HAUPTMAN, H. & KARLE, J. (1957). *Acta Cryst.* **10**, 267.
HUGHES, E. W. (1953). *Acta Cryst.* **6**, 871.
KARLE, I., HAUPTMAN, H., KARLE, J. & WING, A. B. (1957). *Acta Cryst.* **10**, 481.
KARLE, J. & HAUPTMAN, H. (1950). *Acta Cryst.* **3**, 181.
KENDALL, M. G. (1943). *The Advanced Theory of Statistics*, vol. 1. London: Griffin.
KITAIGORODSKI, A. I. (1954). *Travaux de l'Institut de Cristallographie* (Communications au IIIe. Congres International de Cristallographie). Moscou: Academie des Sciences de l'U.R.S.S.
SAYRE, D. (1952). *Acta Cryst.* **5**, 60.
SAYRE, D. (1953). *Acta Cryst.* **6**, 430.
SIMERSKÁ, M. (1956). *Czech. J. Phys.* **6**, 1.
SLACK, M. (1946). *J. Inst. Elec. Eng.* **93** (Part III), 76.
VAND, V. & PEPINSKY, R. (1953). *The Statistical Approach to X-ray Structure Analysis*. State College: X-ray and Crystal Analysis Laboratory of the Pennsylvania State University.
VAND, V. & PEPINSKY, R. (1954). *Acta Cryst.* **7**, 451.
WICKSELL, S. D. (1917). *Kungl. Sv. Vet. Akad. Handl.* **58**, Nr. 3.
WOOLFSON, M. M. (1954). *Acta Cryst.* **7**, 61.
WOOLFSON, M. M. (1957). *Acta Cryst.* **10**, 116.

Acta Cryst. (1958). **11**, 543

Matrix-Theoretical Derivation of Inequalities

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Using the matrix theory, a general type of inequalities for $P1$ and $P\bar{1}$ is derived. The results obtained for $P1$ are the same as those derived by Karle & Hauptman.

1. Introduction

Since Harker & Kasper (1948) derived inequalities among the structure factors, the same problem has been examined by many authors. Based upon the fact that the electron densities are non-negative, Karle & Hauptman (1950) derived some general limitations to be imposed upon the relations among the structure factors. Using their method, Bouman (1956) derived a complete set of fundamental inequalities for the structure factors possessing a centre of symmetry. The present paper will show that the same relations can be derived in a more compact form by means of the matrix representation of the Fourier transformation between the structure factors and electron densities.

2. Representation in matrix form

For the sake of simplicity we shall treat the one-dimensional case, though the method can be extended to the cases of more than one dimension without encountering any great difficulty. The electron density $\varrho(x)$ in the unit of the length L is represented by

$$\varrho(x) = \frac{1}{L}\sum_{h=-\infty}^{+\infty} F_h \exp[2\pi i h x], \quad h = 0, \pm1, \pm2, \ldots; \quad -\tfrac{1}{2} \le x \le \tfrac{1}{2}. \quad (1)$$

Let us divide our unit cell into $(2N+1)$ equal ranges, $\Delta \equiv L/(2N+1)$. Then the following formula approaches to equation (1) asymptotically with increase of the number of division:

$$\varrho\left(\frac{r}{2N+1}\right) = \frac{1}{(2N+1)\Delta}\sum_{h=-N}^{+N} F_h \exp\left[2\pi i h \frac{r}{2N+1}\right], \quad h, r = -N, \ldots, 0, \ldots, N. \quad (2)$$

Defining

$$\varrho_r \equiv \varrho\left(\frac{r}{2N+1}\right)\Delta,$$

(2) is replaced by

$$\varrho_r = \frac{1}{2N+1}\sum_{h=-N}^{+N} F_h \exp\left[2\pi i \frac{hr}{2N+1}\right], \quad h, r = -N, \ldots, 0, \ldots, N. \quad (3)$$

The Fourier series (3) can now be represented in terms of a $(2N+1)$-dimensional matrix product as follows:

$$\boldsymbol{\varrho} = \boldsymbol{U}\boldsymbol{F}\boldsymbol{U}^{-1}, \quad (4)$$

where

$$\boldsymbol{\varrho} = \begin{bmatrix} \varrho_{-N} & & & & & & \\ & \ddots & & & & & \\ & & \varrho_{-r} & & 0 & & \\ & & & \ddots & & & \\ & & & & \varrho_0 & & \\ & & 0 & & & \varrho_r & \\ & & & & & & \ddots \\ & & & & & & & \varrho_N \end{bmatrix}, \quad (5)$$

$$F=\frac{1}{2N+1}\begin{bmatrix} F_0 & F_{-1} & \dots & F_{-N} & F_N & \dots & F_2 & F_1 \\ F_1 & F_0 & \dots & F_{-N+1} & F_{-N} & \dots & F_3 & F_2 \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ F_{-1} & F_{-2} & \dots & F_N & F_{N-1} & \dots & F_1 & F_0 \end{bmatrix} \tag{6}$$

and

$$U=\frac{1}{\sqrt{(2N+1)}}\begin{bmatrix} \omega^{N^2} & \omega^{N(N-1)} & \dots & \omega^{-N^2} \\ \omega^{(N-1)N} & \omega^{(N-1)^2} & \dots & \omega^{-(N-1)N} \\ \dots & \dots & \dots & \dots \\ \omega^{-N^2} & \omega^{-N(N-1)} & \dots & \omega^{N^2} \end{bmatrix}$$

$$\left(\omega=\exp\left[\frac{2\pi i}{2N+1}\right]\right). \tag{7}$$

ϱ, F and $U=\tilde{U}^{-1}$ are the diagonal, cyclic and unitary matrices respectively.

3. The derivation of a general type of inequality for *P*1

The expression (4) indicates the fact that the cyclic matrix F composed of the structure factors has the eigenvalues ϱ_r when it is diagonalized by the unitary transformation U. Now, the diagonal matrix ϱ is Hermitian and non-negative because the eigenvalues are all real and non-negative. Therefore, the matrix F is also Hermitian and non-negative by virtue of the well known fact that the Hermitian and non-negative characters of any matrix are invariant under unitary transformation. The postulate for F to be Hermitian corresponds to the well known Friedel law:

$$F_h=F^*_{-h}\,. \tag{8}$$

On the other hand, the non-negative character of F directly leads to the following inequalities:

$$\begin{vmatrix} F_{h_1-h_1} & F_{h_1-h_2} & \dots & F_{h_1-h_n} \\ F_{h_2-h_1} & F_{h_2-h_2} & \dots & F_{h_2-h_n} \\ \dots & \dots & \dots & \dots \\ F_{h_n-h_1} & F_{h_n-h_2} & \dots & F_{h_n-h_n} \end{vmatrix} \geq 0 \quad (n=1,2,3,\dots)\,. \tag{9}$$

Relation (9) is nothing but what Karle & Hauptman derived for *P*1.

4. The derivation of a general type of inequality for $P\bar{1}$

We now consider the case where the electron density $\varrho(x)$ or ϱ_r show certain kinds of symmetry as demanded by a space group. The matrix ϱ will be invariant under a symmetry operation T:

$$\varrho=T\varrho T^{-1}\,. \tag{10}$$

Operating the unitary transformation U on the expression (10), we obtain

$$F=RFR^{-1},\quad \text{where}\quad R=U^{-1}TU\,. \tag{11}$$

Thus we see F is invariant under the transformation R. Therefore, we can reduce our matrix F to the direct sum of partial matrices which belong respectively to different eigenvalues of R, because F and R are commutative with each other.

In the case of centrosymmetric structures $\varrho_r=\varrho_{-r}$, T is found to be as follows:

$$T=T^{-1}=\begin{bmatrix} 0 & & & 1 \\ & & 1 & \\ & \cdot^{\cdot^{\cdot}} & & \\ 1 & & & 0 \end{bmatrix} \quad \text{and} \quad T^2=1\,. \tag{12}$$

Then R is

$$R=U^{-1}TU=T=R^{-1} \quad \text{and} \quad R^2=1\,. \tag{13}$$

The formulae (11) and (13) show that the matrix F is doubly symmetric. This corresponds to the well known result:

$$F_h=F_{-h}\,, \tag{14}$$

which holds in the case of centrosymmetric structures. Corresponding to the two eigenvalues ± 1 of R given by (13), F is reduced to the direct sum of two partial matrices F^+ and F^-. Namely, in such a representation where R is diagonal,

$$VRV^{-1}=\left[\begin{array}{ccccc|ccccc} 1 & & & & 0 & & & & & \\ & 1 & & & & & & & & \\ & & \ddots & & & & & 0 & & \\ & & & 1 & & & & & & \\ 0 & & & & 1 & & & & & \\ \hline & & & & & -1 & & & & 0 \\ & & & & & & -1 & & & \\ & & 0 & & & & & \ddots & & \\ & & & & & & & & -1 & \\ & & & & & 0 & & & & -1 \end{array}\right] \begin{array}{l} \}\ \text{Dimension } N+1 \\ \\ \}\ \text{Dimension } N \end{array}, \tag{15}$$

$$V=V^{-1}=1/\sqrt{2}\begin{bmatrix} 1 & & & & & & 1 \\ & 1 & & 0 & & 1 & \\ & & \ddots & & \cdot^{\cdot^{\cdot}} & & \\ & & 1 & & 1 & & \\ & 0 & & \sqrt{2} & & 0 & \\ & & 1 & & -1 & & \\ & \cdot^{\cdot^{\cdot}} & & & & \ddots & \\ & 1 & & 0 & & -1 & \\ 1 & & & & & & -1 \end{bmatrix}, \tag{16}$$

and

$$VFV^{-1}=\left[\begin{array}{c|c} F^+ & 0 \\ \hline 0 & F^- \end{array}\right] \begin{array}{l} \}\ \text{Dimension } N+1 \\ \}\ \text{Dimension } N \end{array} \tag{17}$$

where

$$
\boldsymbol{F}^{+}=\frac{1}{2N+1}\begin{bmatrix}
F_0+F_1 & F_1+F_2 & F_2+F_3 & \cdots & F_{N-3}+F_{N-2} & F_{N-2}+F_{N-1} & F_{N-1}+F_N & \sqrt{2}F_N \\
F_1+F_2 & F_0+F_3 & F_1+F_4 & \cdots & F_{N-4}+F_{N-1} & F_{N-3}+F_N & F_{N-2}+F_N & \sqrt{2}F_{N-1} \\
F_2+F_3 & F_1+F_4 & F_0+F_5 & \cdots & F_{N-5}+F_N & F_{N-4}+F_N & F_{N-3}+F_{N-1} & \sqrt{2}F_{N-2} \\
\cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\
F_{N-3}+F_{N-2} & F_{N-4}+F_{N-1} & F_{N-5}+F_N & \cdots & F_0+F_6 & F_1+F_5 & F_2+F_4 & \sqrt{2}F_3 \\
F_{N-2}+F_{N-1} & F_{N-3}+F_N & F_{N-4}+F_N & \cdots & F_1+F_5 & F_0+F_4 & F_1+F_3 & \sqrt{2}F_2 \\
F_{N-1}+F_N & F_{N-2}+F_N & F_{N-3}+F_{N-1} & \cdots & F_2+F_4 & F_1+F_3 & F_0+F_2 & \sqrt{2}F_1 \\
\sqrt{2}F_N & \sqrt{2}F_{N-1} & \sqrt{2}F_{N-2} & \cdots & \sqrt{2}F_3 & \sqrt{2}F_2 & \sqrt{2}F_1 & F_0
\end{bmatrix} \quad (18)
$$

and

$$
\boldsymbol{F}^{-}=\frac{1}{2N+1}\begin{bmatrix}
F_0-F_2 & F_1-F_3 & F_2-F_4 & \cdots & F_{N-3}-F_{N-1} & F_{N-2}-F_N & F_{N-1}-F_N \\
F_1-F_3 & F_0-F_4 & F_1-F_5 & \cdots & F_{N-4}-F_N & F_{N-3}-F_N & F_{N-2}-F_{N-1} \\
F_2-F_4 & F_1-F_5 & F_0-F_6 & \cdots & F_{N-5}-F_N & F_{N-4}-F_{N-1} & F_{N-3}-F_{N-2} \\
\cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\
F_{N-3}-F_{N-1} & F_{N-4}-F_N & F_{N-5}-F_N & \cdots & F_0-F_5 & F_1-F_4 & F_2-F_3 \\
F_{N-2}-F_N & F_{N-3}-F_N & F_{N-4}-F_{N-1} & \cdots & F_1-F_4 & F_0-F_3 & F_1-F_2 \\
F_{N-1}-F_N & F_{N-2}-F_{N-1} & F_{N-3}-F_{N-2} & \cdots & F_2-F_3 & F_1-F_2 & F_0-F_1
\end{bmatrix}. \quad (19)
$$

Since $\boldsymbol{V}$ is orthogonal, (17) is also Hermitian and non-negative. Let us consider the case $N\rightarrow\infty$. F_h is practically zero when h is very large. Therefore, from (17), (18) and (19), the following inequalities will be obtained:

$$
\begin{vmatrix}
F_{(h_1-h_1)/2}\pm F_{(h_1+h_1)/2} & F_{(h_1-h_2)/2}\pm F_{(h_1+h_2)/2} & \cdots & F_{(h_1-h_n)/2}\pm F_{(h_1+h_n)/2} \\
F_{(h_2-h_1)/2}\pm F_{(h_2+h_1)/2} & F_{(h_2-h_2)/2}\pm F_{(h_2+h_2)/2} & \cdots & F_{(h_2-h_n)/2}\pm F_{(h_2+h_n)/2} \\
\cdots & \cdots & \cdots & \cdots \\
F_{(h_n-h_1)/2}\pm F_{(h_n+h_1)/2} & F_{(h_n-h_2)/2}\pm F_{(h_n+h_2)/2} & \cdots & F_{(h_n-h_n)/2}\pm F_{(h_n+h_n)/2}
\end{vmatrix} \geq 0 \quad (n=1,2,3,\ldots), \quad (20)
$$

where $(h_i\pm h_j)/2$ are all integers. Relation (20) is a general type of inequality for the structures possessing a centre of symmetry.

The determinants of the first degree of (20) give rise to the inequalities

$$\text{(I)} \qquad F_0\pm F_{h_1}\geq 0\,.$$

The determinants of the second degree,

$$
\begin{vmatrix}
F_0\pm F_{h_1} & F_{(h_1-h_2)/2}\pm F_{(h_1+h_2)/2} \\
F_{(h_2-h_1)/2}\pm F_{(h_2+h_1)/2} & F_0\pm F_{h_2}
\end{vmatrix} \geq 0 \quad (21)
$$

include all of the well known results of Harker & Kasper in the case of centrosymmetric structures. Putting $h_2\equiv 0$, $h_1\equiv 2h$ in (21), we can find the non-trivial inequality

$$\text{(II)} \qquad F_0(F_0+F_{2h})\geq 2F_h^2\,.$$

For even h_1 and even h_2, by putting $h_1\equiv 2h$ and $h_2\equiv 2h'$, we obtain

$$\text{(III)} \qquad (F_0\pm F_{2h})(F_0\pm F_{2h'})\geq(F_{h-h'}\pm F_{h+h'})^2\,,$$

and for odd h_1 and odd h_2, by putting $(h_1-h_2)/2\equiv h$ and $(h_1+h_2)/2\equiv h'$, we obtain

$$\text{(IV)} \qquad (F_0\pm F_{h+h'})(F_0\pm F_{h-h'})\geq(F_h\pm F_{h'})^2\,.$$

The third-degree determinants of (20) include the only non-trivial one, found by de Wolf & Bouman (1954), with respect to the two indices h_1 and h_2. This is obtained for the case $h_3\equiv 0$, $h_1\equiv 2h$, $h_2\equiv 2h'$ and for plus sign:

$$
\begin{vmatrix}
F_0+F_{2h} & F_{h-h'}+F_{h+h'} & 2F_h \\
F_{h-h'}+F_{h+h'} & F_0+F_{2h'} & 2F_{h'} \\
2F_h & 2F_{h'} & 2F_0
\end{vmatrix} \geq 0 \quad (22)
$$

or

$$\text{(V)} \qquad [F_0(F_0+F_{2h})-2F_h^2][F_0(F_0+F_{2h'})-2F_{h'}^2] \geq [F_0(F_{h-h'}+F_{h+h'})-2F_hF_{h'}]^2\,.$$

(I)–(V) are in accordance with the inequalities of Bouman (1956).

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References

Bouman, J. (1956). *Acta Cryst.* **9**, 777.
Harker, D. & Kasper, J. S. (1948). *Acta Cryst.* **1**, 70.
Karle, J. & Hauptman, H. (1950). *Acta Cryst.* **3**, 181.
Wolf, P. M. de & Bouman, J. (1954). *Acta Cryst.* **7**, 328.