Acta Crystallographica Section A
Foundations of Crystallography
ISSN 0108-7673

Received 23 June 2008
Accepted 29 July 2008

Bijective mapping between the eigenvalue spectrum of a Karle–Hauptman matrix and its phases

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For any particular Karle–Hauptman matrix, a bijective mapping exists between the eigenvalue spectrum of the matrix and the set of structure-factor phases, given enough phase constraints. For a matrix of order $n + 1$ with no symmetry-equivalent reflections, $n + 1$ phases need to be fixed. Only a small subset of matrices derived from centric reflections from trigonal or hexagonal space groups have a bijective mapping without any fixed phases.

1. Introduction

The matrix A below is a Karle–Hauptman matrix of order $(n + 1)$ (Karle & Hauptman, 1950). The matrix comprises a set of structure factors of which the top-row reflections are arbitrary but completely determine the rest. The matrix is Hermitian, thus the set of $(n + 1)$ eigenvalues are real; the ordered set of these is the eigenvalue spectrum.

$$A = \begin{pmatrix} E_0 & E_1 & E_2 & E_3 & \cdots & E_n \\ E_{\bar{1}} & E_0 & E_{2-1} & E_{3-1} & \cdots & E_{n-1} \\ E_{\bar{2}} & E_{1-2} & E_0 & E_{3-2} & \cdots & E_{n-2} \\ \vdots & & & & \ddots & \\ E_{\bar{n}} & & \cdots & & & E_0 \end{pmatrix}.$$

See Table 1 for definitions of the symbols in this paper. For an equal-atom structure with N atoms, it can be shown that there will be N nonzero eigenvalues and $(n+1) - N$ zero eigenvalues (van der Plas *et al.*, 1998*a*). Using the properties of the eigenvalues or determinant, phase sets can be refined (de Gelder, de Graaf & Schenk, 1993; van der Plas *et al.*, 1998*b*; Tsoucaris, 1970). When refining phases in this manner it is important to ensure that multiple phase sets do not generate the same set of eigenvalues.

This paper examines the necessary constraints in order to ensure a bijective mapping between the eigenvalue spectrum of a KH and the included structure-factor phases. By this we mean that given a particular set of phases (which conform to the constraints) the eigenvalue spectrum is unique, *i.e.* no other phase set gives the same spectrum. Conversely, given an appropriate eigenvalue spectrum the set of phases is determined uniquely. The eigenvalue spectrum is related to the electron density but it is not necessarily the same, therefore in general the constraints are not just fixing the origin and enantiomorph.

2. Matrix transforms

2.1. Eigenvalue-invariant transformations

Let A and B be square matrices of the same order with $\sigma(A) = \sigma(B)$. B can be transformed onto A using the similarity transform

$$P^{-1}BP = A. \qquad (1)$$

For Hermitian matrices it is also true that $\sigma(A^T) = \sigma(A)$. No other type of transformation needs to be considered.

2.2. Similarity transform

If A and B are KHs of the same order and are constructed from the same structure factors, and we assume that the magnitudes are fixed, any valid ST must maintain each magnitude. If the matrix is positive-definite then the columns and rows of the matrix are linearly independent. This means that the general form of P must be diagonal. Given these constraints, the matrix C below will be the general form of P (de Gelder, Elout *et al.*, 1993; Ralph, 1991).

$$C = \begin{pmatrix} \exp(i\varphi_0) & 0 & \cdots & 0 & 0 \\ 0 & \exp(i\varphi_1) & \cdots & 0 & 0 \\ 0 & 0 & \exp(i\varphi_2) & \cdots & 0 \\ \vdots & & & \ddots & \\ 0 & 0 & \cdots & 0 & \exp(i\varphi_n) \end{pmatrix}.$$

Applying C as an ST using equation (1),

$$C^{-1}AC = \begin{pmatrix} E_0 & E_1\exp(i\Phi_{10}) & E_2\exp(i\Phi_{20}) & \cdots & E_n\exp(i\Phi_{n0}) \\ \cdots & E_0 & E_{2-1}\exp(i\Phi_{21}) & \cdots & E_n\exp(i\Phi_{n1}) \\ \vdots & & & \ddots & \\ \cdots & & & & E_0 \end{pmatrix}.$$

Each φ_i is arbitrary thus generating an infinite number of different matrices with the same $\sigma(A)$. To limit the STs to the identity only, all Φ's need to be set to zero. This can be achieved by fixing a number of phases.

Let $\mathcal{R} = \{\Phi_{ij} : i, j \in \{0, \cdots, n\}; i > j\}$ which contains $n(n-1)/2$ elements. These $\Phi_{ij} \in \mathcal{R}$ are associated with the reflections above the

Table 1
Nomenclature and abbreviations.

$E_{k-j} = E[(h(k) - h(j)]$	normalized structure factors
θ_{k-j}	the phase of the reflection E_{k-j}
$h(0)$	the origin reflection
$\alpha, \varphi_i, \Phi_{ij}$	reals modulo 2π
k_i	have the values ± 1
$\Phi_{ij} = \varphi_i - \varphi_j$	each Φ_{ij} is associated with E_{i-j}
A, B, C, P	square matrices of order $(n + 1)$
$\sigma(A)$	the eigenvalue spectrum of matrix A
ST	similarity transform
KH	Karle–Hauptman matrix

leading diagonal. Some elements of $\mathcal{R}$ can be combined to form others, using the equations below.

$$\begin{aligned} \Phi_{ik} + \Phi_{kj} &= \varphi_i - \varphi_k + \varphi_k - \varphi_j = \Phi_{ij}, \\ \Phi_{ki} + \Phi_{jk} &= \varphi_k - \varphi_i + \varphi_j - \varphi_k = -\Phi_{ij}, \\ \Phi_{ik} - \Phi_{jk} &= \varphi_i - \varphi_k - (\varphi_j - \varphi_k) = \Phi_{ij}, \\ \Phi_{kj} - \Phi_{ki} &= \varphi_k - \varphi_j - (\varphi_k - \varphi_i) = \Phi_{ij}. \end{aligned} \quad (2)$$

Let $\mathcal{T}$ be a subset of $\mathcal{R}$ for which all elements of $\mathcal{R}$ can be generated by those in $\mathcal{T}$ using equations (2). In order to set each $\Phi_{ij} = 0$ it is sufficient to set each $\varphi_i = \varphi_j = \alpha$. Each Φ represents an equation involving φ's and the set of those obtained from $\mathcal{T}$ form a set of linearly independent equations. As each φ needs to be set to the same value, resulting in a one-dimensional solution set, only n equations would be needed to solve for $(n + 1)$ unknowns, as stated in de Gelder, Elout *et al.* (1993). This translates into $\mathcal{T}$ having exactly n elements.

There is at least one choice for $\mathcal{T}$, which is $\mathcal{T} = \{\Phi_{i0} \in \mathcal{R} : i \in \{1, \ldots, n\}\}$. However, for orders greater than two, this is not the only choice. For example for a 4 × 4 KH, $\mathcal{R} = \{\Phi_{10}, \Phi_{20}, \Phi_{30}, \Phi_{21}, \Phi_{31}, \Phi_{32}\}$ and $\mathcal{T}$ could be $\{\Phi_{10}, \Phi_{20}, \Phi_{30}\}$ or $\{\Phi_{21}, \Phi_{32}, \Phi_{10}\}$. However, it is clear that $\mathcal{T} \neq \{\Phi_{10}, \Phi_{20}, \Phi_{21}\}$.

Fixing the phase of E_{i-j} implies that E_{i-j} is unchanged after applying a valid ST, hence $\Phi_{ij} = 0$. Setting

$$\Phi_{ij} = 0 \quad \forall \Phi_{ij} \in \mathcal{T} \;\Rightarrow\; \Phi_{ij} = 0 \quad \forall \Phi_{ij} \in \mathcal{R}. \quad (3)$$

This is true because all members of $\mathcal{R}$ are generated by those in $\mathcal{T}$. Such an ST would be equivalent to the identity. Setting elements of $\mathcal{T}$ equal to multiples of 2π would also lead to the identity transformation.

2.3. Transpose

Taking the transpose of a KH reverses the sign of all phases, hence swapping the enantiomorph but leaving $\sigma(A)$ unchanged. This transformation is distinct from an ST and fixing phases from reflections associated with $\mathcal{T}$ is not sufficient to give a bijective mapping. In most cases applying a nontrivial ST to the transpose will keep the fixed phases invariant but change the others that were not fixed, hence giving two phase sets that generate the same spectrum. In this case fixing an additional acentric phase, to those required by §2.2, is required. An obvious exception to this is when all the associated reflections in $\mathcal{R}\backslash\mathcal{T}$ are real, *i.e.* phases are limited to 0 or π.

3. Symmetry

The use of symmetry-related reflections (instead of fixing phases) to limit STs has already been considered (de Gelder, Elout *et al.*, 1993). However, the question remains as to what conditions are necessary to guarantee a bijective mapping.

If there are two symmetry-related reflections E_{i-j} and E_{k-l}, then $\Phi_{ij} = \Phi_{kl}$ or $\Phi_{ij} = -\Phi_{kl}$ (Bijvoet opposite). Any number of these relationships is not sufficient for a bijective mapping. A set of symmetry-related Φ_{ij} can be limited if at least two are in $\mathcal{T}$ and a third is related through equations (2). For example, assume that E_{i-k}, E_{k-j} and E_{i-j} are symmetry equivalents and that $\Phi_{ik}, \Phi_{kj} \in \mathcal{T}$. From equations (2) we have that $\Phi_{ik} + \Phi_{kj} = \Phi_{ij}$. After applying an ST, the phases of the symmetry equivalents are shifted by $\pm\alpha$, implying the following:

$$k_1\Phi_{ik} = k_2\Phi_{kj} = k_3\Phi_{ij} = \alpha, \text{ where } k_i = \pm 1, \quad (4)$$

$$\Rightarrow (k_1 + k_2 - k_3)\alpha = 0 \pmod{2\pi}. \quad (5)$$

If $k_1 + k_2 - k_3 = \pm 1$, equation (5) has only one solution *i.e.* $\alpha = 0$, making a bijective mapping possible. Given the conditions above, E_{i-k}, E_{k-j} and E_{i-j} must be symmetry equivalents and with the correct Bijvoet sign. The only Laue groups which fit these criteria are $\overline{3}m$, $6/m$ and $6/mmm$. For example, reflections 110, $\overline{1}20$ and $\overline{2}10$ from a $P321$ structure.

The next case to consider is that E_{i-k}, E_{k-l}, E_{l-j} and E_{i-j} are all symmetry equivalents and that $\Phi_{ik}, \Phi_{kl}, \Phi_{lj} \in \mathcal{T}$. Thus equation (5) would be modified to give

$$(k_1 + k_2 + k_3 - k_4)\alpha = 0 \pmod{2\pi}.$$

This type of relationship will never lead to a bijective mapping because $k_1 + k_2 + k_3 - k_4 \neq \pm 1$, thus there will always be more than one solution. The same will be true for all relationships that have an even number of Φ's.

The next scenario could be that E_{ik}, E_{kl}, E_{lm}, E_{mj} and E_{ij} are symmetry equivalents. Also that all the associated Φ's are in $\mathcal{T}$ except Φ_{ij}. The symmetry relationships derived from the corresponding equations to (4) and (5) are only satisfied by the same three Laue groups. Although not all possible combinations of equations in (2) and sets of Φ have been tested, it is assumed that the symmetry restrictions are the same as above.

To complete the necessary constraints to ensure a bijective mapping using symmetry only, each set of symmetry-related reflections must form relationships like those described above and $\mathcal{T}$ must have a full compliment of n members. The above constraints still might not be sufficient for a bijective mapping. From §2.3 the matrix transpose must also be considered. Thus to achieve a bijective mapping, from symmetry constraints alone, the KH must be real and satisfy the above conditions.

4. Example

Below is a KH matrix, of order five, constructed from an artificial structure in $P2_12_12_1$. The reflection indices are shown for the upper triangle of the matrix.

$$\begin{pmatrix} 000 & 111 & \overline{111} & 333 & \overline{333} \\ & 000 & \overline{220} & 222 & \overline{442} \\ & & 000 & 442 & \overline{222} \\ & & & 000 & \overline{660} \\ & & & & 000 \end{pmatrix}$$

If we examine the symmetry equivalents in the matrix, we get the following:

$$\begin{aligned} \Phi_{10} = \Phi_{20} = \alpha, \quad \Phi_{30} = \Phi_{40} = \beta, \\ \Phi_{32} = \Phi_{41} = \gamma, \quad \Phi_{31} = \Phi_{42} = \delta, \end{aligned}$$

where $\alpha, \beta, \gamma, \delta$ are reals modulo 2π. Using equations (2), we can find relationships between the Φ's:

$$\begin{aligned} &\Phi_{20} - \Phi_{10} = \Phi_{21} = 0, \\ &\Phi_{40} - \Phi_{30} = \Phi_{43} = 0, \\ &\Phi_{42} - \Phi_{32} = \Phi_{43} = 0 \Rightarrow \gamma = \delta, \\ &\Phi_{40} - \Phi_{20} = \Phi_{42} \Rightarrow \beta - \alpha = \delta. \end{aligned}$$

From this we can deduce that the phases for $\overline{220}$ and $\overline{660}$ are fixed but there are an infinite number of possible phase sets generating the same spectrum.

Table 2 shows different possible phase sets that give the same eigenvalue spectrum. The solutions were verified numerically by starting from random phases (except for those that were fixed) and

Table 2
Different possible phase sets that give the same eigenvalue spectrum.

Phase sets are shown which have the same eigenvalue spectrum as the reference one, given the indicated fixed phases.

Case	Fixed phases	Phases for (111, 333, 442, 222, 220, 660)
1	None	320, 70, 180, 30, 0, 180
		160, 180, 90, 300, 0, 180 plus others
2	111, 333	340, 350, 80, 290, 0, 180†
		240, 250, 300, 90, 0, 180
3	111, 222	340, 350, 80, 290, 0, 180†
		340, 190, 140, 290, 0, 180
4	222, 422	230, 240, 80, 290, 0, 180
		120, 130, 80, 290, 0, 180 plus others
5	111, 333, 222	340, 350, 80, 290, 0, 180†

† The correct phase set.

refining phases by minimizing the current to the reference spectrum, over many trials. One particular phase set was used to generate the reference spectrum; this phase set is referred to as the correct set.

Case 1, where no phases were fixed, verifies that there is more than one matrix with the same spectrum. From §2.2, four matrix elements need to be fixed to limit the STs to the identity. As there are four pairs of symmetry-related reflections, fixing two phases should be sufficient. In cases 2 and 3 the associated Φ's form a complete set $\mathcal{T}$, thus the STs are limited. However, in each case there was an additional solution to the correct one. This confirms the assertion in §2.3 that a transform of the transpose will also give the same spectrum. Case 4 shows that although the correct number of matrix elements were fixed, this does not limit the STs. Case 5 shows that a bijective mapping is possible by fixing three phases in this case.

5. Discussion

It has been shown here that it is possible to generate a bijective mapping by fixing reflections. For a KH with no symmetry-equivalent reflections then $n + 1$ phases need to be fixed in a matrix of order $(n + 1)$. It is clear from the example that arbitrarily selecting phase constraints does not necessarily lead to a bijective mapping. Sections 2.2 and 2.3 provide a method to ensure a bijective mapping by fixing matrix elements associated with $\Phi_{ij} \in \mathcal{T}$ and possibly one other. The patterns of matrix elements that form $\mathcal{T}$ are the same for any KH of a particular order.

The inclusion of symmetry-related reflections in a KH complicates matters. It is clear from §3 that only in a small number of cases will symmetry by itself be able to ensure a bijective mapping. For most KHs at least one phase must be fixed. This can be verified by replacing the top-row reflections in the example by 111, $\bar{1}\bar{1}1$, $\bar{1}1\bar{1}$, $1\bar{1}\bar{1}$. In general there is no way of predicting the exact number of phases that must be fixed.

If the eigenvalue spectrum of a particular KH could be predicted *a priori* and sufficient phase constraints are applied to give a bijective mapping, then all other phases would be determined uniquely. Such a scenario could lead to a phase-extension method.

I thank the staff of the Mathematics Department and ICARUS, NUI Maynooth, Ireland, and Professors P. Main and M. M. Woolfson, University of York, England. Special thanks go to Dr D. Dickinson, NUI Maynooth, Ireland.

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