through electron microscopy, which shows that the antiphase domains are rounded to blocky in shape, about 50–100 Å in diameter (see Fig. 6 in Ghose, Ng & Walter, 1972).

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References

BAILEY, J. C., CHAMPNESS, P. E., DUNHAM, A. C., ESSON, J., FYFE, W. S., MACKENZIE, W. S., STUMPFL, E. F. & ZUSSMAN, J. (1970). *Proc. Apollo* 11 *Lunar Sci. Conf., Geochim. Cosmochim. Acta, Suppl.* 1, Vol. 1, pp. 169–194.
BOWN, M. G. & GAY, P. (1957). *Acta Cryst.* 10, 440–441.
BROWN, G. E., PREWITT, C. T., PAPIKE, J. J. & SUENO, S. (1972). *J. Geophys. Res.* 77, 5778–5789.
CHAMPNESS, P. E. & LORIMER, G. W. (1971). *Contrib. Miner. Petrol.* 33, 171–183.
CHRISTIE, J. M., LALLY, J. S., HEUER, A. H., FISHER, R. M., GRIGGS, D. T. & RADCLIFFE, S. V. (1971). *Proc. 2nd Lunar Sci. Conf.,* Vol. 1, pp. 68–89. Boston: M.I.T. Press.
CLARK, J., ROSS, M.& APPLEMAN, D. (1971). *Amer. Min.* 56, 888–908.
GHOSE, S., NG, G. & WALTER, L. S. (1972). *Proc. 3rd Lunar Sci. Conf. Geochim. Cosmochim. Acta, Suppl.* 3, Vol. 1, pp. 507–531.
GUINIER, A. (1963). *X-Ray Diffraction.* San Francisco: W. H. Freeman.
HOHN, F. E. (1964). *Elementary Matrix Algebra,* 2nd ed. New York: Macmillan.
MORIMOTO, N. (1956). *Proc. Japan Acad.* 32, 750–752.
MORIMOTO, N. & GÜVEN, N. (1970). *Amer. Min.,* 55, 1195–1209.
MORIMOTO, N. & TOKONAMI, M. (1969). *Amer. Min.* 54, 725–740.
NYE, J. F. (1957). *Physical Properties of Crystals.* Oxford Univ. Press.
PREWITT, C. T., PAPIKE, J. J. & ROSS, M. (1970). *Earth Planet. Sci. Lett.* 8, 448.
TAKEDA, H. (1971). *Earth Planet. Sci. Lett.* 15, 65–71.

Direct Structure Determination of Asymmetric Membrane Systems from X-ray Diffraction*

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A theoretical analysis of X-ray diffraction from asymmetric planar systems is given. Phase information is obtained from the continuous intensity function from such a system. Although a unique phase function cannot be determined, it is possible to derive the relatively small number of phase solutions which are consistent with the observed diffraction.

The structure of biological membranes is a subject of considerable current interest. X-ray diffraction studies have had a dominant role and have yielded much valuable information on this important topic. One fact about membrane ultrastructure which is slowly becoming evident is that most natural biological membranes (in contrast to artificial model lipid bilayer systems) are asymmetric. This asymmetry appears to be predominantly due to the protein component of the membrane (which is not to rule out the possibility of an asymmetric distribution of lipids in the membrane). Hence, the functional properties of the membrane are determined, in large part, from this property. Fortunately, many of the naturally occurring asymmetric membrane systems consist of repeating units of membrane pairs which have a center of symmetry. This permits the rather well developed theory of diffraction by centrosymmetric structures to be utilized in structure determination. However, there are many other membrane systems of interest where the former theory is not applicable (*e.g.*, dispersions of membrane vesicles and sheets). The present paper is concerned with the analysis of these asymmetric systems.

The difficulty in analyzing diffraction data from asymmetric structures (as well as symmetric ones) lies in the well-known phase problem of X-ray diffraction theory. There are indirect methods of obtaining this phase information (*e.g.* isomorphous replacement), but they have not been very useful for membranes. There has been much work in the past few years on direct methods of phase determination. These studies have shown that the X-ray diffraction intensity data contain some of the phase information necessary for a structural determination. Crucial to these studies has been the realization that the rather simple mathematical properties of positivity and boundedness of the electron density distribution (e.d.d.) place a severe restriction on its Fourier transform. In particular, the property

of boundedness is of utmost importance since it means that the Fourier transform must be an analytic function. Hence, the full power of analytic-function theory can be utilized to attack the problem.

Consider a planar multilayer system of a finite number, n, of membrane sheets of infinite radius. One is interested in determining the projection $t_n(x)$ of the electron density of the multilayer onto the normal to the sheets. The observed diffracted intensity from such a system will be denoted

$$I_n(X) \propto K(X) \cdot |T_n(X)|^2 \tag{1}$$

where $T_n(X)$ is the Fourier transform of $t_n(x)$ and $K(X)$ is a correction factor appropriate for the particular experimental arrangement being used. Of great importance is the autocorrelation function of $t_n(x)$, denoted by Hosemann & Bagchi (1962) as

$$Q(x) = t_n(x) * t_n(-x) = \text{F.T.}|T_n(X)|^2, \tag{2}$$

where the $*$ denotes the convolution product,

$$t_n(x) * t_n(-x) = \int t_n(x')t_n(x'+x)dx' . \tag{3}$$

The above authors showed that if n is not too large, the function

$$Q_0(x) = t(x) * t(-x) \tag{4}$$

can be obtained from $Q(x)$, where $t(x)$ is the desired e.d.d. of one membrane sheet. When $t(x)$ contains a center of symmetry, it can be determined by a deconvolution of equation (4). Analyses of this type have been carried out for artificial lipid bilayer systems by Lesslauer & Blasie (1972) and for naturally occurring membrane pair systems by Worthington, King & McIntosh (1973). In the case of membrane dispersions, one observes the function $|T(X)|^2$ from which $Q_0(x)$ can be obtained directly by Fourier transformation. Hence, the structure analysis of asymmetric systems to be discussed in this paper will assume that one has obtained the $Q_0(x)$ function in one of these two ways. Thus, the problem is to obtain $t(x)$ from $Q_0(x)$ by some sort of deconvolution procedure, analogous to the centrosymmetric case, or equivalently, to determine the phase function directly from the $|T(X)|^2$ function.

From the convolution theorem of Fourier transform theory it is known that the autocorrelation function is

$$Q_0(x) = 2\int_0^\infty |T(X)|^2 \cos 2\pi Xx dX . \tag{5}$$

Therefore, if the continuous intensity function $|T(X)|^2$ can be obtained, it is a simple matter to obtain $Q_0(x)$ by the above transformation (it is assumed that one has calculated the necessary factors to obtain $|T(X)|^2$ from the observed intensity function). One potential problem that becomes apparent is that $|T(X)|^2$ is only known from some lower limit $X_0 > 0$ to an upper limit X_1. In principle one can extrapolate the $|T(X)|^2$ function both to zero and infinity (King & Worthington, 1971), apparently circumventing this problem. How-

ever, in practice the extrapolation to infinity is frustrated by the presence of experimental errors in the data.

Furthermore, in this procedure one must know the width of the 'structure' under consideration in order to satisfactorily accomplish the extrapolations. Various methods of extrapolating $|T(X)|^2$ to $X=0$ have been used. For the purpose of this discussion, I shall assume a method has been found that leads to correct values of $Q_0(x)$.

Deconvolution of $Q_0(x)$

Before progressing to the asymmetric deconvolution, it might prove useful to summarize briefly the procedure for symmetric structures. After Hosemann & Bagchi, the structure of width d is divided into $2(m+1)$ equal strips of width δ. The width δ is made small enough so that $Q_0(x)$ and $t(x)$ are approximately constant in δ. The electron densities of the strips are denoted $[t_0, t_1, \ldots t_{m-1}, t_m, t_m, t_{m-1}, \ldots t_i \ldots t_0]$. The e.d.d. can be obtained uniquely from $Q_0(x)$ through the following recursion relation

$$Q_0(d-n\delta) = \delta^2 \sum_{i=0}^n t_i t_{n-i} \quad n=0,1,\ldots 2(m+1) . \tag{7}$$

The only ambiguity in the recursion relation is that the first value t_0 is obtained by taking the square root of $Q_0(d)$ and either the $+$ or $-$ root can be taken. The two structures so obtained are the negatives of each other. If the property of positivity of the electron density is invoked, the negative structure is ruled out and uniqueness is obtained.

In the case of biological membranes, one is usually interested in the e.d.d. of the membrane relative to the density of the surrounding fluid layer. Thus the negative structure cannot be immediately ruled out and must be considered. However, in practice it can usually be eliminated on general physical and chemical grounds.

In the case of an asymmetric structure, the same sort of latticing procedure is applied, i.e. the structure of width d is divided into $(m+1)$ strips of equal width δ. The electron densities of the strips are denoted $[t_0, t_1, t_2, \ldots t_i \ldots t_{m-1}, t_m]$. The electron-density distribution can be obtained from the following set of nonlinear algebraic equations

$$Q_0(n\delta) = \delta^2 \sum_{i=0}^{m-n} t_i t_{i+n} \quad n=0,1,\ldots,m . \tag{8}$$

The equations are no longer in the form of a recursion relation. They no longer possess a unique solution and, moreover, may contain an immense manifold of solutions.

The nature of this ambiguity in the solution of the autocorrelation function has been discussed by Calderon & Pepinsky (1952). Using the notation here, they showed that if $T(X)$ can be factored as

$$T(X) = T_1(X) \cdot T_2(X) \cdot T_3(X) \ldots T_i(X) \ldots , \tag{9}$$

that there is an indeterminacy in $t(x)$ given by

$$t(x) = t_1(\pm x) * t_2(\pm x) * t_3(\pm x) * \ldots t_i(\pm x) * \ldots (10)$$

In the situation where $t(x)$ possesses a center of symmetry, there is no ambiguity in equation (10) and a unique solution is obtained. In the asymmetric case each sign combination of equation (10) generally gives a different solution to the autocorrelation function $Q_0(x)$. However, this result does not answer the key question of how $T(X)$ can be factored or even whether it is always possible to factor it at all. It will be seen that a much more meaningful investigation can be made by looking at the mathematical properties of $T(X)$ directly.

Solution of the phase problem

Given the generally complex function $T(X)$ which is the Fourier transform of a bounded real function $t(x)$, the latter defined for a finite interval $(0, d)$ on the real axis and zero elsewhere, the possibility of reconstructing the function $T(X)$ when only its amplitude $|T(X)|$ is known should first be considered. The quantities are related by the expression

$$T(X) = |T(X)| \exp [i\alpha(X)] . \tag{11}$$

One can analytically continue this function from the real axis to the entire complex plane by defining

$$T(Z) = \int_0^d t(x) \exp (i2\pi Zx)dx , \tag{12}$$

where $Z = X + iY$. It is apparent from equation (12) that $T(Z)$ is analytic in the entire complex plane except for $Y \to -\infty$. In fact, from Titchmarsh (1939) it is seen that $T(Z)$ is an integral function of order one. Furthermore, from the asymptotic behavior of Fourier transforms (Dettman, 1965), one sees that

$$T(Z) \sim i/Z \tag{13}$$

for $|Z| \to \infty$ in the lower half plane (denoted by D_-).

By considering Cauchy's integral around a contour, L, consisting of the real axis and an infinite semicircle in the upper half plane (denoted by D_+), one can write

$$T(Z) = \frac{1}{2\pi i} \int_L \frac{T(Z')dZ'}{Z' - Z} , \quad Z \text{ in } D_+ . \tag{14}$$

From this integral it follows that

$$T(X) = \frac{1}{\pi i} \int_{-\infty}^{\infty} \frac{i \exp [-i\alpha(X')] \sin \alpha(X')T(X')dX'}{X' - X - i\varepsilon} \tag{15}$$

in the limit where $(\varepsilon > 0) \to 0$. It will now be useful to consider the following analytic function

$$\Phi(Z) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{B(X')T(X')dX'}{X' - Z} . \tag{16}$$

If we denote its limiting value in D_+ as $\Phi^+(X)$ and in D_- as $\Phi^-(X)$, Gakhov (1966) has shown that

$$\Phi^+(X) - \Phi^-(X) = B(X)T(X) . \tag{17}$$

Furthermore, it is apparent that

$$2\Phi^+(X) = \frac{1}{\pi i} \int_{\infty-}^{\infty} \frac{B(X')T(X')dX'}{X' - X - i\varepsilon} . \tag{18}$$

Hence, if we identify $B(X)$ as the factor in front of $T(X')$ in the integral of equation (15), it follows that

$$\Phi^+(X) = \exp [2i\alpha(X)]\Phi^-(X) . \tag{19}$$

The solution to the boundary value problem defined by equation (19) will be necessary in order to find an expression for the phase function. That is, it is required to find two functions $\Phi^+(Z)$ and $\Phi^-(Z)$ which are analytic in D_+ and D_-, respectively, and which also satisfy equation (19). The boundary-value problem thus stated is an established one in complex function theory, and is known as the homogeneous Riemann problem for the semi-plane. The solution to the problem is discussed by Gakhov (1966), and since it is not a familiar problem to many readers a brief summary of it is given in the Appendix.

With the solution to the homogeneous Riemann problem from the Appendix, given by equation (A-7), it is possible to solve for $|T(X)|$ in terms of the phase function. The solution is accomplished by utilizing the Plemelj formulae, given by equation (17). Hence,

$$|T(X)| = \frac{2P_{m-1}(X)}{(X^2+1)^{m/2}} \exp \left[\frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\ln G_0(X')dX'}{X' - X} \right] . \tag{20}$$

If we take the natural logarithm of both sides of equation (20), it follows that

$$\ln \left\{ \frac{(X^2+1)^{m/2}|T(X)|}{2P_{m-1}(X)} \right\} = \frac{1}{\pi i} \int_{-\infty}^{\infty} \frac{\frac{1}{2} \ln G_0(X')dX'}{X' - X} . \tag{21}$$

If the following analytic function is considered:

$$\Phi(Z) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\frac{1}{2} \ln G_0(X')dX'}{X' - Z} , \tag{22}$$

the Plemelj formulae indicate that

$$\Phi^+(X) - \Phi^-(X) = \frac{1}{2} \ln G_0(X) , \tag{23}$$

$$\Phi^+(X) + \Phi^-(X) = \ln \left\{ \frac{(X^2+1)^{m/2}|T(X)|}{2P_{m-1}(X)} \right\} . \tag{24}$$

It is now necessary to investigate the boundary-value problem designated as the non-homogeneous Riemann problem,

$$\Phi^+(X) = (-1) \cdot \Phi^-(X) + g(X) \tag{25}$$

where the right-hand side of equation (24) is $g(X)$. The solution to this problem is given in the Appendix for the case where $T(X)$ has no zeros and also for the situation when $T(X)$ does have zeros.

The simplest case, and the one which will be discussed first, is when $T(Z)$ has no zeros on the finite real axis or in the upper half plane. The solution can be obtained simply from the solution of the non-homogeneous problem in the semi-plane given by equation (A-12) in the Appendix, for $m=0$. Again, utilizing the Plemelj formulae, one can calculate the phase function to be

$$a_1(X) = -\frac{2X}{\pi} \int_0^\infty \frac{\ln|T(X')| dX'}{X'^2 - X^2}. \quad (26)$$

Since this solution will be seen to be of great importance, it will be designated as the fundamental solution, type I, of the phase problem. It is apparent that, given the magnitude of the transform along the real axis, the fundamental phase solution can be directly calculated. Unfortunately, the fundamental solution is not the only solution to the phase problem. Since it was assumed that all of the zeros of $T(Z)$ were in the lower half plane to obtain equation (26), it should be apparent that the non-uniqueness of the phase problem is somehow intimately linked to the distribution of zeros in the complex plane.

Another important case, designated as the fundamental solution, type II, is when there are zeros on the real axis but none in D_+. The solution to the phase problem can be obtained in this case from the solution of the non-homogeneous problem on the segmented real axis, given by equation (A-15) in the Appendix. Once again, the Plemlj formulae are utilized to obtain the phase function

$$\alpha_{II}(X) = -\frac{2XR(X)}{\pi} \int_0^\infty \frac{\ln\left\{\dfrac{(X'^2+1)^{m/2}|T(X')|}{R(X')}\right\} dX'}{R(X')(X'^2 - X^2)}$$
$$+ m[\pi/2 - \tan^{-1}(1/X)] \quad (27)$$

where $R(X)$ is the polynomial coinciding with the zeros of $T(X)$ and the index m is also determined by the real zeros. Hence, as long as there are no zeros in D_+ a unique solution to the phase problem exists, given by either equation (26) or equation (27), depending on whether $T(X)$ has zeros or not.

In the general situation there may be zeros anywhere in the whole complex plane, including D_+. Titchmarsh (1926) has shown that a function such as $T(Z)$ defined by equation (12) can be written

$$T(Z) = T(0) \exp[i\pi dZ] \prod_{n=1}^\infty (1 - Z/Z_n), \quad (28)$$

where the product is extended over all of the zeros Z_n. Hence, $T(Z)$ is completely determined by a knowledge

of all of its zeros. Since only the positions of the real zeros of $T(X)$ are known experimentally, the non-uniqueness of the solution to the phase problem is due to ignorance of the positions of the non-real zeros. Although equation (28) is completely general it will prove useful to have another expression for $T(Z)$. In the special case where there are no zeros on the real axis, but there are some in D_+, Toll (1956) has shown that $T(X)$ can be written as

$$T(X) = |T(X)| \exp[i\alpha_1(X)] \prod_n \left(\frac{X-Z_n}{X-Z_n^*}\right), \quad (29)$$

where Z_h^* is the conjugate zero to Z_n and the product is over all zeros in D_+. In the most general case, with zeros on the real axis, $\alpha_1(X)$ should be replaced by $\alpha_{II}(X)$ in equation (29).

The rather complex mathematical argument given in the preceding discussion is necessary for the rigorous development of the concepts presented. However, a rather intuitive argument based on elementary complex-function theory might be appropriate at this point. It is well known that the real and imaginary parts of an analytic function are uniquely determinable from each other. If one takes the natural logarithm of $T(Z)$ and assumes that it is also analytic, then from the preceding argument it follows that $\ln|T|$ and the phase function α are also uniquely determinable from each other. Equations (20) and (26) illustrate such a relationship between these two functions. Since the logarithm of zero is not defined, it is not surprising that the zeros of $T(Z)$ pose special problems for the analysis. When these singular points are properly taken into account, the results of the preceding discussion follow.

Discussion

What has been obtained is the fact that the zeros of the analytic continuation of $T(X)$ to the complex plane are of supreme importance in the solution of the phase problem. If the positions of all of these zeros were known, the phase could be uniquely determined from the experimentally obtainable intensity data. However, since only the real zeros are amenable to experimental location, it is only possible to calculate one of the two special cases considered in the preceding discussion, i.e. the fundamental solutions of type I or type II. In general, there is no guarantee that either of these solutions is the correct one. Hence, there is a fundamental indeterminancy in the phase problem.

In order to gain further insight into the nature of the phase problem, it would be informative to know how many zeros $T(Z)$ has. Titchmarsh (1926) has shown that functions of this type possess an infinite number of zeros. Thus one is faced with the fact that there is an infinite manifold of phase functions which are consistent with the observed intensity! Fortunately, the situation is not quite as discouraging as this result would indicate, for all but a finite number of these

zeros have essentially no effect on the calculated e.d.d. The rationale for this conclusion was discussed by Nussenzveig (1967), who showed that the asymptotic distribution of zeros is determined by the properties of the cutoff of $t(x)$ and contains no information about the shape of $t(x)$ within its interval of definition. This information is contained in the zeros located relatively closer to the origin. Since the number of zeros of $T(Z)$ within a radius $|Z| \leq R$ is

$$n(R) \sim \frac{d}{\pi} R , \qquad (30)$$

as was shown by Titchmarsh (1926), it is apparent that there are a relatively small number of zeros in the region of experimental interest. Thus, in a practical sense, there are only a small finite number of phase solutions consistent with the data.

There are other properties of $t(x)$ which allow one to calculate all of the various phase possibilities inherent in the preceding discussion, but do not allow one to specify which of them is the correct one. Up to this point, the reality condition has not been explicitly imposed on $t(x)$. When it is specified that $t(x)$ must be real, it is apparent that

$$T^*(Z^*) = T(-Z) . \qquad (31)$$

Hence, we can conclude that the zeros of $T(Z)$ are either purely imaginary or occur in pairs symmetrically located about the imaginary axis; i.e., if Z is a zero, then so is $-Z^*$. Furthermore, if it is specified that $t(x)$ is positive, there can be no purely imaginary zeros. Although the positions of the zeros of $T(Z)$ are severely restricted by the reality and positivity conditions on $t(x)$, they are apparently not determinable from the magnitude of $T(X)$. This appears to be an inherent obstacle in the phase problem.

Despite the ambiguity of the correct phase solution from the intensity data, one can nevertheless calculate all of the possibilities. Hence, a knowledge of the chemical composition and other structural features may allow one to choose the correct solution from the given possibilities. Walther (1963) has shown that all possible solutions to the phase problem differ only by 'zero flips' about the real axis where Z_n and $-Z_n^*$ are replaced by Z_n^* and $-Z_n$, respectively. Thus, the fundamental solutions I or II can be calculated directly from equation (12) and the resulting distribution of zeros in the lower half plane determined by inspection. Each of the possible $T(X)$ functions can then be calculated from equation (29) by systematically transposing all combinations of the equivalent pairs of zeros across the real axis. As a final point, it should be noted that when the structure $t(x)$ has a center of symmetry, it follows that if Z_n and $-Z_n^*$ are zeros, then Z_n^* and $-Z_n$ must also be zeros. Thus, there is no ambiguity in this case and the phase problem has a unique solution.

The usefulness of these theoretical developments in solving asymmetric one-dimensional structures has been verified for a biological membrane of considerable interest (King, Blaurock & Stoeckenius, in preparation). Purple membrane, a cell membrane fragment isolated from *Halobacterium halobium*, apparently acts as a light-driven proton pump which may function as an alternative energy-conversion mechanism to the normal metabolic processes. These functional considerations, combined with electron microscopy (Blaurock & Stoeckenius, 1971), indicate that the membrane must have an asymmetric distribution in a direction perpendicular to its surface. The continuous X-ray intensity from the profile can be obtained and hence this system is an ideal subject for interpretation by the theory described in this paper. Although a unique structure could not be calculated from the theory, it does yield a possible profile which is consistent with the known chemical and physical properties of the membrane.

APPENDIX

Riemann boundary-value problem

For a more detailed discussion of the Riemann boundary-value problem see Gakhov (1966) or Muskhelishvili (1953). In this Appendix the treatment of the problem by Gakhov will be discussed and its essential features outlined. Although the problem can be defined and solved for a complicated system of intersecting arcs and contours in the complex plane, only the special cases of the problem for the real axis and the segmented real axis will be covered here.

The Riemann problem consists of finding two functions $\Phi^+(Z)$ and $\Phi^-(Z)$, analytic in the upper and lower half planes respectively, whose limiting values satisfy the equation

$$\Phi^+(X) = G(X)\Phi^-(X) + g(X) \quad (-\infty < X < \infty) . \quad \text{(A-1)}$$

It is assumed that $G(X)$ and $g(X)$ satisfy a Lipschitz condition of order λ and also that $G(X) \neq 0$. Of fundamental importance is the index m of the function $G(X)$, which is defined by

$$m = \frac{1}{2\pi i} \int d \ln G(X) . \qquad \text{(A-2)}$$

It follows from the properties of the logarithmic residue of the function $G(X)$ that m is determined by the number of zeros of $\Phi^+(Z)$ and $\Phi^-(Z)$.

If one first considers the homogeneous Riemann problem, where $g(X) = 0$ in equation (A-1), one can formally obtain the solution by taking logarithms of both sides

$$\ln \Phi^+(X) - \ln \Phi^-(X) = \ln G(X) . \qquad \text{(A-3)}$$

From this so-called Plemelj formula, it follows that the solution to the problem is given by

$$\Phi^+(Z) = A \exp [\Gamma^+(Z)] , \quad \Phi^-(Z) = A \exp [\Gamma^-(Z)] ,$$
$$\text{(A-4)}$$

where

$$\Gamma(Z) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\ln G(X')\mathrm{d}X'}{X'-Z} . \qquad \text{(A-5)}$$

For this formal result to be correct, $\ln G(X)$ must be a single-valued function, that is, m must be zero. In the more general case, with non-zero m, the solution can be obtained by considering the function

$$G_0(X) = G(X) \left(\frac{X-i}{X+i}\right)^{-m} \qquad \text{(A-6)}$$

which has zero index and whose logarithm is a one-valued function. Thus, the solution to the homogeneous problem with positive index m can be written

$$\Phi^+(Z) = P_m(Z)(Z+i)^{-m} \exp \Gamma^+(Z) ,$$
$$\Phi^-(Z) = P_m(Z)(Z-i)^{-m} \exp \Gamma^-(Z), \qquad \text{(A-7)}$$
where

$$\Gamma(Z) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\ln G_0(X')\mathrm{d}X'}{X'-Z} , \qquad \text{(A-8)}$$

and where $P_m(Z)$ is a polynomial of degree not higher than m.

The solution to the non-homogeneous problem, with $g(X) \neq 0$, is easily obtained from the preceding. We introduce the analytic function

$$\Psi(Z) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{g(X')\mathrm{d}X'}{\theta^+(X')(X'-Z)} , \qquad \text{(A-9)}$$

where $\theta(Z)$, the so-called canonical function, is defined

$$\theta^+(Z) = \exp \Gamma^+(Z) ,$$
$$\theta^-(Z) = \left(\frac{Z-i}{Z+i}\right)^{-m} \exp \Gamma^-(Z) . \qquad \text{(A-10)}$$

This enables us to write the analytic continuation of the boundary condition (A-1) as

$$\frac{\Phi^+(Z)}{\theta^+(Z)} - \Psi^+(Z) = \frac{\Phi^-(Z)}{\theta^-(Z)} - \Psi^-(Z)$$
$$= \frac{P_m(Z)}{(Z+i)^m} \ (m \geq 0) . \qquad \text{(A-11)}$$

From this follows the general solution to the problem

$$\Phi(Z) = \theta(Z) \left[\Psi(Z) + \frac{P_m(Z)}{(Z+i)^m}\right] \ m \geq 0 \quad \text{(A-12)}$$

$$\Phi(Z) = \theta(Z)[\Psi(Z) + C] \ m < 0 . \qquad \text{(A-13)}$$

If the solution is required to vanish at infinity, P_m is replaced by P_{m-1} in equation (A-12) and $C=0$ in

equation (A-13). Furthermore, for the case where $m < 0$, it is required for the analyticity of $\Phi(Z)$ at infinity that

$$C = -\Psi^-(-i) ,$$

and $\int_{-\infty}^{\infty} \frac{g(X')\mathrm{d}X'}{\theta^+(X')(X'+i)^k} = 0 \ (K=2,\ldots,-m) . \quad \text{(A-14)}$

Finally we will consider the Riemann boundary-value problem for the segmented real axis. This is necessary when the function $G(X)$ has points of discontinuity on the real axis. The segments consist of the intervals between these points. In the solution to the phase problem where $T(Z)$ has real zeros, the phase function $\alpha(X)$ is not continuous at the zero points. The procedure is similar to the previous case except that the behavior of $\Phi^\pm(X)$ at the points of discontinuity must be considered. Since the solution is rather lengthy, the reader is referred to Gakhov (1966) for the details of the calculation. The solution to the non-homogeneous problem for the special case with $G(X) = -1$ is given by

$$\Phi(Z) = \frac{R(Z)}{2\pi i} \int_{-\infty}^{\infty} \frac{g(X')}{R(X')} \frac{\mathrm{d}X'}{X'-Z} , \qquad \text{(A-15)}$$

where

$$R(Z) = \prod_{j=1}^{N} (Z^2 - X_j^2) , \qquad \text{(A-16)}$$

and the points X_j are the zeros.

References

BLAUROCK, A. E. & STOECKENIUS, W. (1971). *Nature New Biol.* **233**, 152–155.

CALDERON, A. & PEPINSKY, R. (1952). *Computing Methods and the Phase Problem in X-ray Crystal Analysis.* Edited by R. PEPINSKY. State College, Pa.

DETTMAN, J. W. (1965). *Applied Complex Variables.* New York: Macmillan.

GAKHOV, F. D. (1966). *Boundary Value Problems.* London: Pergamon Press.

HOSEMANN, R. & BAGCHI, S. N. (1962). *Direct Analysis of Diffraction by Matter.* Amsterdam: North Holland.

KING, G. I. & WORTHINGTON, C. R. (1971). *Phys. Lett.* **35**, 259–260.

LESSLAUER, W. & BLASIE, J. K. (1972). *Biophys. J.* **12**, 175–190.

MUSKHELISHVILI, N. I. (1953). *Singular Integral Equations.* Groningen: Noordhoff.

NUSSENZVEIG, H. M. (1967). *J. Math. Phys.* **8**, 561–573.

TITCHMARSH, E. C. (1926). *Proc. Lond. Math. Soc.* **25**, 283–296.

TITCHMARSH, E. C. (1939). *The Theory of Functions.* Oxford Univ. Press.

TOLL, J. S. (1956). *Phys. Rev.* **104**, 1760–1772.

WALTHER, A. (1963). *Opt. Acta,* **10**, 41–53.

WORTHINGTON, C. R., KING, G. I. & McINTOSH, T. J. (1973). *Biophys. J.* **13**, 480–494.