Acta Cryst. (1989). A45, 144

An alternative approach to the Hauptman–Karle determinantal inequalities. By E. PRINCE, *Institute for Materials Science and Engineering, National Bureau of Standards, Gaithersburg, MD* 20899, *USA*

(*Received* 4 *May* 1988; *accepted* 9 *June* 1988)

Abstract

A procedure is described for constructing a series of progressively stronger restrictions on the magnitudes and phases of individual structure factors in terms of sets of other structure factors. The existence of the Cholesky factors of Hauptman–Karle matrices is used to ensure that the electron density is everywhere positive.

Karle & Hauptman (1950; also Karle, 1985; Woolfson, 1987) showed that the condition that electron densities may never be negative implies that the determinants of all possible matrices of the form

$$\mathbf{A}=\begin{pmatrix} 1 & U(\mathbf{h}_1) & U(\mathbf{h}_2) & \cdots & U(\mathbf{h}_n) \\ U(-\mathbf{h}_1) & 1 & U(\mathbf{h}_2-\mathbf{h}_1) & \cdots & U(\mathbf{h}_n-\mathbf{h}_1) \\ U(-\mathbf{h}_2) & U(\mathbf{h}_1-\mathbf{h}_2) & 1 & \cdots & U(\mathbf{h}_n-\mathbf{h}_2) \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ U(-\mathbf{h}_n) & U(\mathbf{h}_1-\mathbf{h}_n) & U(\mathbf{h}_2-\mathbf{h}_n) & \cdots & 1 \end{pmatrix}, \quad (1)$$

where $U(\mathbf{h})$ is the unitary structure factor of reflection $\mathbf{h}$, must be greater than or equal to zero. This statement is equivalent to the statement that all such matrices are positive semidefinite, and it is an immaterial further restriction to require actual positive definiteness. For convenience we shall restrict this discussion to real structure factors, so that $U(-\mathbf{h})=U(\mathbf{h})$. The extension to complex structure factors, for which $\mathbf{A}$ is Hermitian, is straightforward. If and only if $\mathbf{A}$ is positive definite, there exists (Stewart, 1973) an upper triangular matrix, $\mathbf{R}$, with positive diagonal elements (a so-called *Cholesky factor*) such that $\mathbf{R}^T\mathbf{R}=\mathbf{A}$, so that to show that $\mathbf{A}$ is positive definite it is sufficient to show that $\mathbf{R}$ exists. (Note that the determinant of $\mathbf{A}$ is the square of the product of the diagonal elements of $\mathbf{R}$.) For the 3×3 submatrix in the upper left corner of $\mathbf{A}$,

$$\mathbf{R}=\begin{pmatrix} 1 & U(\mathbf{h}_1) & U(\mathbf{h}_2) \\ 0 & [1-U(\mathbf{h}_1)^2]^{1/2} & [U(\mathbf{h}_1-\mathbf{h}_2)-U(\mathbf{h}_1)U(\mathbf{h}_2)] \times[1-U(\mathbf{h}_1)^2]^{-1/2} \\ 0 & 0 & \{1-U(\mathbf{h}_2)^2-[U(\mathbf{h}_1-\mathbf{h}_2)-U(\mathbf{h}_1)U(\mathbf{h}_2)]^2 \times[1-U(\mathbf{h}_1)^2]^{-1}\}^{1/2} \end{pmatrix}. \quad (2)$$

For $\mathbf{R}$ to exist all quantities whose square roots must be extracted to form its diagonal elements must be positive. Thus, for R_{33}

$$[U(\mathbf{h}_1-\mathbf{h}_2)-U(\mathbf{h}_1)U(\mathbf{h}_2)]^2<[1-U(\mathbf{h}_1)^2][1-U(\mathbf{h}_2)^2], \quad (3)$$

which is equivalent to a fundamental inequality given by Karle & Hauptman (1950) that puts restrictions on the phase of $U(\mathbf{h}_1-\mathbf{h}_2)$ if those of $U(\mathbf{h}_1)$ and $U(\mathbf{h}_2)$ are known.

Inequality (3) has the form $|U(\mathbf{h})-\delta|^2<r^2$; where r is the geometric mean of two positive real numbers, both of which are less than one. If $U(\mathbf{h}_1)$ and $U(\mathbf{h}_2)$ are both close to one, r can be small, and the phase of $U(\mathbf{h}_1-\mathbf{h}_2)$ correspondingly well defined. As the number of atoms in the asymmetric unit increases, however, the probability of finding large values of $|U(\mathbf{h})|$ decreases, and the phase-determining power of inequality (3) also decreases. Karle & Hauptman (1950) give expressions like inequality (3) involving matrices larger than 3×3 that have greater power. These expressions, however, are given in terms of minors of determinants, and, because of the rapid increase in the complexity of expansions of determinants as their size gets larger, have rarely if ever been put to practical use. Cholesky factorization, by contrast, is a sequential process (Prince, 1982) in which each element of the jth row of $\mathbf{R}$ is formed by subtracting from the corresponding element of $\mathbf{A}$ $j-1$ terms, each of which is a product of two elements of a higher row of $\mathbf{R}$, and then dividing by R_{jj}. Higher-power expressions like inequality (3) can therefore be derived explicitly fairly easily. For example, the corresponding expression from a 4×4 matrix, which relates one unitary structure factor to five others, is

$$\begin{aligned}&\{U(\mathbf{h}_2-\mathbf{h}_3)-U(\mathbf{h}_2)U(\mathbf{h}_3)\\&\quad-[U(\mathbf{h}_1-\mathbf{h}_2)-U(\mathbf{h}_1)U(\mathbf{h}_2)]\\&\quad\times[U(\mathbf{h}_1-\mathbf{h}_3)-U(\mathbf{h}_1)U(\mathbf{h}_3)]\}^2\\&\qquad<\{1-U(\mathbf{h}_2)^2-[U(\mathbf{h}_1-\mathbf{h}_2)\\&\qquad\quad-U(\mathbf{h}_1)U(\mathbf{h}_2)]^2/[1-U(\mathbf{h}_1)^2]\}\\&\qquad\quad\times\{1-U(\mathbf{h}_3)^2-[U(\mathbf{h}_1-\mathbf{h}_3)\\&\qquad\quad-U(\mathbf{h}_1)U(\mathbf{h}_3)]^2/[1-U(\mathbf{h}_1)^2]\}. \end{aligned}\quad (4)$$

The two factors on the right-hand side of inequality (4) are both less than or equal to the corresponding factors in inequality (3), and the positive definiteness of $\mathbf{A}$ ensures that they are positive.

Because of the inherent symmetry of the Cholesky factorization procedure, the only elements of $\mathbf{R}$ that have a new form in a still bigger matrix are the last two in the final column. Thus, the explicit derivation of still more powerful relations, while tedious, is straightforward and does not increase rapidly in complexity.

References

KARLE, J. (1985). *Structure and Statistics in Crystallography*, edited by A. J. C. WILSON, pp. 1-22. Guilderland, NY: Adenine Press.

KARLE, J. & HAUPTMAN, H. (1950). *Acta Cryst.* 3, 181-187.

PRINCE, E. (1982). *Mathematical Techniques in Crystallography and Materials Science.* New York: Springer.

STEWART, G. W. (1973). *Introduction to Matrix Computations.* New York: Academic Press.

WOOLFSON, M. M. (1987). *Acta Cryst.* A43, 593-612.

0108-7673/89/010144-01$03.00