Phonon spectrum of metallic glasses in an icosahedral model

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The phonon spectrum of metallic glasses is analyzed for the case of an icosahedral crystal on a three-dimensional sphere: a regular {3; 3; 5} polyhedron in four-dimensional space. A dynamic operator is shown to be determined unambiguously by the symmetry and by the conditions that the energy remains constant under rotations and translations of the system as a whole. The eigenfunctions and the associated frequencies of the normal modes of atoms are found by projection onto irreducible representations of the symmetry group. The spectrum found agrees with that calculated by a recursion method for a numerical model of amorphous iron.

1. INTRODUCTION

A study of the short-range order in metallic glasses shows that a significant number of atoms have icosahedral surroundings, which lead to an energy lower than that for other groupings of 13 atoms. We know that an ideal crystal cannot be constructed from icosahedra because the point symmetry group, Y, is incompatible with lattice translations. The local surroundings of individual atoms, in contrast, can exhibit such a symmetry. This situation is encountered not only in amorphous metal but also in certain alloys of transition metals with tetrahedral close packing of atoms (Frank-Kasper phases) and in quasicrystals of the Al–Mn type. On this basis we might expect that many properties of such systems (especially those which depend on the local configuration of atoms) would be determined by the icosahedral symmetry, among other things.

The icosahedral short-range order in metallic glasses can be studied by considering a hypothetical system of atoms each of which is surrounded by 12 nearest neighbors, which form an icosahedron. In ordinary three-dimensional space R^3, such a configuration would not be possible, but it can be realized on a three-dimensional sphere S^3. As a result we obtain a regular polyhedron in four-dimensional space, which is customarily designated {3; 3; 5}, which indicates that there are five tetrahedra around each edge, and three trihedral faces converge at each vertex of these tetrahedra.

A polyhedron of this sort has been proposed[1,2] as a model of an amorphous metal in a curved space. A theory has been derived for an icosahedral order in glasses.[3] That theory treats the glasses as a result of the projection of a {3; 3; 5} polyhedron from a sphere S^3 onto a plane three-dimensional space R^3. The result is an icosahedral structure penetrated by a large number of defect lines, disclinations.[4] The structure factor calculated on this basis reproduces the experimentally observed structure factor well.[5] Calculations carried out for various properties also yield encouraging results.

In the present paper we study the dynamic properties of an icosahedral crystal. In Sec. 2 we briefly describe the structure of the polyhedron, its symmetry group G, and its irreducible representations. In Sec. 3 we determine the action G in the vibrational representation F, its nature, and the expansion of F in irreducible representations of group G. In Sec. 4 we examine the structure of the dynamic operator and the conditions which are imposed on it by the symmetry of the polyhedron. The frequencies of the normal modes are calculated in Sec. 5.

2. THE {3; 3; 5} POLYHEDRON, ITS SYMMETRY GROUP, AND ITS IRREDUCIBLE REPRESENTATIONS

The polyhedron (or polytope) of interest here has 120 vertices, which lie on the three-dimensional sphere S^3. If we make use of the well-known isomorphism between S^3 and the group SU(2) of 2×2 complex unitary matrices with a unit determinant, we can show[3] that the {3; 3; 5} vertices correspond to the elements of a discrete subgroup of SU(2): the icosahedral spinor group Y', which is the inverse transform of the ordinary icosahedral group under the mapping

$$S^3=SU(2)\to SO(3)=SU(2)/Z_2.$$

The positions of the atoms of the icosahedral crystal on S^3 can then be specified by means of the parameters of the group SU(2) (the angle φ and the unit vector $\mathbf{n}$), on the basis of the well-known formulas:

$$v=\exp\{{}^1/_2 i\mathbf{n}_v\boldsymbol{\sigma}\varphi_v\},$$

where φ_v and $\mathbf{n}_v$ are the angle and direction of the rotation axis on which v falls due to the projection $SU(2)\to SO(3)$, and σ is a formal vector constructed from Pauli matrices.

As the distance on the sphere we should use the arc length along a great circle. The distance from an atom corresponding to the element $(\mathbf{n}, \varphi)$ to the unit element $1_{Y'}$ of the Y' group is then equal to the angle φ. The arc length between the atoms corresponding to elements w and v is

$$\varphi_{w^{-1}v}=\varphi_{v^{-1}w}=2\arccos\left(\cos\frac{\varphi_v}{2}\cos\frac{\varphi_w}{2}+\mathbf{n}_v\mathbf{n}_w\sin\frac{\varphi_v}{2}\sin\frac{\varphi_w}{2}\right).$$

The 120 elements of group Y' form nine conjugate classes V_j, which are characterized by an identical angle φ and by equivalent directions of the vectors $\mathbf{n}$ (i.e., they are coupled by a symmetry transformation from Y) to the vertices, to the centers of the faces, or to the middles of edges of the icosahedron.[3] The nearest neighbors of each atom are the 12 vertices of the icosahedron which correspond to the conjugate class V_6, with an angle φ equal to $2\pi/5$ (Fig. 1).

The symmetry group of the S^3 sphere is

$$SO(4)=S^3\times SO(3)=SU(2)\times SU(2)/Z_2.$$

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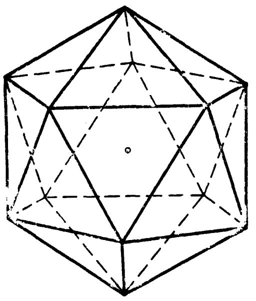

FIG. 1. Icosahedral arrangement of nearest neighbors of a central atom.

Correspondingly, for the polyhedron we have

$$G=Y\times Y'=Y'\times Y'/Z_2. \quad (2.1)$$

The elements of G are then specified by a pair of elements of Y'; $(l,r)\in G$ sends $v\in Y'$ into lvr^{-1} for l and $r\in Y'$. Furthermore, the actions of (l,r) and $(-l,-r)$ are equivalent. If an element $l\in Y'$ corresponds to $(\varphi_l,\mathbf{n}_l)$, then $(l,l)\in G$ acts on $v\in Y'$ as a rotation of $\mathbf{n}_v$ through an angle φ_l around the axis $\mathbf{n}_l$. We thus find a natural splitting of G into "rotations" of the type (l,l) and "displacements" $(1_{Y'},r^{-1})$. The order of G is 7200.

It is clear from (2.1) that the irreducible representations of the group G are the product of two irreducible representations of the group Y', which we denote by $\Gamma_\alpha^{Y'}$, $\alpha=1,\ldots,9$ (Table I). The character $(l,r)\in G$ in the representation $\Gamma_{\alpha\beta}^{G}$ is equal to the product of the characters of Y':

$$\chi_{\alpha\beta}{}^{G}(l,r)=\chi_\alpha{}^{Y'}(l)\chi_\beta{}^{Y'}(r^{-1}). \quad (2.2)$$

The equivalence of (l,r) and $(-l,-r)$ means that the following relation must hold

$$\chi_{\alpha\beta}{}^{G}(l,r)=\chi_{\alpha\beta}{}^{G}(-l,-r).$$

Since the characters of Y' are real, we find from (2.2)

$$\chi_\alpha{}^{Y'}(l)\chi_\beta{}^{Y'}(r)=\chi_\alpha{}^{Y'}(-l)\chi_\beta{}^{Y'}(-r).$$

The irreducible representations of group G are thus formed by a pair of irreducible representations of group Y' of identical parity:

$$\chi_\alpha{}^{Y'}(l)=\chi_\alpha{}^{Y'}(-l),\quad \chi_\beta{}^{Y'}(l)=\chi_\beta{}^{Y'}(-l) \quad (2.3)$$

or

$$\chi_\alpha{}^{Y'}(l)=-\chi_\alpha{}^{Y'}(-l),\quad \chi_\beta{}^{Y'}(l)=-\chi_\beta{}^{Y'}(-l).$$

3. VIBRATIONAL REPRESENTATION OF *F* AND ACTION OF GROUP *G* ON IT

The vibrational-representation space consists of all possible sets of small displacements of atoms from their equilibrium positions. To find the degree of degeneracy of the frequencies of the normal vibration modes, we need to expand F in the irreducible representations of group G (Ref. 6).

In our case, the displacement of each atom, w, is a vector in the space tangent to the sphere T_wS^3. By virtue of the group structure of Y', the basis of the tangent spaces to S^3 at all vertices {3; 3; 5} can be specified in a consistent way, in such a way that under displacements $g_v\in G{:}w\to vw$ the basis vectors T_wS^3 would be converted into the basis $T_{vw}S^3$.

Let us examine the action of an arbitrary element of G, of the form $(u,v^{-1}u)$, on the representation F. An element w transforms into $uwu^{-1}v$, and the tangent space to the sphere at the corresponding point, T_wS^3, rotates through an angle φ_u around $\mathbf{n}_u$ and is then displaced by v.

If we represent this by a 360×360 matrix, it turns out to be of block form with 3×3 cells. In the block row corresponding to w, only a single block is nonzero: that at the intersection with the block column of the element $uwu^{-1}v$.

The character of the element (l,r) in the F representation is the trace of a matrix of this sort. It is clear that only those blocks which lie on the diagonals, i.e., only those for which the relations

$$lwr^{-1}=w,\quad w^{-1}lw=r$$

hold will contribute to it.

We thus see that l and r lie in the same conjugate class. The characters of G are constant on its conjugate classes, which are formed by a pair of conjugate classes of Y'. This means that the characters of the elements (l,r) and (l,l) are identical, and since (l,l) specifies a rotation through an angle φ_l around the n_l axis they are equal to the product of the number of fixed atoms and the trace of the matrix $\Lambda_l=\Gamma_4^{Y'}(l)$ which performs this rotation:

$$\chi_F{}^{G}(l,r)=\delta_{\{l\},\{r\}}\frac{|Y'|}{|\{l\}|}\chi_4{}^{Y'}(l). \quad (3.1)$$

Here $|Y'|=120$ is the order of Y'; $|\{l\}|$ is the number of elements in the conjugate class containing l; we have $\delta_{\{l\},\{r\}}=1$ if l and r lie in the same conjugate class or $\delta_{\{l\},\{r\}}=0$ in the opposite case; and $|Y'|/|\{l\}|$ is the number of fixed points under the action of (l,l). The irreducible representation $\Gamma_4^{Y'}(l)$ specifies a natural representation of Y' by

TABLE I. Characters of the Y' group.

α	Class								
	$1V_1$	$1V_2$	$30V_3$	$20V_4$	$20V_5$	$12V_6$	$12V_7$	$12V_8$	$12V_9$
1	1	1	1	1	1	1	1	1	1
2	2	−2	0	1	−1	τ	$-\tau$	τ^{-1}	$-\tau^{-1}$
3	2	−2	0	1	−1	$-\tau^{-1}$	τ^{-1}	$-\tau$	τ
4	3	3	−1	0	0	τ	τ	$-\tau^{-1}$	$-\tau^{-1}$
5	3	3	−1	0	0	$-\tau^{-1}$	$-\tau^{-1}$	τ	τ
6	4	−4	0	−1	1	1	−1	−1	1
7	4	4	0	1	1	−1	−1	−1	−1
8	5	5	1	−1	−1	0	0	0	0
9	6	−6	0	0	0	−1	1	1	1

Note: $\tau=(\sqrt{5}+1)/2$.

rotations of the three-dimensional space. The multiplicity with which the irreducible representation of the group G, $\Gamma^G_{\alpha\beta} = \Gamma^{Y'}_\alpha \otimes \Gamma^{Y'}_\beta$, enters the expansion of the representation of F in irreducible terms is[6]

$$m(\alpha,\beta)=\frac{1}{|G|}\sum_{g\in G}\chi_F{}^G(g)\overline{\chi_{\alpha\beta}{}^G(g)}. \qquad (3.2)$$

Since the character $\chi^G_{\alpha\beta}$ is real, and by virtue of (2.2), we find

$$m(\alpha,\beta)=\frac{1}{2|G|}\sum_{l,r}\delta_{\{l\},\{r\}}\frac{|Y'|}{|\{l\}|}\chi_4{}^{Y'}(l)\chi_\alpha{}^{Y'}(l)\chi_\beta{}^{Y'}(l)$$

$$=\frac{1}{|Y'|}\sum_{l\in Y'}\chi_4{}^{Y'}(l)\chi_\alpha{}^{Y'}(l)\chi_\beta{}^{Y'}(l). \qquad (3.3)$$

Consequently, $m(\alpha,\beta)$ is equal to the multiplicity with which $\Gamma^{Y'}_4$ enters the tensor product $\Gamma^{Y'}_\alpha \otimes \Gamma^{Y'}_\beta$.

4. THE DYNAMIC MATRIX

A force matrix with elements

$$D_{\mu\nu}^{ij}=\frac{1}{M}\frac{\partial^2 E}{\partial x_i{}^\mu\,\partial x_j{}^\nu}$$

acts on F in the manner of the matrices of a representation of G, and it decomposes in a corresponding way into 3×3 blocks: $\hat{D}(w,v)$, $w, v\in Y'$. By virtue of the homogeneity of our system (invariance under displacements), the blocks depend on the relative positions of the elements w and v; i.e.,

$$\hat{D}(w,v)=\hat{D}(1_{Y'},w^{-1}v)=\hat{D}(w^{-1}v). \qquad (4.1)$$

We adopt the customary assumption (Refs. 7 and 8, for example) that the part of the force matrix which is not diagonal in the atoms is nonzero only at nearest neighbors. This assumption means that we have $\hat{D}(v)\neq 0$ only if $v = 1_{Y'}$ or $v\in V_6$.

Let us consider $\hat{D}(v)$ for $v\in V_6$, $\varphi_v = 2\pi/5$. The vectors $\mathbf{n}_v$ for $v\in V_6$ are directed to the vertices of the icosahedron. The elements of G which leave $1_{Y'}$ and v fixed must not change $\hat{D}(v)$. Since we have $\hat{D}(v)\to\hat{\Lambda}_l\hat{D}(v)\hat{\Lambda}'_l$ as $v\to lvl^{-1}$, by choosing as v an element v_0 with $\mathbf{n}_{v_0} = (0,0,1)$ and by considering rotations through angles which are multiples of $2\pi/5$ around the z axis (such rotations send the icosahedron into itself), we find

$$\hat{D}(v_0)=\begin{pmatrix} c_1 & 0 & 0\\ 0 & c_1 & 0\\ 0 & 0 & c_2\end{pmatrix}=c_1\hat{E}+(c_2-c_1)\hat{P}_{\mathbf{n}_{v_0}},$$

where $\hat{E}$ is the 3×3 unit matrix, and the projection operator $\hat{P}_\mathbf{n}$ projects onto the direction of the vector $\mathbf{n}$:

$$\hat{P}_\mathbf{n}\mathbf{a}=\mathbf{n}(\mathbf{a}\mathbf{n}).$$

To determine $\hat{D}(v)$ on an arbitrary $v\in V_6$ we need to perform a corresponding symmetry transformation (l,l) such that v_0 is sent into v. We then find

$$\hat{D}(v)=\hat{\Lambda}_l\hat{D}(v_0)\hat{\Lambda}_l^{\prime}=c_1\hat{E}+(c_2-c_1)\hat{P}_{\mathbf{n}_v}. \qquad (4.2)$$

The dynamic matrix must satisfy the condition that the energy is constant under parallel translations:

$$\sum_{v\in Y'}\hat{D}(v)=0. \qquad (4.3)$$

Under our assumptions this condition means

$$\hat{D}(1_{Y'})+\sum_{v\in V_6}\hat{D}(v)=0,$$

i.e.,

$$\hat{D}(1_{Y'})=-\Big(12c_1\hat{E}+(c_2-c_1)\sum_{v\in V_6}\hat{P}_{\mathbf{n}_v}\Big).$$

By virtue of the transformation law for a projection operator under the action of Y', a sum over a conjugate class commutes with any operator from $\Gamma^{Y'}_4$. From Shur's lemma,[9] we then have

$$\sum_{v\in V_6}\hat{P}_{\mathbf{n}_v}=\frac{|V_6|}{3}(\mathrm{tr}\hat{P}_{\mathbf{n}_v})\hat{E}=4\hat{E}, \qquad (4.4)$$

where $\hat{E}$ is the identity operator. We can then write

$$\hat{D}(1_{Y'})=-4(2c_1+c_2)\hat{E}. \qquad (4.5)$$

This result corresponds to the situation in an ordinary crystal with a point group of symmetry high enough to make the three-dimensional representation Λ irreducible. A block of the force matrix which is diagonal in the particle indices is then a scalar:

$$\frac{1}{M}\frac{\partial^2 E}{\partial x_i{}^\mu\,\partial x_i{}^\lambda}=\lambda_i\delta_{\mu\nu}.$$

5. CALCULATION OF THE NORMAL VIBRATION FREQUENCIES

We need to find the eigenvalues of the force matrix. For this purpose we should examine the effect of the dynamic operator on the characteristic functions $|\alpha\beta\rangle$, which transform under an irreducible representation of group G (Ref. 6):

$$|\alpha\beta\rangle=\frac{d_{\alpha\beta}}{|G|}\sum_{g\in G}\chi_{\alpha\beta}{}^G(g)\hat{g}|a\rangle, \qquad (5.1)$$

where $d_{\alpha\beta}$ is the dimensionality of the representation $\Gamma^G_{\alpha\beta}$, and $|a\rangle$ is an arbitrary function from F of fairly general form.

As $|a\rangle$ we choose $|\mathbf{n};1_{Y'}\rangle$: a state for which an atom at the identity of the group is displaced a small distance along the direction of the unit vector $\mathbf{n}$, while the other atoms remain at rest. We can then write

$$D|\alpha\beta\rangle=\omega_{\alpha\beta}{}^2|\alpha\beta\rangle, \qquad (5.2)$$

where $\omega_{\alpha\beta}$ is the frequency which corresponds to $\Gamma^G_{\alpha\beta}$. Using (5.1) and (2.2), we find

$$|\alpha\beta\rangle=\sum_{v\in Y'}|\hat{C}^{(v)}_{\alpha\beta}\mathbf{n};v\rangle,$$

where the operator

$$\hat{C}^{(v)}_{\alpha\beta}=\frac{d_\alpha d_\beta}{2|G|}\sum_{u\in Y'}\chi_\alpha{}^{Y'}(u)\chi_\beta{}^{Y'}(u^{-1}v)\hat{\Lambda}_u \qquad (5.3)$$

in R^3 specifies the displacement of atom v in the characteristic function of the form $|\alpha\beta\rangle$.

Relation (5.2) must hold for all the $|\alpha\beta\rangle$ components, i.e., for the displacements of all the atoms, including that corresponding to unity. We then find

$$\sum_{v\in Y'}\hat{D}(v)\hat{C}^{(v)}_{\alpha\beta}=\omega^2_{\alpha\beta}\hat{C}^{(1_{Y'})}_{\alpha\beta}. \qquad (5.4)$$

From the constancy of the characters on the conjugate classes and (5.3) we find

$$\hat{C}_{\alpha\beta}^{(1_{Y'})}=\frac{d_\alpha d_\beta}{|Y'|^2}\sum_{A\subset Y'}\chi_\alpha^{Y'}(A)\chi_\beta^{Y'}(A)\sum_{u\in A}\hat{\Lambda}_u, \qquad (5.5)$$

where A runs over all nine conjugate classes V'. The sum within a conjugate class in (5.5) commutes with all the $\hat{\Lambda}_l$. Using Schur's lemma, working by analogy with (4.4), and comparing the traces of the matrices,[9] we find

$$\sum_{u\in A}\hat{\Lambda}_u=\frac{|A|}{3}\chi_4^{Y'}(A)\hat{E}.$$

We must then have

$$\sum_{u\in Y'}\chi_\alpha^{Y'}(u)\chi_\beta^{Y'}(u)\hat{\Lambda}_u=40m(\alpha,\beta)\hat{E}, \qquad (5.6)$$

which leads to the expression

$$\hat{C}_{\alpha\beta}^{(1_{Y'})}=\frac{d_\alpha d_\beta}{360}m(\alpha,\beta)\hat{E}.$$

Relation (5.4) then becomes

$$\sum_{v\in Y'}\hat{D}(v)\hat{C}_{\alpha\beta}^{(v)}=\omega_{\alpha\beta}^2\frac{d_\alpha d_\beta}{360}m(\alpha,\beta)\hat{E}. \qquad (5.7)$$

In the case of multiple terms in the expansion of F [i.e., in the case $m(\alpha,\beta)>1$] the problem becomes slightly more complicated, because $|\alpha\beta\rangle$ is the sum of several different functions, which transform in the same way but which correspond to different eigenvalues. In this case (5.2) is replaced by

$$D|\alpha\beta\rangle=\sum_{i=1}^{m(\alpha,\beta)}\omega_{\alpha\beta(i)}^2|\alpha\beta\rangle_i, \qquad (5.8)$$

where

$$|\alpha\beta\rangle=\sum_{i=1}^{m(\alpha,\beta)}|\alpha\beta\rangle_i=\sum_{i=1}^{m(\alpha,\beta)}\sum_{v\in Y'}|\hat{C}_{\alpha\beta(i)}^{(v)}\mathbf{n};v\rangle. \qquad (5.9)$$

For a sum operator of the type (5.1), relations (5.5) and (5.7) again perform projections onto this representation. The frequencies are found from an equation analogous to (5.4):

$$\sum_{v\in Y'}\hat{D}(v)\hat{C}_{\alpha\beta}^{(v)}=\sum_{i=1}^{m(\alpha,\beta)}\omega_{\alpha\beta(i)}^2\hat{C}_{\alpha\beta(i)}^{(1_{Y'})}, \qquad (5.10)$$

where the operator

$$\hat{C}_{\alpha\beta}^{(v)}=\sum_{i=1}^{m(\alpha,\beta)}\hat{C}_{\alpha\beta(i)}^{(v)}$$

is given by expression (5.3).

In order to solve Eq. (5.10) with several unknowns we should act on $|\alpha\beta\rangle$ with a set of commuting operators $\{D^k\}$ which have identical eigenfunctions corresponding to eigenvalues $\{\omega_{\alpha\beta(i)}^{2k}\}$. If $m(\alpha,\beta)=1$, we find the following expressions for the frequencies:

$$\omega_{\alpha\beta}^2\hat{E}=\frac{1}{40}\sum_{u,v\in Y'}\chi_\alpha^{Y'}(u)\chi_\beta^{Y'}(u^{-1}v)\hat{D}(v)\hat{\Lambda}_u. \qquad (5.11)$$

The expansion of F includes two three-dimensional representations Γ_{14}^G and Γ_{41}^G, which correspond to translations and rotations of the entire polyhedron and which therefore have vanishing frequencies.

For $\alpha=4$, and $\beta=1$ we find from (4.3)

$$\frac{1}{40}\sum_{u,v\in Y'}\hat{D}(v)\chi_4^{Y'}(u)\chi_1^{Y'}(u^{-1}v)\hat{\Lambda}_u$$

$$=\frac{1}{40}\sum_u\chi_4^{Y'}(u)\hat{\Lambda}_u\sum_v\hat{D}(v)=0.$$

For $\alpha=1$ and $\beta=4$ the following condition must hold:

$$\frac{1}{40}\sum_{u,v\in Y'}\hat{D}(v)\chi_4^{Y'}(u^{-1}v)\hat{\Lambda}_u=0.$$

The change of variables $w=u^{-1}v$ yields

$$\sum_v\hat{D}(v)\hat{\Lambda}_v=0. \qquad (5.12)$$

This result could be obtained by examining the change in the energy upon a small rotation. With $w\to lwl^{-1}$, a displacement with $\varphi_l\ll 1$ gives us

$$\Delta w\approx\varphi_l\sin\varphi_w\hat{\Lambda}_w[\mathbf{n}_w\mathbf{n}_l].$$

Expanding the energy change in a series in the displacements, and equating the terms for all powers of φ_l to zero, we find condition (5.12).

Carrying out summation (5.12) for the $D(v)$ given by (4.20) and (4.5), we find

$$\sum_{v\in Y'}\hat{D}(v)\hat{\Lambda}_v=-4(2c_1+c_2)\hat{E}+4\chi_4^{Y'}(V_6)\hat{E}+4(c_2-c_1)\hat{E}.$$

We thus find $4c_1(\tau-3)=0$, where $\tau=(\sqrt{5}+1)/2$ is the golden section. In order to obtain $\omega_{14}=0$, we must require $c_1=0$.

Substituting (4.2), (4.5) and (5.6) into (5.11), we find the following expressions for the frequencies of normal vibrations corresponding to simple terms:

$$\omega_{\alpha\beta}^2\hat{E}=\hat{D}(1_{Y'})+\frac{1}{40}\Big[c_1\sum_{u,v\in Y'}\chi_\alpha^{Y'}(u)\chi_\beta^{Y'}(u^{-1}v)\hat{\Lambda}_u$$

$$+(c_2-c_1)\sum_{u,v\in Y'}\chi_\alpha^{Y'}(u)\chi_\beta^{Y'}(u^{-1}v)\hat{P}_{\mathbf{n}_v}\hat{\Lambda}_u\,.\Big] \qquad (5.13)$$

Introducing the notation

$$\sum_{v\in V_6}\chi_\beta^{Y'}(u^{-1}v)=B_\beta(u),$$

$$\sum_{v\in V_6}\chi_\beta^{Y'}(u^{-1}v)\hat{P}_{\mathbf{n}_v}=\hat{Q}_\beta(u), \qquad (5.14)$$

we can write

$$\omega_{\alpha\beta}^2\hat{E}=\hat{D}(1_{Y'})+\frac{1}{40}\Big[c_1\sum_{u\in Y'}\chi_\alpha^{Y'}(u)B_\beta(u)\hat{\Lambda}_u$$

$$+(c_2-c_1)\sum_{u\in Y'}\chi_\alpha^{Y'}(u)\hat{Q}_\beta(u)\hat{\Lambda}_u\Big]. \qquad (5.15)$$

It is easy to show (see the Appendix) that

$$B_\beta(u)=\frac{12}{d_\beta}\chi_\beta^{Y'}(V_6)\chi_\beta^{Y'}(u),$$

$$\sum_{u\in\{u\}}\hat{Q}_\beta(u)\hat{\Lambda}_u=\sum_{\gamma=1}^{9}f_\gamma^\beta\chi_\gamma^{Y'}(u)\frac{|\{u\}|}{3}\hat{E}.$$

Substituting these expressions into (5.15), transforming the sum in the last term by analogy with (5.5), and using (5.6),

TABLE II. The coefficients f_α^β of the expansion of $\mathrm{tr}(\hat{Q}_\beta(u)\hat{\Lambda}_u)$ in the characters of group Y'; the dashed correspond to representations $\Gamma_{\alpha\beta}^G$ which do not appear in the expansion of the vibrational representation.

	1	2	3	4	5	6	7	8	9
1	–	–	–	4	–	–	–	–	–
2	–	2τ	–	–	–	2τ	–	–	–
3	–	–	–	–	–	–	–	–	$-2\tau^{-1}$
4	4	–	–	$2\tau^{-1}$	–	–	–	$\frac{2\tau^2}{\sqrt{5}}$	–
5	–	–	–	–	–	–	-2τ	$\frac{4\tau^{-1}}{\sqrt{5}}$	–
6	–	2τ	–	–	–	$\frac{2\tau-5}{\sqrt{5}}$	–	–	$\frac{2\tau}{\sqrt{5}}$
7	–	–	–	–	-2τ	–	-1	$\frac{2(2-\tau)}{\sqrt{5}}$	–
8	–	–	–	$\frac{2\tau^2}{\sqrt{5}}$	$\frac{4\tau^{-1}}{\sqrt{5}}$	–	$\frac{2(2-\tau)}{\sqrt{5}}$	$\frac{-2\tau^2}{\sqrt{5}}$	–
9	–	–	$-2\tau^{-1}$	–	–	$\frac{2\tau}{\sqrt{5}}$	–	–	$\frac{2(2-3\tau)}{\sqrt{5}}$

we find expressions for the square of the frequency corresponding to a simple term in the expansion of F in irreducible representations of group G:

$$\omega_{\alpha\beta}{}^2=-4(2c_1+c_2)+\frac{12c_1}{d_\beta}\chi_\beta{}^{Y'}(V_6)+f_\alpha{}^\beta(c_2-c_1). \quad (5.16)$$

The values of f_α^β are given in Table II; their meaning is discussed in the Appendix. Since $c_1=0$, and since the force matrix is positive definite, we can write $c_2=-\omega_0^2<0$.

We finally find the following result for simple terms:

$$\omega_{\alpha\beta}=\omega_0(4-f_\alpha{}^\beta)^{1/2}.$$

In order to find two different frequencies of a unique multiple term $[m(9,9)=2]$, we need to evaluate the sum (5.7) with the square and cube of the dynamic operator.

As a result we find the system of four equations

$$S_1+S_2=(d_9d_9/360)\,m(9,9)={}^1/_5, \quad \omega_{99(1)}^2S_1+\omega_{99(2)}^2S_2=X_1,$$
$$\omega_{99(1)}^4S_1+\omega_{99(2)}^4S_2=X_2, \quad \omega_{99(1)}^6S_1+\omega_{99(2)}^6S_2=X_3, \quad (5.17)$$

where S_1 and S_2 are some matrix elements of the operators $\hat{C}_{99(1)}^{(1_{Y'})}$ and $\hat{C}_{99(2)}^{(1_{Y'})}$, and X_k are the corresponding matrix elements of the sum on the left side of (5.7) for D^k. It is easy to derive

$$X_1={}^1/_5(4-{}^1/_2f_9{}^9). \quad (5.18)$$

To find X_2 and X_3 we need to calculate $\hat{D}^2(v)$ and $\hat{D}^3(v)$ from

$$\hat{D}^2(v)=\sum_{w\in Y'}\hat{D}(w)\hat{D}'(w^{-1}v),$$
$$\hat{D}^3(v)=\sum_{w\in Y'}\hat{D}^2(w)\hat{D}'(w^{-1}v); \quad (5.19)$$

as a result they are expressed in terms of $\hat{E}$, $\hat{P}_{n_v}$, and $\hat{\Lambda}_v$.

The system (5.17) can be solved easily by switching to the new variables

$$d={}^1/_2(\omega_{99(1)}^2+\omega_{99(2)}^2), \quad \Delta d=\omega_{99(2)}^2-\omega_{99(1)}^2. \quad (5.20)$$

Using (5.18), we then find

$$d=\frac{X_3-5X_1X_2}{2(X_2-5X_1{}^2)}, \quad \left(\frac{\Delta d}{2}\right)^2=d^2-10dX_1+5X_2,$$

from which we easily find $\omega_{99(1)}$ and $\omega_{99(2)}$.

6. RESULTS AND DISCUSSION

Table III shows the normal-vibration frequencies calculated from (5.16) and (5.20), along with their degrees of degeneracy. Also shown are the irreducible representations to which they correspond. The state density is shown in Fig. 2, where each frequency corresponds to a peak, whose height is equal to the degree of degeneracy.

Significantly, the condition $c_1=0$ leads to a twofold degeneracy with respect to permutations; the effect is to dou-

TABLE III. Frequencies of the normal vibrations of the atoms of a polyhedron and degrees of degeneracy.

Frequency, in units of ω_0	Irreducible representations (α,β)	Degree of frequency degeneracy	Frequency, in units of ω_0	Irreducible representations (α,β)	Degree of frequency degeneracy
0	(1.4) and (4.1)	6	1.913	(8.7) and (7.8)	40
0.874	(2.2), (2.6) and (6.2)	20	2.188	(6.6)	16
1.288	(8.4) and (4.8)	30	2 236	(7.7)	16
1.598	(9.6) and (6,9)	48	2.288	(9.3) and (3.9)	24
1.663	(4.4)	9	2.518	(8.8)	25
1.701	(8.5) and (5.8)	30	2.669	$(9.9)_2$	36
1.851	$(9.9)_1$	36	2.690	(5.7) and (7.5)	24

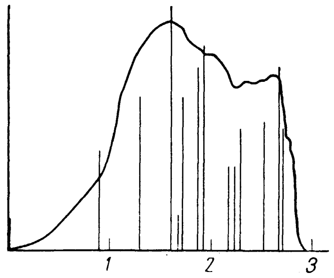

FIG. 2. Spectrum of a dynamic operator on a {3; 3; 5} polyhedron in comparison with the frequency density of the normal vibrations of atoms in a numerical model of amorphous iron[11] (solid line). The frequencies are expressed in arbitrary units ω_0. The height of each peak is proportional to the degree of degeneracy of the corresponding frequency of the normal mode of the polyhedron.

ble the weight of the frequencies corresponding to the nondiagonal representations $\Gamma^G_{\alpha\beta}$, i.e., corresponding to representations in which we have $\alpha \neq \beta$.

When an amorphous medium is constructed from icosahedral blocks by means of "projections," the low-frequency part of the spectrum should undergo the greatest change. This part of the spectrum if formed primarily by long-wave excitations of the acoustic-phonon type. On a polyhedron, such excitations cannot be reproduced because of the finite number of atoms, but they are totally insensitive to the structure. In the Debye model they are determined by the macroscopic elastic constants.

In contrast, the high-frequency part of the spectrum, which includes most of the frequencies according to the estimates of Ref. 10, depends on the short-range order, i.e., on local atomic configurations, which are modeled quite well by an icosahedral crystal. The considerations are supported by a comparison of the calculated spectrum with the spectral density of the vibrations of atoms which was obtained in Ref. 11 by the method of continued fractions for a numerical model of amorphous iron (containing 500 atoms). The spectrum is shown in Fig. 2. Despite the diversity of spectra, we see that the qualitative features are reproduced satisfactorily.

After this work had been completed, I learned of Ref. 12, where similar results were obtained by another method: an explicit construction of eigenstates from the basis functions of irreducible representations of the SO(4) group, hyperspherical harmonics. The approach of the present paper appears to be more physical, since it makes more extensive use of the "crystallography" of a polyhedron. As a result, the entire spectrum is determined by a single force parameter and is calculated analytically. There is no need to resort to a numerical solution of an equation like (5.2) for a binary potential of a particular type. The quasimomentum concept which was introduced by Widom[12] seems extraneous since this quantity is conserved only in the long-wave region, where the frequency is insensitive to the local structure, upon a projection of the polytope onto R^3.

I sincerely thank A. Ya. Belen'kiĭ and A. L. Roĭtburd for useful discussions of these questions.

APPENDIX

Calculation of $B_\beta(u)$ and $\hat{Q}_\beta(u)$. We will first show that $B_\beta(u)$ is a central function of group Y', i.e., that it is constant on the conjugate classes:

$$B_\beta(wuw^{-1}) = \sum_{v\in V_6} \chi_\beta^{Y'}(wu^{-1}w^{-1}v).$$

Since the character $\chi_\beta^{Y'}$ is not changed by a cyclic permutation of the factors (like the trace of the corresponding matrix), we find

$$B_\beta(wuw^{-1}) = \sum_{v\in V_6} \chi_\beta^{Y'}(u^{-1}w^{-1}vw). \tag{A1}$$

Clearly, when v runs over V_6, $w^{-1}vw$ also runs over all of V_6, and the sum does not change:

$$B_\beta(wuw^{-1}) = B_\beta(u). \tag{A2}$$

As a central function, $B_\beta(u)$ can be expanded in a linear combination of characters[13] (a Fourier transformation):

$$B_\beta(u) = \sum_{\zeta=1}^{9} b_\zeta^\beta \chi_\zeta^{Y'}(u). \tag{A3}$$

The expansion coefficients b_ζ^β are determined by the inverse Fourier transformation:

$$b_\zeta^\beta = \frac{1}{|Y'|}\sum_{u\in Y'} B_\beta(u)\overline{\chi_\zeta^{Y'}(u)}$$
$$= \sum_{v\in V_6}\Big[\frac{1}{|Y'|}\sum_{u\in Y'}\chi_\beta^{Y'}(u^{-1}v)\chi_\zeta^{Y'}(u)\Big] = \sum_{v\in V_6}[\chi_\beta^{Y'} * \chi_\zeta^{Y'}](v), \tag{A4}$$

where the asterisk indicates convolution of functions.

The coefficients of a Fourier expansion of the convolution of two functions in the matrix elements of irreducible representations of a group are known[14] to be equal to the product of the corresponding coefficients for the functions being convolved. We can thus write

$$b_\zeta^\beta = \frac{12}{d_\beta}\delta_{\beta\zeta}\chi_\zeta^{Y'}(V_6),$$

$$B_\beta(u) = \sum_{\zeta=1}^{9} b_\zeta^\beta \chi_\zeta^{Y'}(u) = \frac{12}{d_\beta}\chi_\beta^{Y'}(V_6)\chi_\beta^{Y'}(u). \tag{A5}$$

To evaluate the sum

$$\sum_{\{u\}} \hat{Q}_\beta(u)\hat{\Lambda}_u = \sum_{u\in\{u\}}\sum_{v\in V_6} \chi_\beta^{Y'}(u^{-1}v)\hat{P}_{n_v}\hat{\Lambda}_u \tag{A6}$$

we need to study how the terms transform upon conjugation. It is easy to show that we have

$$\hat{Q}_\beta(wuw^{-1}) = \sum_{v\in V_6}\chi_\beta^{Y'}(wu^{-1}w^{-1}v)\hat{P}_{n_v} = \hat{\Lambda}_w\hat{Q}_\beta(u)\hat{\Lambda}_w'. \tag{A7}$$

Since $\hat{\Lambda}_u$ transforms in the same way, the sum over the conjugate class of the product $\hat{Q}_\beta(u)\hat{\Lambda}_u$ commutes with any operator $\Gamma_4^{Y'}$, and from Schur's lemma[9] it is

$$\sum_{u\in\{u\}} \hat{Q}_\beta(u)\hat{\Lambda}_u = \frac{|\{u\}|}{3}\operatorname{tr}(\hat{Q}_\beta(u)\hat{\Lambda}_u)\hat{E}. \tag{A8}$$

By virtue of the transformation law for $\hat{Q}_\beta(u)\hat{\Lambda}_u$ under conjugations, the function $\operatorname{tr}(\hat{Q}_\beta(u)\hat{\Lambda}_u)$ is constant on the conjugate classes and can be written as a linear combination of characters:

$$\mathrm{tr}(\hat{Q}_\beta(u)\hat{\Lambda}_u)=\sum_{\gamma=1}^{9} f_\gamma^\beta \chi_\gamma^{Y'}(u), \tag{A9}$$

where the expansion coefficients

$$f_\gamma^\beta \hat{E}=\frac{1}{40}\sum_{u\in Y'}\chi_\gamma^{Y'}(u)\hat{Q}_\beta(u)\hat{\Lambda}_u$$

are the same, to within a factor $c_2 - c_1$, as the last term in the sum in (5.15).

The central function $\mathrm{tr}(\hat{Q}_\beta(u)\hat{\Lambda}_u)$ is found by direct calculation from (5.14), with allowance for the symmetry of the icosahedron; this symmetry facilitates the derivation of $\hat{Q}_\beta(u)$. Expansion (A9) is found from the table of characters of Y' (Table I).

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