becomes

$$\langle \delta U(\mathbf{r}_1)\delta U(\mathbf{r}_2)\rangle = (2\pi)^{-6} \iint d\mathbf{k}_1\, d\mathbf{k}_2 \exp(i\mathbf{k}_1\cdot\mathbf{r}_1)\exp(i\mathbf{k}_2\cdot\mathbf{r}_2) \times u(\mathbf{k}_1)u(\mathbf{k}_2)S(\mathbf{k}_1,\mathbf{k}_2)\sum_{\mathbf{l}} \exp[-i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{l}], \quad (A6)$$

where the correlation function $S$ is given by

$$S(\mathbf{k}_1,\mathbf{k}_2) = \exp[-M(\mathbf{k}_1+\mathbf{k}_2)^2] - \exp(-Mk_1^2)\exp(-Mk_2^2). \quad (A7)$$

In both examples discussed in the paper the $f$ functions of ($A1$) take the form of plane waves and so we write

$$f_1(\mathbf{r}_1) = \exp(-i\mathbf{q}_1\cdot\mathbf{r}_1), \quad f_2(\mathbf{r}_2) = \exp(i\mathbf{q}_2\cdot\mathbf{r}_2). \quad (A8)$$

When ($A6$) is substituted into ($A1$) we obtain

$$\mathcal{I} = u(\mathbf{q}_1)u^*(\mathbf{q}_2)S(\mathbf{q}_1-\mathbf{q}_2)\sum_{\mathbf{l}} \exp[i(\mathbf{q}_2-\mathbf{q}_1)\cdot\mathbf{l}] \quad (A9)$$

which is the basic result for the matrix elements. It only remains to look at the sum in ($A9$), which covers all lattice sites in the crystal. It follows that the transverse parts, $\mathbf{Q}_1$ and $\mathbf{Q}_2$, of $\mathbf{q}_1$ and $\mathbf{q}_2$ must be equal to within a two-dimensional reciprocal-lattice vector, $\mathbf{G}$. The sum in the $z$ direction is however only over the finite thickness of the crystal. Provided $(q_2)_z - (q_1)_z$ is small (as it is in practice) the sum can be replaced by an integral to yield

$$(1/l_z)\int_0^t dz \exp[i(q_{2z}-q_{1z})z] = (t/l_z)\exp[i(q_{2z}-q_{1z})t/2] \times \sin[(q_{2z}-q_{1z})t/2]/[(q_{2z}-q_{1z})t/2], \quad (A10)$$

where $l_z$ is the repeat distance in the $z$ direction. It is this oscillatory thickness-dependent factor which controls the degree of coherence between different states. If $q_{1z} = q_{2z}$, ($A10$) gives a contribution which increases linearly with $t$. However, if $q_{1z}$ and $q_{2z}$ are different the $(\sin x)/x$ factor in ($A10$) falls off with increasing thickness, which indicates a reduction in the off-diagonal elements of the density matrix and a correspondingly weaker coherence.

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# Orientational Parameters of *n*-Dimensional Symmetry Operations

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## Abstract

The number of orientational parameters is evaluated for a general point-symmetry operation in $n$ dimensions. When the operation contains orthogonal identical crypto-components some of the parameters become free and this phenomenon is investigated.

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## 1. Introduction

It is a commonplace that combinations of orthogonal mirror reflections give rise to symmetry operations (twofold rotation, centre of inversion *etc.*) whose effects are independent of the orientation of their constituent (crypto-) mirrors. When the detailed geometric effects of double rotations were worked out in four dimensions it was found that some of these were partially independent of the orientation of their crypto-rotation planes (Whittaker, 1984). The evidence for this came primarily from a geometrical representation of the symmetry in hyperstereograms, but in the case of the double fourfold rotation (symbolized **44** or **IV**) it was substantiated algebraically. If the crypto-rotation planes are supposed to lie on the planes of the *wx* and *yz* axes, matrices for the two 90° rotations multiply together to give the matrix of the **IV** operation as

$$\begin{pmatrix} 0 & -1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \end{pmatrix} = \begin{pmatrix} 0 & -1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \end{pmatrix}. \quad (1)$$

However, the same product is obtained by multiplying together

$$\frac{1}{2}\begin{pmatrix} 1-\cos p\cos q & -1-\cos p\cos q & \sin p-\cos p\sin q & \sin p+\cos p\sin q \\ 1+\cos p\cos q & 1-\cos p\cos q & -\sin p-\cos p\sin q & \sin p-\cos p\sin q \\ \sin p+\cos p\sin q & \sin p-\cos p\sin q & 1+\cos p\cos q & -1+\cos p\cos q \\ -\sin p+\cos p\sin q & \sin p+\cos p\sin q & 1-\cos p\cos q & 1+\cos p\cos q \end{pmatrix} \quad (2)$$

and

$$\frac{1}{2}\begin{pmatrix} 1+\cos p\cos q & -1+\cos p\cos q & -\sin p+\cos p\sin q & -\sin p-\cos p\sin q \\ 1-\cos p\cos q & 1+\cos p\cos q & \sin p+\cos p\sin q & -\sin p+\cos p\sin q \\ -\sin p-\cos p\sin q & -\sin p+\cos p\sin q & 1-\cos p\cos q & -1-\cos p\cos q \\ \sin p-\cos p\sin q & -\sin p-\cos p\sin q & 1+\cos p\cos q & 1-\cos p\cos q \end{pmatrix}. \quad (3)$$

These two matrices* also represent 90° rotations about a pair of orthogonal planes whose orientation depends on the free variables $p$ and $q$. Since the orientation of a plane (and therefore of a pair of orthogonal planes) in four dimensions requires four parameters to specify it (Whittaker, 1985, p. 46), it is evident that the effect of the **IV** operation is independent of at least two out of the four parameters that would be expected to be required to specify its orientation.

The above factorization of the **IV** operation was performed in an *ad hoc* manner that could not easily be extended to operations of order other than 4, and the meaning of the parameters $p$ and $q$ was not ascertained. However, Weigel, Veysseyre, Phan, Effantin & Billiet (1984) showed more generally that, for any four-dimensional symmetry operation involving two orthogonal rotations through equal angles on the *wx* and *yz* planes, the matrix is invariant under a transformation of the axes by a double rotation through equal arbitrary angles (say $\theta$) parallel to the planes *wy* and *xz*. They therefore concluded that the two (crypto-) 'planes of rotation are not unique but belong to a one-parameter family of pairs of orthogonal planes'. They did not observe that the invariance also exists for a further transformation by a double rotation through different equal arbitrary angles (say $\varphi$) parallel to the planes *xy* and *zw*, and that the crypto-planes of rotation therefore belong to a two-parameter family, at least. Furthermore, it can be shown that the two parameters $\theta$, $\varphi$ introduced in this analysis can be related to those appearing in the factorization of the **IV** operation (1) above by the relations

$$p=2\theta \quad \text{and} \quad q=-2\varphi.$$

It is to be noted that the **IV** operation (1) was of the type $\mathbf{IV}_+$ (Whittaker 1990), and the double-rotation transformations of the axes in that case must both be of negative hand. For an invariant transformation of a $\mathbf{IV}_-$ operation they must both be of positive hand, and the two matrix factors in that case may be obtained from (2) and (3) by multiplying both the right-hand column and the bottom row by −1, thereby leaving the bottom term of the diagonal unchanged.

## 2. Partitioning of *n* dimensions

Following Weigel *et al.* (1984) we may express a general point-symmetry operation in $n$ dimensions on a suitable basis in terms of a unimodular matrix

* The first of the two matrices was given by Whittaker (1984) but contained errors in the signs of some of the terms. Both matrices were given correctly by Whittaker (1985).

$\mathbf{M}$ of the form

$$\mathbf{M}=\begin{pmatrix} 1 & 0 & \dots & 0 & 0 & \dots & 0 & 0 & 0 & 0 & 0 & 0 & \dots \\ 0 & 1 & \dots & 0 & 0 & \dots & 0 & 0 & 0 & 0 & 0 & 0 & \dots \\ \vdots & \vdots & \ddots & \vdots & \vdots & \dots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \dots \\ 0 & 0 & \dots & -1 & 0 & \dots & 0 & 0 & 0 & 0 & 0 & 0 & \dots \\ 0 & 0 & \dots & 0 & -1 & \dots & 0 & 0 & 0 & 0 & 0 & 0 & \dots \\ \vdots & \vdots & \dots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \dots \\ 0 & 0 & \dots & 0 & 0 & \dots & & \mathbf{N}_1 & 0 & 0 & 0 & 0 & \dots \\ 0 & 0 & \dots & 0 & 0 & \dots & & & 0 & 0 & 0 & 0 & \dots \\ 0 & 0 & \dots & 0 & 0 & \dots & 0 & 0 & & \mathbf{N}_2 & 0 & 0 & \dots \\ 0 & 0 & \dots & 0 & 0 & \dots & 0 & 0 & & & 0 & 0 & \dots \\ 0 & 0 & \dots & 0 & 0 & \dots & 0 & 0 & 0 & 0 & & \mathbf{N}_3 & \dots \\ 0 & 0 & \dots & 0 & 0 & \dots & 0 & 0 & 0 & 0 & & & \dots \\ \vdots & \vdots & \dots & \vdots & \vdots & \dots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \end{pmatrix} \quad (4)$$

where each $2\times2$ matrix $\mathbf{N}_i$ is given by

$$\mathbf{N}_i=\begin{pmatrix} \cos 2\pi\, q_i/p_i & -\sin 2\pi\, q_i/p_i \\ \sin 2\pi\, q_i/p_i & \cos 2\pi\, q_i/p_i \end{pmatrix}$$

and represents a rotation of order $p_i$ ($>2$).

The $n$ dimensions may then be partitioned for our purpose into four sets:

(i) $f$ dimensions corresponding to the number of $+1$s on the diagonal of $\mathbf{M}$;

(ii) $g$ dimensions corresponding to the number of $-1$s on the diagonal of $\mathbf{M}$, being the number of orthogonal crypto-mirror operations;

(iii) $2H$ dimensions containing subsets of $2h_j$ dimensions, each of which supports a multiple rotation containing $h_j$ identical crypto-rotations having the same values of $p_i$ ($>2$) and $|q_i|$;

(iv) $2k$ dimensions supporting a multiple rotation involving $k$ crypto-rotations all differing from one another and from those involved in (iii) in respect of $p_i$ ($>2$) or $|q_i|$ or both.

Any one, two or three of these sets may be absent.

The problem then consists in finding the number of parameters required to define the external orientation of each of the sets, so defined, that are present, and then any additional parameters required to define the internal orientations of the crypto-components within the sets. When all the sets but one have been oriented, no further external parameters are required since the last set is defined as the complement of the totality of the others.

The orientation of an $m$-dimensional subspace through the origin is defined if $m$ linearly independent points in it are specified. In $n$ dimensions each of these points has $n$ coordinates, making $mn$ parameters. However, once the subspace has been defined its orientation is unaffected by free movement of the points within it, so that each of the $m$ points has $m$ free parameters. Thus the number of parameters required to specify the orientation of a partition of $n$ dimensions into two conjugate subspaces of $m$ and $(n-m)$ dimensions is $m(n-m)$. In particular the orientation of a plane in $n$-dimensional space is specified by $2(n-2)$ parameters.

It is clear that when we have partitioned $m$ out of $n$ dimensions, any further partitioning is to be applied to the remaining $(n-m)$ dimensions. Thus if we partition out a set of $2h$ dimensions followed by a set of $2k$ dimensions the total number of orientational parameters is

$$\begin{aligned} &2h(n-2h)+2k(n-2h-2k) \\ &\quad=2n(h+k)-4(h^2+hk+k^2). \end{aligned}$$

The symmetry of this result shows that the order of the partitioning is irrelevant.

## 3. Internal orientation

### 3.1. *k crypto-rotations all different* ($p_i>2$)

It is convenient to treat this case first, although in defining the set of $k$ different crypto-rotations in the previous section it was necessary to do so after removing from consideration any rotation which occurs more than once in the whole operation.

Since the crypto-rotations are all different the orientation of the operation clearly requires the plane supporting each one to be defined unequivocally. Thus the problem is simply that of partitioning successive planes from the set of $2k$ dimensions. This proceeds as in the previous section, and the number of parameters is given by $2(2k-2)+2(2k-4)+\ldots+2[2k-2(k-1)]=2k(k-1)$.

### 3.2. *h crypto-rotations all identical* [*with identical* $p_i$ ($>2$) *and* $|q_i|$]

The relevant portion of the matrix $\mathbf{M}$ containing $h$ $2\times2$ matrices on the diagonal, identical but for the sign of $q_i$, may be written

$$\mathbf{M}_1=\begin{pmatrix} \cos\alpha_1 & -\sin\alpha_1 & 0 & 0 & \dots & 0 & 0 \\ \sin\alpha_1 & \cos\alpha_1 & 0 & 0 & \dots & 0 & 0 \\ 0 & 0 & \cos\alpha_2 & -\sin\alpha_2 & \dots & 0 & 0 \\ 0 & 0 & \sin\alpha_2 & \cos\alpha_2 & \dots & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \dots & \cos\alpha_h & -\sin\alpha_h \\ 0 & 0 & 0 & 0 & \dots & \sin\alpha_h & \cos\alpha_h \end{pmatrix}$$

where $\alpha_i=2\pi q_i/p_i$, and varies only in its sign. This transforms an arbitrary unit vector

$$\mathbf{v}=(c_1, c_2, c_3, \ldots, c_{2h})$$

to a vector

$$\begin{aligned} \mathbf{v}'=(&c_1\cos\alpha_1-c_2\sin\alpha_1,\ c_1\sin\alpha_1 \\ &+c_2\cos\alpha_1,\ c_3\cos\alpha_2 \\ &-c_4\sin\alpha_2, \ldots, c_{2h-1}\sin\alpha_h+c_{2h}\cos\alpha_h) \end{aligned}$$

and the angle between $\mathbf{v}$ and $\mathbf{v}'$ is given by

$$\begin{aligned}\cos\theta &= c_1^2\cos\alpha_1 - c_1c_2\sin\alpha_1 + c_1c_2\sin\alpha_1\\ &\quad + c_2^2\cos\alpha_1 + \ldots + c_{2h}^2\cos\alpha_h\\ &= \cos\alpha\end{aligned}$$

where $\alpha = |\alpha_i|$.

Thus the vector is turned through $2\pi|q_i|/p_i$ by each application of the operations, and after $p_i$ applications it coincides with itself, so all the images are coplanar. Their plane ($\Pi$) is therefore a possible supporting plane of one of the crypto-rotations of the operation.

In order to consider the orientation of $\Pi$ we note that it is spanned by any two linear combinations of $\mathbf{v}$ and $\mathbf{v}'$, and we take $(c_2 - c_1\cot\alpha_1)\mathbf{v} + c_1\operatorname{cosec}\alpha_1\mathbf{v}'$ and $(c_1 + c_2\cot\alpha_1)\mathbf{v} - c_2\operatorname{cosec}\alpha_1\mathbf{v}'$ whose components after renormalizing are of the form $(0, C_1, C_2, C_3, \ldots, C_{2h-2}, C_{2h-1})$ and $(C_1, 0, C_3, -C_2, \ldots, C_{2h-1}, -C_{2h-2})$ with $\sum_{i=1}^{2h-1} C_i^2 = 1$. The orientation of the plane $\Pi$ is thus specified by $2(h-1)$ parameters, whereas a general plane in $2h$-dimensional space requires $4(h-1)$ parameters to specify its orientation uniquely. Thus $\Pi$ belongs to a set of planes specified by $2(h-1)$ parameters and having $2(h-1)$ free parameters.

Conjugate to the plane $\Pi$ is a $(2h-2)$-dimensional subspace containing the remaining $(h-1)$ supporting planes. The parameters required to specify each of these in turn can be considered in the same way, so that the total number of internal parameters of the $h$-tuple rotation is given by $2(h-1) + 2(h-2) + \ldots + 2 = h(h-1)$. This is half the number that would be required if the rotations were all different, as shown in § 3.1, and the remaining $h(h-1)$ parameters are free.

### 3.3. *g orthogonal crypto-reflections*

In this case the relevant part of the matrix $\mathbf{M}$ is

$$\mathbf{M}_2 = \begin{pmatrix} -1 & 0 & 0 & \ldots & 0\\ 0 & -1 & 0 & \ldots & 0\\ 0 & 0 & -1 & \ldots & 0\\ \vdots & \vdots & \vdots & \ddots & 0\\ 0 & 0 & 0 & \ldots & -1\end{pmatrix}$$

and it is evident that the symmetry has no internal orientational parameters since every vector $\mathbf{v}$ is transformed to $-\mathbf{v}$. That is, $\mathbf{v}$ and its image $\mathbf{v}'$ are collinear and are related by a reflection operation in a one-dimensional subspace having no fixed parameters; its orientational parameters are all free and are defined by $\mathbf{v}$, just as half of the parameters of the plane in which $\mathbf{v}$ was rotated were found to be free and defined by $\mathbf{v}$ in § 3.2.

## 4. Discussion

Although the properties of the matrix $\mathbf{M}$ are such that it is not necessary to analyse symmetry operations in terms of any crypto-components more complex than rotations, the conclusions regarding free parameters can be illuminated by considering an analysis in terms of double rotations. For example, in eight dimensions one could consider the operation $\mathbf{5}^1\mathbf{5}^2\mathbf{8}^1\mathbf{8}^3$ as the dual double rotation **V VIII**.* The parameters involved would be 16 for the partitioning into two four-dimensional subspaces, making 24 in all. The whole operation would repeat an arbitrary vector to span the whole eight-dimensional space. On the other hand a dual double rotation with two identical crypto-components such as **VV** can be rearranged from $\mathbf{5}^1\mathbf{5}^2 \cdot \mathbf{5}^1\mathbf{5}^2$ to $\mathbf{5}^1\mathbf{5}^1 \cdot \mathbf{5}^2\mathbf{5}^2$. Again the partitioning into two four-dimensional subspaces requires 16 parameters, but the orientation of the double equal rotation within each only requires two, making 20 in all and leaving 4 parameters free. Furthermore we see from the rearranged form that a vector operated on by $\mathbf{5}^1\mathbf{5}^1$ is rotated through $2\pi/5$ in a plane and is then rotated through $4\pi/5$ in an orthogonal plane by the $\mathbf{5}^2\mathbf{5}^2$ operation. The vector is therefore repeated to the vertices of a pentatope just as if it were operated on by a single **V** in an appropriate (not completely determined) orientation. The initial vector and its four images spans only a four-dimensional subspace whose orientation varies with the initial vector and this variable subspace can be regarded as a possible support of one of the two crypto-**V** operations.

The purpose of this discussion is to point out the generality of the phenomenon that when identical crypto-components are repeated a vector and its images span a subspace of the same dimensionality as if only one of the cryptocomponents were acting alone in a subspace containing the vector: if the repeated crypto-components are one-dimensional mirror operations the subspace is one-dimensional; if they are rotations it is a plane; if they are double rotations it is four-dimensional, and so on. Whenever this occurs some orientational parameters become free - in the first case all of them, in the second case half of them, and in more complex cases a fraction less than half. This fraction can be evaluated but is of little interest because such situations can be fully treated in terms of repeated component crypto-rotation planes.

## 5. Concluding remarks

The number of parameters required to specify the orientation of any $n$-dimensional point-symmetry

* The superscript numerals in $\mathbf{5}^1\mathbf{5}^2$ *etc.* denote the values of $|q_i|$ in the crypto-rotation components.

operation has been derived. For four-dimensional operations involving a pair of identical rotations two, and only two, of the expected four orientational parameters are free parameters. This confirms what has hitherto been believed but not proven.

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# Interference of X-ray Diffraction Trajectories in a Strained Crystal Undergoing Ultrasonic Excitation

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### Abstract

The X-ray optics in a strained single crystal under ultrasonic excitation are considered. The anomalous behavior of diffraction intensity, depending on sound amplitude, is analyzed. The interference of X-rays moving along different trajectories is demonstrated, which leads to a new type of *Pendellösung* effect, depending on the strain gradient and sound frequency. Experimental data agree with the theoretical predictions.

## 1. Introduction

The problem of ultrasonic influence on X-ray and neutron diffraction is now under intensive investigation. The most interesting phenomena arise when the magnitude of the ultrasound wave vector $k$ is of the order of the gap $\Delta k_0$ between the branches of the dispersion surface (DS) (in the two-beam approximation). In this case diffraction is of a multiwave nature, since, together with the nodes 0 and $H$ of the reciprocal lattice, the points $\pm mk$ and $H \pm mk$ will be located near the Ewald sphere (Entin, 1979). Formally, if

$$k > \Delta k_0 \quad (1)$$

one speaks about the creation of an ultrasonic superlattice, which strongly modifies the eigenstates of diffracted quanta inside the crystal. Such ultrasound we will call here a high-frequency wave.

Interaction between modified Bloch states by means of high-frequency ultrasonic perturbation leads to new physical effects, such as the resonant ultrasonic suppression of the Borrmann transmission (Entin, 1977), the ultrasound-induced *Pendellösung* beatings in diffraction intensity (Iolin, Zolotoyabko, Raitman, Kuvaldin & Gavrilov, 1986; Entin & Puchkova, 1984) and anomalous behavior of diffraction intensity in elastically deformed crystals in the presence of acoustic waves (Iolin, Raitman, Kuvaldin & Zolotoyabko, 1988).

The latter effect consists of a substantial decrease (up to 50%) in the diffraction intensity $I$ at small sound amplitudes $w$ ($Hw < 1$, where $H$ is the magnitude of the reciprocal-lattice vector), in contrast to the intensity growth in a thin nondistorted crystal undergoing ultrasonic excitation. Such curves were first obtained in a neutron diffraction experiment and were theoretically explained by E. Iolin in terms of the violation of the adiabaticity condition for quanta movement, taking into account the inelastic multiphonon scattering (Iolin, 1987).

Moreover, an additional *Pendellösung* effect was predicted, due to the interference of waves travelling along different trajectories inside the distorted crystal under ultrasonic excitation. In contrast with the well known results for elastically strained crystals without ultrasound (Kato, 1964; Hart, 1966), the new *Pendellösung* effect reveals itself in the form of diffraction intensity oscillations, measured at definite sound amplitudes. The oscillation period depends on the strain gradient $b$ and sound frequency $\nu$ (more precisely on the parameter $k/\Delta k_0$). Preliminary results in this field, obtained with both X-ray and neutron beams, were reported at the Twelfth European Crystallographic Meeting (Iolin, Zolotoyabko, Raitman & Kuvaldin, 1989).

Here we present the detailed data concerning X-ray diffraction in elastically strained crystals undergoing high-frequency ultrasound.

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