group.

The continuum elastic free energy is thus

$$F = \frac{1}{2} \int_{S^3} d\Omega\, \lambda_{abcd} u^a{}_b u^c{}_d . \quad (2.11)$$

Evaluation of (2.11) requires the following integrals:

$$I_a = \int_{S^3} d\Omega\, \psi^\alpha{}_{;\alpha} \psi^\beta{}_{;\beta} , \quad (2.12a)$$

$$I_b = \int_{S^3} d\Omega\, \psi^\alpha{}_{;\beta} \psi^\beta{}_{;\alpha} , \quad (2.12b)$$

$$I_c = \int_{S^3} d\Omega\, \psi^\alpha{}_{;\beta} \psi_\alpha{}^{;\beta} , \quad (2.12c)$$

which can be integrated by parts

$$I_a = -\int_{S^3} d\Omega\, \psi^\alpha \psi^\beta{}_{;\beta\alpha} + \int_{S^3} d\Omega (\psi^\alpha \psi^\beta{}_{;\beta})_{;\alpha} , \quad (2.13a)$$

$$I_b = -\int_{S^3} d\Omega\, \psi^\alpha \psi^\beta{}_{;\alpha\beta} + \int_{S^3} d\Omega (\psi^\alpha \psi^\beta{}_{;\alpha})_{;\beta} , \quad (2.13b)$$

$$I_c = -\int_{S^3} d\Omega\, \psi^\alpha \psi_\alpha{}^{;\beta}{}_{;\beta} + \int_{S^3} d\Omega (\psi^\alpha \psi_\alpha{}^{;\beta})_{;\beta} . \quad (2.13c)$$

The second terms in Eqs. (2.13) are exact divergences and hence vanish.

In Eq. (2.13a) we recognize

$$\psi^\beta{}_{;\beta} = \nabla \cdot \boldsymbol{\psi} \quad (2.14)$$

and hence

$$\psi^\beta{}_{;\beta\alpha} = \nabla_\alpha \nabla \cdot \boldsymbol{\psi} . \quad (2.15)$$

Noting Eq. (1.23) we find

$$I_a = -\int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \nabla\nabla \cdot \boldsymbol{\psi} . \quad (2.16)$$

In Eq. (2.13b) we commute the derivatives[13] on α and β to find

$$\psi^\beta{}_{;\alpha\beta} = \psi^\beta{}_{;\beta\alpha} + R^{\ \beta}_{\nu\beta\alpha} \psi^\nu . \quad (2.17)$$

Recognizing the contraction of the Riemann tensor as the Ricci tensor

$$R^{\ \beta}_{\nu\beta\alpha} = R_{\nu\alpha} = 2\delta_{\nu\alpha} , \quad (2.18)$$

we obtain

$$I_b = -\int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \nabla\nabla \cdot \boldsymbol{\psi} - 2\int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \boldsymbol{\psi} . \quad (2.19)$$

Finally, Eq. (2.13c) may be simplified using the identity

$$\nabla \cdot \nabla \boldsymbol{\psi} = \nabla\nabla \cdot \boldsymbol{\psi} + 2\boldsymbol{\psi} - \nabla \times \nabla \times \boldsymbol{\psi} , \quad (2.20)$$

which is the analogue on S^3 of

$$\nabla^2 \boldsymbol{\psi} = \nabla\nabla \cdot \boldsymbol{\psi} - \nabla \times \nabla \times \boldsymbol{\psi} . \quad (2.21)$$

We obtain

$$I_c = \int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \nabla \times \nabla \times \boldsymbol{\psi} - \int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \nabla\nabla \cdot \boldsymbol{\psi} - 2\int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \boldsymbol{\psi} . \quad (2.22)$$

Substituting the form (2.10) of λ_{abcd} into the formula for the free energy (2.11),

$$F = \tfrac{1}{2}(\lambda I_a + \mu I_b + \mu I_c) . \quad (2.23)$$

Using the identities (1.22) and (1.23), we find

$$F_M^L = [(\tfrac{1}{2}\lambda + \mu) M(M+2) - 2\mu] \int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \boldsymbol{\psi} \quad (2.24)$$

for longitudinal phonons and

$$F_M^T = [\tfrac{1}{2}\mu (M-1)(M+3)] \int_{S^3} d\Omega\, \boldsymbol{\psi} \cdot \boldsymbol{\psi} \quad (2.25)$$

for transverse phonons. Inspecting Eqs. (2.24) and (2.25), we find for large M that continuum elastic theory on S^3 is identical to $\mathbb{R}^3$, provided that we identify

$$k_L = \sqrt{M(M+2)} , \quad (2.26)$$

$$k_T = \sqrt{(M-1)(M+3)} , \quad (2.27)$$

as the longitudinal and transverse wave numbers. The resulting sound speeds are simply

$$c_L = \sqrt{(\lambda + 2\mu)/\rho}, \quad c_T = \sqrt{\mu/\rho} .$$

III. POLYTOPE 120

At long wavelengths, vibrations of Polytope 120 resemble vibrations of the continuum. At short wavelengths, however, the discrete symmetry group[10,11] $G = (Y' \times Y')/Z_2$ of the polytope reduces the high dimensional vector hyperspherical harmonics into smaller, irreducible representations. The vibrational frequencies of each irreducible representation may be very different from the predictions of continuum elastic theory. In this section we classify vibrational modes of Polytope 120 according to irreducible representations of G. We construct wave functions and numerically evaluate their frequencies.

Isotropy of acoustical properties simplifies the calculation of elastic constants of real glass in $\mathbb{R}^3$. Assuming a microcrystalline model of glass in which fragments of

TABLE I. Character table of icosahedral symmetry group $Y' \subset SU(2)$. $\Omega = (\sqrt{5}+1)/2$ is the golden mean.

Y'	$1C_0$	$12V_1$	$20F_2$	$12V_3$	$30E_4$	$12V_5$	$20F_6$	$12V_7$	$1C_8$
A	1	1	1	1	1	1	1	1	1
E_1	2	Ω	1	Ω^{-1}	0	$-\Omega^{-1}$	-1	$-\Omega$	-2
E_2	2	$-\Omega^{-1}$	1	$-\Omega$	0	Ω	-1	Ω^{-1}	-2
F_1	3	Ω	0	$-\Omega^{-1}$	-1	$-\Omega^{-1}$	0	Ω	3
F_2	3	$-\Omega^{-1}$	0	Ω	-1	Ω	0	$-\Omega^{-1}$	3
G_1	4	1	-1	-1	0	1	1	-1	-4
G_2	4	-1	1	-1	0	-1	1	-1	4
H	5	0	-1	0	1	0	-1	0	5
I	6	-1	0	1	0	-1	0	1	-6

Polytope 120 pack with random orientations, the elastic constants of the glass are, to a first approximation,[17] equal to the elastic constants of the polytope. This section concludes with a calculation of the elastic constants of the polytope by matching the long-wavelength behavior with continuum elastic theory, and by calculating the sound speeds in a hypothetical perfect icosahedral crystal in $\mathbb{R}^3$.

Techniques from the theory of molecular vibrations[19] may be generalized to four dimensions and applied to Polytope 120. One defines, first of all, the "regular representation" with the 120 vertices of Polytope 120 as basis elements, and characters

$$\chi_R^G(l,r) = \delta_{\Gamma(l)\Gamma(r)}\, {}^o(Y')/{}^o(\Gamma) , \tag{3.1}$$

where $\Gamma(u) \subset Y'$ is the class of $u \in Y'$. Table I presents the character table of Y'. Recall that the regular representation is the union of s-band electronic wave functions.[6] At each vertex we now place a vector which transforms under E_1E_1, which is the (1,1) irreducible representation of SO(4). Thus

$$\chi_{E_1E_1}^G(l,r) = \chi_{E_1}^{Y'}(l)\chi_{E_1}^{Y'}(r) , \tag{3.2}$$

and the resulting 480-dimensional representation, known as the total representation, has characters

$$\chi_T^G(l,r) = \delta_{\Gamma(l)\Gamma(r)}\, {}^o(Y')\chi_{E_1}^{Y'}(l)\chi_{E_1}^{Y'}(r)/{}^o(\Gamma) . \tag{3.3}$$

How many times is the irreducible representation $(\alpha\beta)$ of G contained in the total representation? Using[6]

$$\chi_{\alpha\beta}^G(l,r) = \chi_\alpha^{Y'}(l)\chi_\beta^{Y'}(r) \tag{3.4}$$

and[19]

$$N(\alpha\beta \subset T) = \frac{1}{{}^o(G)} \sum_{(l,r)\in G} \chi_{\alpha\beta}^G(l,r)\chi_T^G(l,r) , \tag{3.5}$$

we obtain

$$N(\alpha\beta \subset T) = \frac{1}{{}^o(Y')} \sum_{\Gamma \subset Y'} {}^o(\Gamma)\chi_\alpha^{Y'}(\Gamma)\chi_\beta^{Y'}(\Gamma)\chi_{E_1}^{Y'}(\Gamma)\chi_{E_1}^{Y'}(\Gamma) . \tag{3.6}$$

Equation (3.6) vanishes unless the quadruple product $\alpha \otimes \beta \otimes E_1 \otimes E_1$ contains the unit representation of Y'. Table II shows the irreducible representation multiplication table[20] of Y'. Using the information in Table II one finds the values of $N(\alpha\beta \subset T)$ presented in Table III.

The 480 modes in Table III include 360 "phonon" modes tangent to S^3 and 120 "electronic" modes normal to S^3. To confirm this, note that the product

$$E_1 \otimes E_1 = F_1 \oplus A . \tag{3.7}$$

Equation (3.6) thus splits into two terms

$$N(\alpha\beta \subset T) = \frac{1}{{}^o(Y')} \sum_{\Gamma \subset Y'} {}^o(\Gamma)[\chi_\alpha^{Y'}(\Gamma)\chi_\beta^{Y'}(\Gamma)\chi_{F_1}^{Y'}(\Gamma) + \chi_\alpha^{Y'}(\Gamma)\chi_\beta^{Y'}(\Gamma)] . \tag{3.8}$$

The first term consists of vector (phonon) representations of G and the second term denotes scalar (electronic) representations;

$$N(\alpha\beta \subset T) = N_v(\alpha\beta) + N_s(\alpha\beta) . \tag{3.9}$$

$N_s(\alpha\beta)$ appears in the s-band electronic properties of Polytope 120 and is known[6] to equal $\delta_{\alpha\beta}$. Thus Table III with 1 subtracted from the diagonal classifies the phonon modes. Adding the two values of l associated with $\alpha\beta$ in Table III yields the principal quantum number M of the vector hyperspherical harmonic containing $\alpha\beta$, and through Eqs. (2.26) and (2.27) yields the wave number.

Choose a vector, $\mathbf{A} \in \mathbb{R}^4$, based at the north pole, $\mathbf{u} = 1 \in S^3$, as a typical basis element of the total representation T. This basis element transforms under

$$(l,r):\ \mathbf{A}\,|\,1\rangle \rightarrow l\mathbf{A}r^{-1}\,|\,lr^{-1}\rangle ; \tag{3.10}$$

thus, we can project an element of the irreducible repre-

TABLE II. Irreducible representation multiplication table of icosahedral group.

Y'	E_1	F_1
A	E_1	F_1
E_1	$A \oplus F_1$	$E_1 \oplus G_1$
E_2	G_2	I
F_1	$E_1 \oplus G_1$	$A \oplus F_1 \oplus H$
F_2	I	$G_2 \oplus H$
G_1	$F_1 \oplus H$	$E_1 \oplus G_1 \oplus I$
G_2	$E_2 \oplus I$	$F_2 \oplus G_2 \oplus I$
H	$G_1 \oplus I$	$F_1 \oplus F_2 \oplus G_2 \oplus H$
I	$F_1 \oplus G_2 \oplus H$	$E_2 \oplus G_1 \oplus 2I$

TABLE III. Occurrences of $\alpha\beta$ in T, Eq. (3.6). Diagonal entries in parentheses denote number of tangential vector representations.

l \ α \ β (l)	0	$\frac{1}{2}$	1	$\frac{3}{2}$	2	$\frac{5}{2}$	3		$\frac{7}{2}$
	A	E_1	F_1	G_1	H	I	F_2	G_2	E_2
0 A	1 (0)	0	1	0	0	0	0	0	0
$\frac{1}{2}$ E_1	0	2 (1)	0	1	0	0	0	0	0
1 F_1	1	0	2 (1)	0	1	0	0	0	0
$\frac{3}{2}$ G_1	0	1	0	2 (1)	0	1	0	0	0
2 H	0	0	1	0	2 (1)	0	1	1	0
$\frac{5}{2}$ I	0	0	0	1	0	3 (2)	0	0	1
3 F_2	0	0	0	0	1	0	1 (0)	1	0
3 G_2	0	0	0	0	1	0	1	2 (1)	0
$\frac{7}{2}$ E_2	0	0	0	0	0	1	0	0	1 (0)

sentation $(\alpha\beta)$ from T using the formula

$$\psi_{\alpha\beta}=\frac{1}{{}^{o}(G)}\sum_{(l,r)\in G}\chi^{G}_{\alpha\beta}(l,r)l\mathbf{A}r^{-1}\,|\,lr^{-1}\rangle\ . \tag{3.11}$$

In view of Eq. (3.4), we find

$$\psi_{\alpha\beta}(\mathbf{u})=\frac{1}{[{}^{o}(Y')]^{2}}\sum_{l\in Y'}\chi^{Y'}_{\alpha}(l)\chi^{Y'}_{\beta}(l^{-1}\mathbf{u})l\mathbf{A}l^{-1}\mathbf{u}\ . \tag{3.12}$$

Equation (3.12) is guaranteed to produce a vibrational eigenstate, provided $\alpha\beta$ is contained only once in $\mathbf{A}\,|\,1\rangle$. Inspecting Table III we see we must work a little harder for the longitudinal modes.

If we choose our vector normal to S^3, $\mathbf{A}=\hat{\mathbf{e}}_0(1)$, $\mathbf{A}$ will commute with all $l\in Y'$ and Eq. (3.12) cannot generate any tangential components. Equation (3.12) yields

$$\psi^{N}_{\alpha\beta,mn}(\mathbf{u})=\delta_{\alpha\beta}Y_{\alpha,mn}(\mathbf{u})\hat{\mathbf{e}}_0(\mathbf{u})\ , \tag{3.13}$$

where $Y_{\alpha,mn}$ are diagonal irreducible representation basis functions. Similarly, if we choose $\mathbf{A}$ tangent to S^3 then Eq. (3.12) cannot generate normal components. This follows because $\mathbf{A}\in\mathbb{R}^3$ implies $l\mathbf{A}l^{-1}\in\mathbb{R}^3$ for all $l\in\mathrm{SU}(2)$ and therefore $l\mathbf{A}l^{-1}u$ belongs to the tangent space of S^3 at u.

Inspecting Table III, we find we can guarantee that $\mathbf{A}\,|\,1\rangle$ contains each diagonal irreducible representation at most once by a suitable choice of $\mathbf{A}\in\mathbb{R}^4$ in all cases but one. One normal and two tangential irreducible representations transform as II. Thus,

$$\psi_{\alpha\beta}(\mathbf{u})=\frac{1}{{}^{o}(Y')}\sum_{l\in Y'}\chi^{Y'}_{\alpha}(l)\chi^{Y'}_{\beta}(l^{-1}\mathbf{u})l\hat{\mathbf{e}}_z l^{-1}\mathbf{u} \tag{3.14}$$

produces a basis element of a tangential irreducible representation of G for all $\alpha\beta\subset T$ except $\alpha\beta=AA$ which is the normal vector yield $\mathbf{Y}_{10000}$ (breathing mode), $\alpha\beta=E_2E_2$, and $\alpha\beta=II$ for which (3.14) produces the sum of basis elements of two occurrences of II in T.

Basis elements of vector irreducible representations are the vibrational eigenstates of Polytope 120. To calculate the vibrational frequencies of the polytope we need to specify the interactions between atoms. Straley[21] performs Monte Carlo simulations of atoms on a sphere in four dimensions interacting along the chords of the sphere with the pair potential

$$\phi(s)=4s^{-12}\ . \tag{3.15}$$

The radius of the sphere is taken as $\Omega=(\sqrt{5}+1)/2$ so that the equilibrium separation of atoms is $s_0=1$. We adopt Straley's interactions to allow comparisons between our results and the simulations. With the interaction (3.15), an atom at $\mathbf{R}'\in\mathbb{R}^4$ exerts a force $\mathbf{F}(\mathbf{R}'-\mathbf{R})$ on an atom at $\mathbf{R}\in\mathbb{R}^4$, where

$$\mathbf{F}(\mathbf{R}'-\mathbf{R})=48(\mathbf{R}'-\mathbf{R})/\,|\,\mathbf{R}'-\mathbf{R}\,|^{14}\ . \tag{3.16}$$

Equation (3.16) allows easy numerical calculation of vibrational frequencies. We construct eigenstates corresponding to an irreducible representation $\alpha\beta$ using Eq. (3.14). We then displace each atom $\mathbf{u}\in Y'$ of Polytope 120 to $\mathbf{u}+\varepsilon\psi_{\alpha\beta}(\mathbf{u})$. The total force on each atom is calculated by summing Eq. (3.16) over all other atoms of the polytope. When $\psi_{\alpha\beta}(\mathbf{u})$ is an eigenstate, the total force takes the form

$$\mathbf{F}_{\varepsilon}[\psi_{\alpha\beta}]=A(\mathbf{u}+\varepsilon\psi_{\alpha\beta})+\varepsilon\lambda_{\alpha\beta}\psi_{\alpha\beta}(\mathbf{u})+O(\varepsilon^2)\ , \tag{3.17}$$

where the component proportional to $\mathbf{u}+\varepsilon\psi_{\alpha\beta}$ is normal to the sphere, and the component proportional to ψ is tangent to the sphere. Because atoms move within a sphere, a constraining force cancels the normal component of $\mathbf{F}$.

The tangential force on an atom may be expressed as an operator

$$\Phi[\psi]=\lim_{\varepsilon\to 0}\mathbf{F}_{\varepsilon}[\psi]\ , \tag{3.18}$$

where $\mathbf{F}$ denotes only the tangential part of Eq. (3.17). Clearly, $\psi_{\alpha\beta}$ are eigenvectors of Φ with eigenvalue $\lambda_{\alpha\beta}$. We can decompose ψ_{II} into two eigenvectors of Φ by writing $\psi_{II}=a(\psi_{II})_1+b(\psi_{II})_2$. Noting $\Phi^{n}[\psi_{II}]=a(\lambda^{n}_{II})_1(\psi_{II})_1+b(\lambda^{n}_{II})_2(\psi_{II})_2$, we generate four equa-

TABLE IV. Vibrational eigenvalues of Polytope 120 classified according to irreducible representation $\alpha\beta$ and degeneracy $d_{\alpha\beta}$.

$\alpha\beta$	$d_{\alpha\beta}$	$-\lambda_{\alpha\beta}$
	Longitudinal	
E_1E_1	4	508.01
F_1F_1	9	1572.67
G_1G_1	16	2638.98
HH	25	3436.11
II_1	36	1487.03
II_2	36	3810.23
G_2G_2	16	2415.68
	Transverse	
F_1A	6	0
G_1E_1	16	309.01
HF_1	30	686.01
IG_1	48	1085.67
F_2H	30	1197.34
G_2H	40	1661.68
G_2F_2	24	3836.24
E_2I	24	2572.66

tions for the four unknowns a, b, $(\lambda_{II})_1$, and $(\lambda_{II})_2$. We find $a=b=1/\sqrt{2}$. Table IV presents the spectrum of Φ classified according to irreducible representations of G. Figure 1 displays the vibrational density of states. The calculations leading to Table IV and Fig. 1 include only nearest-neighbor interactions. When long-range interactions are included the eigenvalues change by a few tenths of a percent.

In the limit of small wave numbers the polytope vibration spectrum should match the continuum elastic theory. Because of the compact topology of S^3, only a discrete set of wave numbers is allowed, so rather than take the limit as wave number goes to zero, we must work with the lowest principal quantum numbers for which the vibrational frequency is nonzero. Thus we identify F_1^L with $-\frac{1}{2}\lambda_{E_1E_1}$ and F_2^T with $-\frac{1}{2}\lambda_{G_1E_1}$. The resulting elastic constants are $\lambda=367.7$ and $\mu=177.3$.

It is interesting to note that we can drive a similar result by considering a hypothetical perfect icosahedral crystal in $\mathbf{R}^3$. If every atom in a crystal has a set of near neighbors, denoted by $\{\mathbf{R}\}$, identical to every other atom, the dynamical matrix of the crystal may be expressed[22]

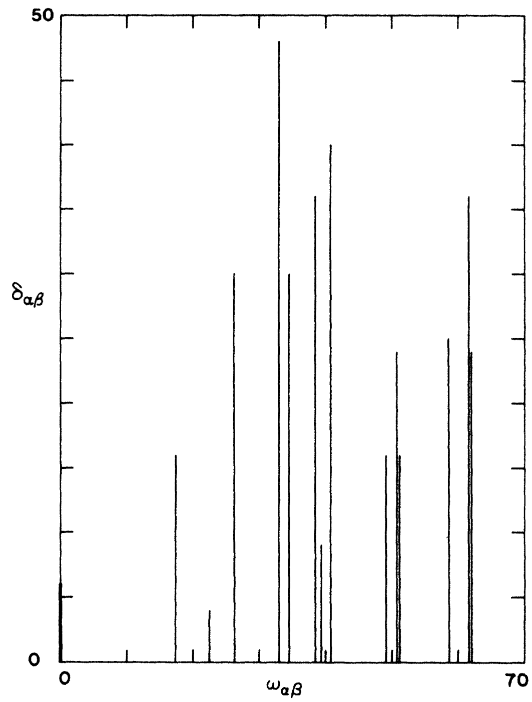

FIG. 1. Density of states of Polytope 120. $\omega_{\alpha\beta}=(-\lambda_{\alpha\beta})^{1/2}$.

$$D(\mathbf{k})=\sum_{\mathbf{R}\in\{\mathbf{R}\}}\sin^2(\tfrac{1}{2}\mathbf{k}\cdot\mathbf{R})(A\,\mathbf{1}+B\hat{\mathbf{R}}\hat{\mathbf{R}}) , \qquad (3.19)$$

where $\mathbf{1}$ is the unit matrix, $A=2\phi'(\mathbf{R})/|\mathbf{R}|$, and $B=2[\phi''(\mathbf{R})-\phi'(\mathbf{R})/|\mathbf{R}|]$. Evaluating the small wave-number limit of Eq. (3.19) in the case where $\{\mathbf{R}\}$ is the set of 12 vertices of an icosahedron yields $\lambda=364.8$ and $\mu=172.8$. In contrast, the fcc crystal has nonisotropic sound speeds leading to soft modes in certain directions.[22] These soft modes significantly reduce the cubic crystal's rigidity compared with the isotropic icosahedral crystal.

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