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Matrix Theoretical Derivation of Inequalities. II.

By T. Oda

Osaka University of Liberal Arts and Education, Tennoji-ku, Osaka, Japan

and S. Naya

Department of Physics, Kinki University, Fuse, Osaka, Japan

and I. Taguchi*

Faculty of Science, Osaka University, Nakanosima, Osaka, Japan

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The present paper shows a way starting from a matrix-theoretical representation of Fourier series and leading to the reduction by the symmetry properties of the matrix of Karle and Hauptman (1950) type, so that a general form of fundamental inequalities involving structure factors can be derived for any given space group.

It has been reported (Taguchi & Naya, 1958) that when the Karle & Hauptman matrix is slightly modified by the cyclic matrix, the matrix $\boldsymbol{F}$ can be diagonalized completely, so that the characteristic numbers may correspond to the electron densities; namely,

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

where $\boldsymbol{\varrho}$ and $\boldsymbol{F}$ are the following N-dimensional matrices with N large:

$$\boldsymbol{\varrho} = \begin{bmatrix} \varrho_0 & & & & 0 \\ & \ddots & & & \\ & & \varrho_r & & \\ & & & \ddots & \\ 0 & & & & \varrho_{N-1} \end{bmatrix},$$

$$\boldsymbol{F} = \frac{1}{N} \begin{bmatrix} F_0 & F_{-1} & F_{-2} & \cdot & \cdot & F_2 & F_1 \\ F_1 & F_0 & F_{-1} & \cdot & \cdot & F_3 & F_2 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ F_{-1} & F_{-2} & F_{-3} & \cdot & \cdot & F_1 & F_0 \end{bmatrix}, \quad (2)$$

$\boldsymbol{U}$ being a unitary matrix used in the transformation. Introducing the regular representation for the cyclic group of order N,

$$\boldsymbol{1}, \boldsymbol{C}, \ldots, \boldsymbol{C}^h, \ldots, \boldsymbol{C}^{N-1}$$

$$\boldsymbol{C} = \begin{bmatrix} 0 & \cdot & \cdot & \cdot & \cdot & 0 & 1 \\ 1 & 0 & \cdot & \cdot & \cdot & \cdot & 0 \\ 0 & 1 & \ddots & & & & \cdot \\ \cdot & 0 & \ddots & \ddots & & & \cdot \\ \cdot & \cdot & & \ddots & \ddots & & \cdot \\ \cdot & \cdot & & & 1 & \ddots & \cdot \\ 0 & 0 & \cdot & \cdot & 0 & 1 & 0 \end{bmatrix} \quad \boldsymbol{C}^N = \boldsymbol{1}\,, \quad (3)$$

$\boldsymbol{F}$ can be written as

$$\boldsymbol{F} = \frac{1}{N} \sum_{h=0}^{N-1} F_h \boldsymbol{C}^h\,. \quad (4)$$

We shall extend equation (4) for the one-dimensional case to the three-dimensional one. The resultant expression becomes to be

$$\boldsymbol{F} = \frac{1}{N^3} \sum_{h=0}^{N-1} \sum_{k=0}^{N-1} \sum_{l=0}^{N-1} F_{hkl} \boldsymbol{C}^h \times \boldsymbol{C}^k \times \boldsymbol{C}^l = \frac{1}{N^3} \sum_{\mathbf{h}=0}^{N-1} \boldsymbol{F}_{\mathbf{h}} \boldsymbol{C}^{\mathbf{h}}\,, \quad (5)$$

* Deceased, 26 May 1959.

where $\boldsymbol{C}^{\mathbf{h}}$ is the N^3-dimensional matrix defined by a direct product of three matrices $\boldsymbol{C}^h$, $\boldsymbol{C}^k$, $\boldsymbol{C}^l$:

$$\boldsymbol{C}^{\mathbf{h}} \equiv \boldsymbol{C}^h \times \boldsymbol{C}^k \times \boldsymbol{C}^l. \tag{6}$$

Now, using the Kronecker symbol δ, the element of matrix $\boldsymbol{C}^h$ in row h_i, column h_j can be expressed from (3) as

$$C^h_{h_i, h_j} = \delta_{h, h_i - h_j} \pmod N , \tag{7}$$

h_i and h_j being $0, 1, 2, \ldots, N-1$ respectively. Hence, taking the $\mathbf{h}_i = (h_i, k_i, l_i)$, $\mathbf{h}_j = (h_j, k_j, l_j)$ element of matrix $\boldsymbol{C}^{\mathbf{h}}$ as

$$\begin{aligned} C^{\mathbf{h}}_{\mathbf{h}_i, \mathbf{h}_j} &= (C^h \times C^k \times C^l)_{(h_i, k_i, l_i)(h_j, k_j, l_j)} = C^h_{h_i, h_j} \cdot C^k_{k_i, k_j} \cdot C^l_{l_i, l_j} \\ &= \delta_{h, h_i - h_j} \cdot \delta_{k, k_i - k_j} \cdot \delta_{l, l_i - l_j} = \delta_{\mathbf{h}, \mathbf{h}_i - \mathbf{h}_j} , \end{aligned} \tag{8}$$

the elements of matrix $\boldsymbol{F}$ in (5) are written as

$$N^3 F_{\mathbf{h}_i, \mathbf{h}_j} = \sum_{\mathbf{h}=0}^{N-1} F_{\mathbf{h}} \cdot C^{\mathbf{h}}_{\mathbf{h}_i, \mathbf{h}_j} = \sum_{\mathbf{h}=0}^{N-1} F_{\mathbf{h}} \cdot \delta_{\mathbf{h}, \mathbf{h}_i - \mathbf{h}_j} = F_{\mathbf{h}_i - \mathbf{h}_j} . \tag{9}$$

From the non-negativity of the matrix $\boldsymbol{F}$ and using relation (9), it follows

$$\begin{vmatrix} F_{\mathbf{h}_1 - \mathbf{h}_1} & \cdots & F_{\mathbf{h}_1 - \mathbf{h}_n} \\ \cdots & \cdots & \cdots \\ \cdots & F_{\mathbf{h}_i - \mathbf{h}_j} & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ F_{\mathbf{h}_n - \mathbf{h}_1} & \cdots & F_{\mathbf{h}_n - \mathbf{h}_n} \end{vmatrix} \geq 0 \quad (n = 1, 2, 3, \ldots) . \tag{10}$$

This is the Karle & Hauptman inequalities in the three-dimensional case.

We shall consider the reduction of the matrix applicable to any space group:

$$\{ \boldsymbol{S}_0 \equiv \boldsymbol{E} = (\mathbf{1}|0), \ldots, \boldsymbol{S}_p = (\boldsymbol{R}_p | \mathbf{t}_p), \ldots \} \quad (p = 0, 1, 2, \ldots, s-1),$$

$\boldsymbol{R}_p$ being the rotational part and $\mathbf{t}_p$ the translational part of the pth symmetry operator $\boldsymbol{S}_p$. Let us rearrange the elements of matrix (5), and then set up the following one (11)

$$\boldsymbol{F}' = \begin{bmatrix} \boldsymbol{F}'_{00} & \cdots & \boldsymbol{F}'_{0, s-1} \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \boldsymbol{F}'_{pq} & \cdots \\ \cdots & \cdots & \cdots \\ \boldsymbol{F}'_{s-1, 0} & \cdots & \boldsymbol{F}'_{s-1, s-1} \end{bmatrix}, \tag{11}$$

where its pq element is

$$\boldsymbol{F}'_{pq} = \begin{bmatrix} F_{\boldsymbol{R}_p \mathbf{h}_1, \boldsymbol{R}_q \mathbf{h}_1} & \cdots & F_{\boldsymbol{R}_p \mathbf{h}_1, \boldsymbol{R}_q \mathbf{h}_m} \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & F_{\boldsymbol{R}_p \mathbf{h}_i, \boldsymbol{R}_q \mathbf{h}_j} & \cdots \\ \cdots & \cdots & \cdots \\ F_{\boldsymbol{R}_p \mathbf{h}_m, \boldsymbol{R}_q \mathbf{h}_1} & \cdots & F_{\boldsymbol{R}_p \mathbf{h}_m, \boldsymbol{R}_q \mathbf{h}_m} \end{bmatrix}, \tag{12}$$

$\mathbf{h}_1, \ldots, \mathbf{h}_m$ being a set of necessary asymmetric vectors in reciprocal lattice. Here it is to be added that the hermitian non-negative character still holds in (11). Furthermore, using the following unitary matrix:

$$\boldsymbol{T} = \begin{bmatrix} \boldsymbol{T}_0 & & & & 0 \\ & \ddots & & & \\ & & \boldsymbol{T}_p & & \\ & & & \boldsymbol{T}_q & \\ 0 & & & \ddots & \boldsymbol{T}_{s-1} \end{bmatrix},$$

$$\boldsymbol{T}_p = \begin{bmatrix} \exp[-2\pi i \mathbf{h}_1 \mathbf{t}_p] & & & 0 \\ & \ddots & & \\ & & \exp[-2\pi i \mathbf{h}_i \mathbf{t}_p] & \\ & & & \ddots \\ 0 & & & \exp[-2\pi i \mathbf{h}_m \mathbf{t}_p] \end{bmatrix}, \tag{13}$$

we shall transform equation (11) to (14)

$$\boldsymbol{T}\boldsymbol{F}'\boldsymbol{T}^{-1} = \begin{bmatrix} \boldsymbol{T}_0 \boldsymbol{F}'_{00} \boldsymbol{T}_0^{-1} & \cdots & \boldsymbol{T}_0 \boldsymbol{F}'_{0, s-1} \boldsymbol{T}_{s-1}^{-1} \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \boldsymbol{T}_p \boldsymbol{F}'_{pq} \boldsymbol{T}_q^{-1} & \cdots \\ \cdots & \cdots & \cdots \\ \boldsymbol{T}_{s-1} \boldsymbol{F}'_{s-1, 0} \boldsymbol{T}_0^{-1} & \cdots & \boldsymbol{T}_{s-1} \boldsymbol{F}'_{s-1, s-1} \boldsymbol{T}_{s-1}^{-1} \end{bmatrix}, \tag{14}$$

where $\boldsymbol{T}_p \boldsymbol{F}'_{pq} \boldsymbol{T}_q^{-1}$ of matrix (14) is

$$\begin{bmatrix} F_{\boldsymbol{R}_p \mathbf{h}_1, \boldsymbol{R}_q \mathbf{h}_1} \exp[-2\pi i (\mathbf{h}_1 \mathbf{t}_p - \mathbf{h}_1 \mathbf{t}_q)] & \cdots & F_{\boldsymbol{R}_p \mathbf{h}_1, \boldsymbol{R}_q \mathbf{h}_m} \exp[-2\pi i (\mathbf{h}_1 \mathbf{t}_p - \mathbf{h}_m \mathbf{t}_q)] \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & F_{\boldsymbol{R}_p \mathbf{h}_i, \boldsymbol{R}_q \mathbf{h}_j} \exp[-2\pi i (\mathbf{h}_i \mathbf{t}_p - \mathbf{h}_j \mathbf{t}_q)] & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ F_{\boldsymbol{R}_p \mathbf{h}_m, \boldsymbol{R}_q \mathbf{h}_1} \exp[-2\pi i (\mathbf{h}_m \mathbf{t}_p - \mathbf{h}_1 \mathbf{t}_q)] & \cdots & F_{\boldsymbol{R}_p \mathbf{h}_m, \boldsymbol{R}_q \mathbf{h}_m} \exp[-2\pi i (\mathbf{h}_m \mathbf{t}_p - \mathbf{h}_m \mathbf{t}_q)] \end{bmatrix}. \tag{15}$$

Now, making use of relation (9) and introducing $\boldsymbol{R}_r=\boldsymbol{R}_p^{-1}\boldsymbol{R}_q$, the elements in matrix (15) can be expressed

$$N^3 F_{\boldsymbol{R}_p\mathbf{h}_i,\,\boldsymbol{R}_q\mathbf{h}_j} \exp\left[-2\pi i(\mathbf{h}_i\mathbf{t}_p-\mathbf{h}_j\mathbf{t}_q)\right]$$
$$=F_{\boldsymbol{R}_p\mathbf{h}_i-\boldsymbol{R}_q\mathbf{h}_j} \exp\left[-2\pi i(\mathbf{h}_i\mathbf{t}_p-\mathbf{h}_j\mathbf{t}_q)\right]$$
$$=F_{\boldsymbol{R}_p(\mathbf{h}_i-\boldsymbol{R}_r\mathbf{h}_j)} \exp\left[-2\pi i(\mathbf{h}_i\mathbf{t}_p-\mathbf{h}_j\mathbf{t}_q)\right],$$

which, using the well known character $(\mathbf{t}_q=\mathbf{t}_p\boldsymbol{R}_r+\mathbf{t}_r)$ and the structure factor relation

$$(F_{\mathbf{h}}=F_{\boldsymbol{R}_p\mathbf{h}} \exp\left[-2\pi i\mathbf{h}\mathbf{t}_p\right]),$$

can be expressed in a simple form

$$F_{\boldsymbol{R}_p(\mathbf{h}_i-\boldsymbol{R}_r\mathbf{h}_j)} \exp\left[-2\pi i(\mathbf{h}_i-\boldsymbol{R}_r\mathbf{h}_j)\mathbf{t}_p\right].\exp\left[2\pi i\mathbf{h}_j\mathbf{t}_r\right]$$
$$=F_{\mathbf{h}_i-\boldsymbol{R}_r\mathbf{h}_j} \exp\left[2\pi i\mathbf{h}_j\mathbf{t}_r\right]. \qquad (16)$$

Hence, equation (15) becomes

$$\boldsymbol{T}_p\boldsymbol{F}'_{pq}\boldsymbol{T}_q^{-1}=\sum_{r=0}^{s-1}\delta_{\boldsymbol{R}_p\boldsymbol{R}_r\boldsymbol{R}_q^{-1},\,\mathbf{1}}.\boldsymbol{F}'_{\boldsymbol{R}_r}, \qquad (17)$$

where

$$\delta_{\boldsymbol{R}_p\boldsymbol{R}_r\boldsymbol{R}_q^{-1},\,\mathbf{1}}=1, \quad \text{if} \quad \boldsymbol{R}_p\boldsymbol{R}_r\boldsymbol{R}_q^{-1}=\mathbf{1},$$
$$\delta_{\boldsymbol{R}_p\boldsymbol{R}_r\boldsymbol{R}_q^{-1},\,\mathbf{1}}=0, \quad \text{if} \quad \boldsymbol{R}_p\boldsymbol{R}_r\boldsymbol{R}_q^{-1}\neq\mathbf{1},$$

and

$$\boldsymbol{F}'_{\boldsymbol{R}_r}\equiv\frac{1}{N^3}\times$$
$$\begin{bmatrix} F_{\mathbf{h}_1-\boldsymbol{R}_r\mathbf{h}_1}\exp\left[2\pi i\mathbf{h}_1\mathbf{t}_r\right] & \ldots & F_{\mathbf{h}_1-\boldsymbol{R}_r\mathbf{h}_m}\exp\left[2\pi i\mathbf{h}_m\mathbf{t}_r\right] \\ \cdots & \cdots & \cdots \\ \cdots & F_{\mathbf{h}_i-\boldsymbol{R}_r\mathbf{h}_j}\exp\left[2\pi i\mathbf{h}_j\mathbf{t}_r\right] & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ F_{\mathbf{h}_m-\boldsymbol{R}_r\mathbf{h}_1}\exp\left[2\pi i\mathbf{h}_1\mathbf{t}_r\right] & \ldots & F_{\mathbf{h}_m-\boldsymbol{R}_r\mathbf{h}_m}\exp\left[2\pi i\mathbf{h}_m\mathbf{t}_r\right] \end{bmatrix}. \qquad (18)$$

As is well known, the elements of the regular representation-matrix $\boldsymbol{P}(\boldsymbol{R}_r)$ for an element $\boldsymbol{R}_r$ in a given point group

$$(\boldsymbol{R}_0\equiv\mathbf{1},\ldots,\boldsymbol{R}_p,\ldots,\boldsymbol{R}_q,\ldots,\boldsymbol{R}_r,\ldots,\boldsymbol{R}_{s-1})$$

can be given by

$$\boldsymbol{P}_{\boldsymbol{R}_p,\boldsymbol{R}_q}(\boldsymbol{R}_r)=\delta_{\boldsymbol{R}_p\boldsymbol{R}_r\boldsymbol{R}_q^{-1},\,\mathbf{1}}. \qquad (19)$$

Combining (14), (17), (18) and (19), we obtain

$$\boldsymbol{T}\boldsymbol{F}'\boldsymbol{T}^{-1}=\sum_{r=0}^{s-1}\boldsymbol{P}(\boldsymbol{R}_r)\times\boldsymbol{F}'_{\boldsymbol{R}_r}. \qquad (20)$$

The reduction of the matrix (20) can be made using the irreducible representation for $\boldsymbol{P}(\boldsymbol{R}_r)$. Therefore, the reduced form of (20) is represented by a form of a direct sum as

$$(\boldsymbol{T}\boldsymbol{F}'\boldsymbol{T}^{-1})_{\text{irred.}}=\sum_{k=1}^{l}{}^{+}\, n_k\boldsymbol{F}^k, \qquad (21)$$

$$\boldsymbol{F}^k=\sum_{r=0}^{s-1}\boldsymbol{P}_k(\boldsymbol{R}_r)\times\boldsymbol{F}'_{\boldsymbol{R}_r}, \qquad (22)$$

where Σ^+ means direct sum of matrices and $\boldsymbol{P}_k(\boldsymbol{R}_r)$ is the kth irreducible representation-matrix obtained from the reduction of $\boldsymbol{P}(\boldsymbol{R}_r)$, n_k its dimension and l the number of classes.

The results (21) and (22) show that the general form for the inequalities is given by the principal subdeterminants of each $\boldsymbol{F}^k$ which can be taken as non-negative; namely, using (18) and (22),

$$\text{princ. subdet.}\begin{bmatrix} \boldsymbol{F}_{11}^k & \cdots & \boldsymbol{F}_{1m}^k \\ \cdots & \cdots & \cdots \\ \cdots & \boldsymbol{F}_{ij}^k & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \boldsymbol{F}_{m1}^k & \cdots & \boldsymbol{F}_{mm}^k \end{bmatrix}\geq 0 \qquad (23)$$

with

$$\boldsymbol{F}_{ij}^k\equiv\sum_{r=0}^{s-1}\boldsymbol{P}_k(\boldsymbol{R}_r)F_{\mathbf{h}_i-\boldsymbol{R}_r\mathbf{h}_j}\exp\left[2\pi i\mathbf{h}_j\mathbf{t}_r\right]. \qquad (24)$$

The result obtained is in harmony with that of Goedkoop (1952*a*, p. 89).

The use of this theory for discussion about other types of inequalities derived by other authors (Harker & Kasper, 1948; Gillis, 1948; MacGillavry, 1950; Okaya & Nitta, 1952; von Eller, 1955; Bouman, 1956) will be given elsewhere, together with the practical applications of the result (23) to some space groups.

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