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X-ray Holographic Interferometry in the Determination of Planar Multilayer Structures. Theory and Experimental Observations

By W. Lesslauer and J. K. Blasie

Department of Biophysics and Physical Biochemistry, Johnson Research Foundation, University of Pennsylvania, Philadelphia, Pennsylvania 19104, *U.S.A.*

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A theoretical treatment of X-ray diffraction from composite multilayer systems is given. These systems consist of two or three structures placed beside one another, each containing a number of different bimolecular leaflets as unit cells. Phase information for one of the structures in the composite system can be obtained, if another structure in the system is known precisely, from the interference of X-ray photons diffracted by the known structure with photons diffracted by the unknown structure. The experimental feasibility is demonstrated in an X-ray diffraction experiment.

1. Introduction

The bimolecular leaflet of polar lipid molecules is a structure of considerable biological interest. It consists of two monomolecular lipid films facing each other so that a plane of symmetry exists between the two films. Such a bimolecular leaflet may be part of the structure of biological membranes. There are several ways to prepare systems which will be called multilayer structures where a varying number of bimolecular leaflets are stacked on top of each other. X-ray diffraction studies on multilayer structures have been done. Discrete lamellar reflections arising from the electron density distribution along a z axis normal to the plane of the leaflets were obtained (Luzzati, 1968; Levine, Bailey & Wilkins, 1968). Structure determinations in these studies involved either assumed phase angles for the lamellar reflections or the fitting of assumed models to observed X-ray intensity data.

A method by which the phase problem can be solved experimentally for these structures is described in this report. It may be briefly stated as follows. A known and an unknown structure are placed beside each other such that X-ray photons of known phase, diffracted by the known (or reference) structure, interfere with photons of unknown phase diffracted by the unknown structure. Phase information for the unknown multilayer structure may be obtained from the resulting intensity data directly, or the generalized Patterson function may be used to analyze the intensity data. The structures of bimolecular leaflets of fatty acids of varying chain length have been determined (Lesslauer & Blasie, 1971) and they can be used as reference structures.

This method differs from isomorphous replacement to the extent that the reference structure is placed outside of the structure to be investigated.

Consider a planar multilayer structure comprising a stack of identical discs of finite thickness and infinite radius. The electron density distribution of the total structure in cylindrical coordinates (r,φ,z), where z is the stacking direction normal to the plane of the discs, may be expressed as:

$$\varrho(\mathbf{r})=\varrho_{\text{u.c.}}(r,\varphi,z)*l_N(z)\,. \qquad (1)$$

$*$ stands for a convolution operation. $l_N(z)$ is a one-dimensional lattice peak function. In order to keep $l_N(z)$ centrosymmetric in systems with a finite number N of discs, $l_N(z)$ is defined in the following way for N even or odd:

$$l_N(z)=\sum_{i=-(N-1)/2}^{(N-1)/2}\delta(z-i\cdot d)$$

where i is an integer and $\delta(z)$ is a Dirac delta function. d is the repeat interval in the z direction, and $\varrho_{\text{u.c.}}(r,\varphi,z)$ is the electron density distribution in a unit cell.

Since we are concerned only with the lamellar reflections arising from the one-dimensional lattice, we consider $\varrho_{\text{u.c.}}(r,\varphi,z)$ to be cylindrically symmetric about z and write for $\varrho(\mathbf{r})$ (Blaurock & Worthington, 1966):

$$\varrho(\mathbf{r})=\{\sigma(r)\cdot t_{\text{u.c.}}(z)\}*l_N(z) \qquad (2)$$

where $\sigma(r)$ is the electron density distribution in the plane and is a constant and $t_{\text{u.c.}}(z)$ is the projection of $\varrho_{\text{u.c.}}(r,z)$ along the r direction onto the z axis; subscript u.c. refers to unit cell.

The diffracted X-ray intensity $I(r^*,z^*)$ in cylindrical reciprocal space coordinates (r^*,φ^*,z^*) where $\mathbf{z}\cdot\mathbf{z}^*/|\mathbf{z}|\cdot|\mathbf{z}^*|=1$, is:

$$I(r^*,z^*)\propto|\Sigma(r^*)\cdot T(z^*)|^2\cdot S(N,z^*) \qquad (3)$$

where

$$\Sigma(r^*)\Leftrightarrow\sigma(r);\; T(z^*)\Leftrightarrow t(z)\dagger$$

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† $\Sigma(r^*)=\int_0^\infty \sigma(r)\,.\,J_0(2\pi rr^*)\,2\pi r\,\mathrm{d}r$

$T(z^*)=\int_{-d/2}^{+d/2} t(z)\exp\{-2\pi izz^*\}\,\mathrm{d}z\,.$

and

$$S(N,z^*)=\frac{\sin^2(\pi\cdot N\cdot z^*\cdot d)}{\sin^2(\pi\cdot z^*\cdot d)}$$

is the interference function for a one-dimensional crystal. Analysis of the lamellar repeat reflections $I(r^*=0,z^*)=I(z^*)$ provides the electron density distribution of the unit cell through the plane of the discs $t(z)$, if the phase of its transform $T(z^*)$ at each sampling point introduced by $S(N,z^*)$ is known.

A theoretical treatment of diffraction from composite multilayer systems with 2 and 3 different components will be given. The treatment applies to symmetric and asymmetric structures and it may gain importance for the latter case, since, in the former, other direct methods can solve the phase problem, provided diffraction from systems with a small number of unit cells is obtained for such structures.

2. Treatment of systems with two structures

A composite multilayer system consisting of a known and an unknown multilayer structure is considered first. The electron density distribution of the composite multilayer structure along the z direction is called $g(z)$. If the center of mass of the known structure is defined as $z=0$,

$$g(z)=[t_k(z)*l_{N,k}(z)]*\delta(z)+[t_u(z)*l_{N,u}(z)]*\delta\left(z-\left\{\frac{N_kd_k+N_ud_u}{2}\right\}\right) \quad (4)$$

where the subscripts k and u refer to the known and unknown multilayer structures and the convolution of these structures with the delta functions places them at their appropriate z coordinate. We shall assume $N_{k,u}$ to be even. For N_k or N_u odd, $N_{k,u}$ has to be replaced by $(N_{k,u}+1)$ everywhere.

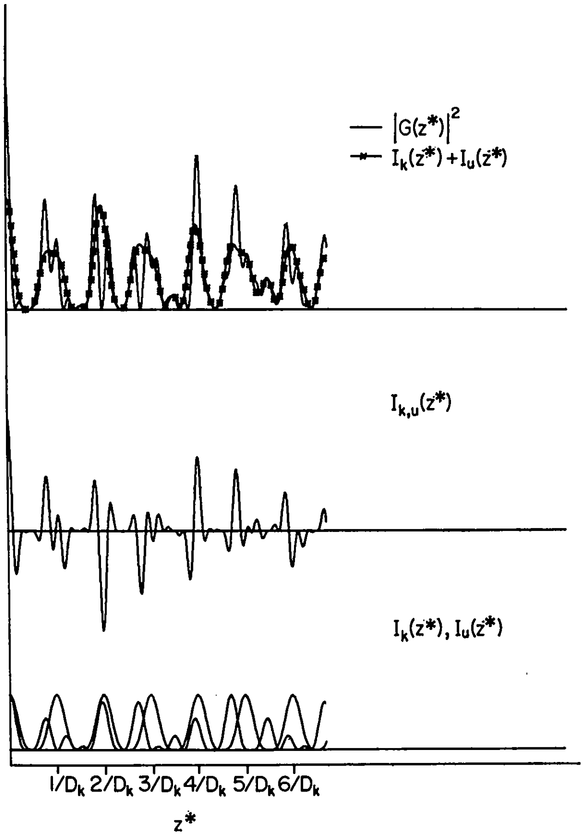

Fig. 1. Diffracted intensity computed for a composite multilayer structure of two unit cells of phosphatidylcholine and two unit cells of barium stearate. I_k, I_u, $I_{k,u}(z^*)$ correspond to the first, second and third terms of equation (6), respectively. $t_k(z)$ and $t_u(z)$ were based on models exhibiting the essential features of barium stearate and phosphatidylcholine bimolecular leaflets.

The intensity $I(z^*)$ diffracted by the composite multilayer is proportional to $|G(z^*)|^2$, where $G(z^*)$ is the one-dimensional Fourier transform of $g(z)$, and

$$\begin{aligned}|G(z^*)|^2=&|T_k(z^*)|^2|L(N_k,z^*)|^2+|T_u(z^*)|^2|L(N_u,z^*)|^2\\&+2|T_k(z^*)||L(N_k,z^*)||T_u(z^*)||L(N_u,z^*)|\\&\times\cos([\psi_k-\psi_u]+[2\pi z^*A_{ku}])\end{aligned} \quad (5)$$

$$\left\{\begin{aligned}&|L(N_{k,u},z^*)|^2=S_{k,u}(N_{k,u},z^*);L(N_{k,u},z^*)\Leftrightarrow l_{N,k,u}(z);\\&T_{k,u}(z^*)=|T_{k,u}|\exp\{-i\alpha_{k,u}\};\\&L(N_{k,u},z^*)=|L_{k,u}|\exp\{-i\beta_{k,u}\};\\&\psi_{k,u}=(\alpha_{k,u}+\beta_{k,u});\\&A_{k,u}=(N_kd_k+N_ud_u)/2\,.\end{aligned}\right\}$$

$\alpha_{k,u}$ is the phase of the unit-cell transform $T_{k,u}$. $\beta_{k,u}$ is the phase of the lattice transform $L_{k,u}$ and $\psi_{k,u}$ is the phase of the 'total structure transform', $T_{k,u}(z^*)\cdot L_{k,u}(z^*)$. We note that $I(z^*)$ data from the known and the unknown structure alone independently provide the first two terms in equation (5) and all the factors in the last term except $\cos([\psi_k-\psi_u]+[2\pi z^*A_{k,u}])$, since $L(N_{k,u},z^*)$ may be directly calculated for known values of $N_{k,u}$ and $d_{k,u}$. The number of unit cells N_k and N_u must be small to ensure overlap of the maxima of L_k and L_u in order to provide appreciable contribution from the third term to $|G(z^*)|^2$, unless d_k and d_u are matched exactly.

We consider the factor $\cos([\psi_k-\psi_u]+[2\pi z^*A_{k,u}])$ for two cases, $t_u(z)$ centrosymmetric *versus* $t_u(z)$ asymmetric. We note that $l_{N,k}(z)$ and $l_{N,u}(z)$ are centrosymmetric for $N_{k,u}$ even or odd and hence $\beta_{k,u}=0,\pi$ as determined by $N_{k,u}$ and $d_{k,u}$ for any z^*.

$t_u(z)$ *centrosymmetric*

Since the reference structure $t_k(z)$ is centrosymmetric also, $\sin(\psi_k-\psi_u)=0$ and $\cos(\psi_k-\psi_u)$ is either $+1$ or -1, depending on whether $\psi_k=\psi_u$ or $\psi_k\neq\psi_u$ where both $\psi_{k,u}=0,\pi$ only. Then equation (5) becomes (the arguments z^* will be dropped for the functions G, T, and L in the following discussion):

$$|G|^2=|T_k|^2|L_k|^2+|T_u|^2|L_u|^2\pm2|T_k||L_k||T_u||L_u|\times\cos(2\pi z^*A_{k,u}) \quad (6)$$

where the ± uncertainty is the only unknown in this expression for $|G|^2$. The first two terms are the diffraction of the known and unknown structures alone, while the third term arises from the interference of photons diffracted by the known structure with those diffracted by the unknown structure. This interference term either adds to or subtracts from the first and second terms, depending on whether the phase difference $(\psi_k-\psi_u)$ for the 'total structure transforms' $(T_k \cdot L_k)$ and $(T_u \cdot L_u)$ is 0 or π, respectively. The sign of $\cos(2\pi z^* A_{k,u})$ at a particular diffraction angle, $z^*=(2\sin\theta)/\lambda$, is known.

The diffracted intensity $I(z^*)$ of the known and the unknown structure alone (*i.e.* $|T_k|^2 \cdot |L_k|^2$ and $|T_u|^2 \cdot |L_u|^2$) can be obtained independently. A direct comparison of these scaled† intensities with those from the composite structure determines whether $(\psi_k-\psi_u)=0$ or π for the experimentally accessible range of z^*. Since β_u is known for all z^*, the phase α_u of the unit cell transform of the unknown structure can be obtained. Fourier transformation of $|T_u| \cdot \exp\{-i\alpha_u\}$ finally gives $t_u(z)$.

Fig. 1 shows a computed example of the effect of the interference term in the calculated diffracted intensity $I(z^*)$ for a composite multilayer structure. Models of barium stearate and phosphatidyl choline multilayer structures were used in the computation for $t_k(z)$ and $t_u(z)$, respectively. The effect of the interference term is seen to depend on the relative phases $(\psi_k-\psi_u)$ of the two 'total structure transforms' $(T_k \cdot L_k)$ and $(T_u \cdot L_u)$ in the manner described above.

$t_u(z)$ asymmetric

For an asymmetric $t_u(z)$ the phase of the total structure transform ψ_u can have all values $0 \leq \psi_u \leq 2\pi$. The reference structure is again centrosymmetric, (*i.e.* $\psi_k=0,\pi$). The observed $I(z^*)$ can be compared with calculated values of $|G|^2$ in equation (5) using the known values for $(|T_{k,u}| \cdot |L_{k,u}|)$, $A_{k,u}$ and ψ_k and varying values of ψ_u until $|G|^2$ matches observed $I(z^*)$. Two values for ψ_u will give an equally good fit. The more probable of the two values may be chosen by intuition, or a second reference structure will resolve the ambiguity in sign. Repeating this process for all z^* values for which $I(z^*)$ is available will determine ψ_u in that range of z^* and $t_u(z)$ can be determined by a Fourier transformation of $|T_u| \cdot \exp\{-i\alpha_u\}$ since β_u is known for all z^* and $\psi_u=\alpha_u+\beta_u$.

3. Treatment of systems with three structures

For reasons that will become apparent below, we introduce an additional multilayer structure acting as a spacer between the reference and unknown structures. We then have for $g(z)$ of the composite multilayer structure

$$\begin{aligned} g(z) &= [t_k(z)*l_{N,k}(z)]*\delta(z) \\ &\quad + [t_s(z)*l_{N,s}(z)]*\delta(z-A_{k,s}) \\ &\quad + [t_u(z)*l_{N,u}(z)]*\delta(z-A_{k,u}) \end{aligned} \tag{7}$$

with

$$A_{k,s}=(N_k d_k+N_s d_s)/2;$$

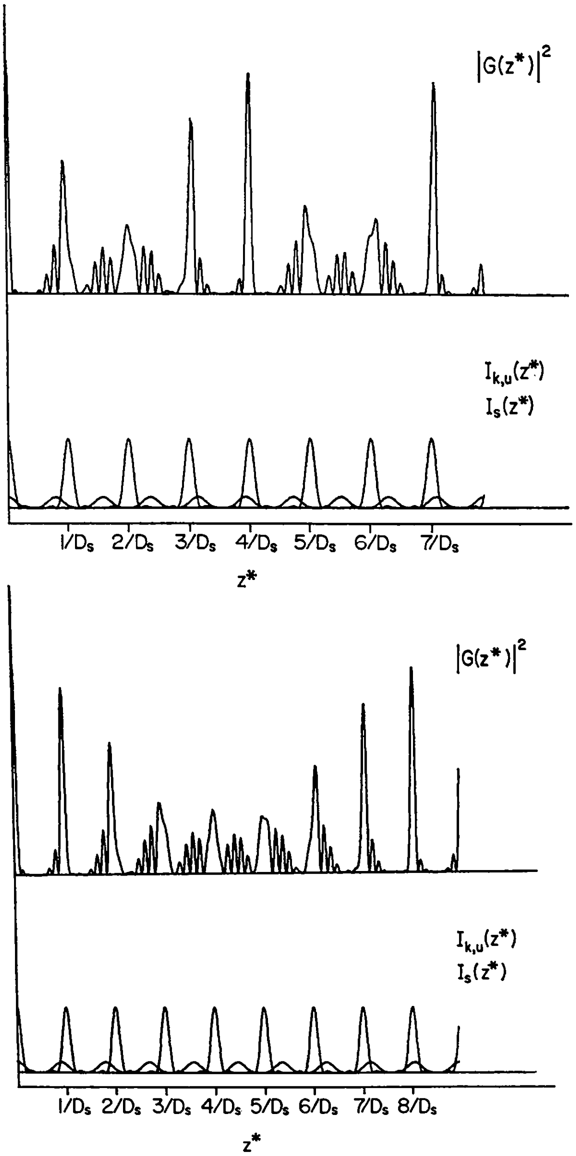

Fig. 2. Diffracted intensities computed for composite multilayer systems with 2 unit cells of barium stearate as known structure and two unit cells of barium stearate as 'unknown' structure. The spacer between the known and unknown structure were (*a*) 5 unit cells of barium myristate, or (*b*) 5 unit cells of barium palmitate. $I_{k,u}(z^*)$ correspond to the first and second terms of equation (10). $I_s(z^*)$ corresponds to the diffracted intensity from the spacer alone at z^* values of h/D_s where D_s is the unit-cell dimension of the spacer structure along z.

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† Correction and scaling factors must be allowed for in the observed diffracted intensities. The scaling of the different diffraction patterns relative to each other may be accomplished by incorporation of an additional structure whose diffraction occurs at angles outside the range of z^* which is of interest for the systems under discussion (*e.g.* multilayers may be built up on evaporated silver mirrors of standard thickness and the diffraction from the silver can be used for scaling).

$$A_{k,u}=N_s d_s+(N_k d_k+N_u d_u)/2$$

and

$$A_{s,u}=A_{k,u}-A_{k,s}\ .$$

We again choose $N_{k,u,s}$ even. For N_k, N_u or N_s odd, $N_{k,u,s}$ has to be replaced by $(N_{k,u,s}+1)$. We obtain for the Fourier transform of $g(z)$:

$$\begin{aligned}|G|^2=&|T_k|^2|L_k|^2+|T_s|^2|L_s|^2+|T_u|^2|L_u|^2\\&+2|T_k|\cdot|L_k|\cdot|T_s|\cdot|L_s|\\&\times\cos\left[(\psi_k-\psi_s)+(2\pi z^* A_{k,s})\right]\\&+2|T_s|\cdot|L_s|\cdot|T_u|\cdot|L_u|\\&\times\cos\left[(\psi_s-\psi_u)+(2\pi z^* A_{s,u})\right]\\&+2|T_k|\cdot|L_k|\cdot|T_u|\cdot|L_u|\\&\times\cos\left[(\psi_k-\psi_u)+(2\pi z^* A_{k,u})\right]\ .\end{aligned}\tag{8}$$

Equation (8) can be simplified with the following approximations. Consider the case where $d_k \simeq d_u$ and both d_k and d_u are considerably different from d_s. Furthermore, we choose N_k and N_u small (~ 2) and N_s larger (≥ 5). $|G|^2$ has then in certain regions of z^* very small relative contributions from the second, fourth, and fifth terms in equation (8) since the maxima of L_s are narrow and the products $|L_k|\cdot|L_s|$ and $|L_s|\cdot|L_u|$ disappear practically everywhere owing to the narrowness of L_s and the inequalities $d_s \neq d_{u,k}$. In these regions of z^*, $|G|^2$ becomes then approximately

$$\begin{aligned}|G|^2\simeq&|T_k|^2|L_k|^2+|T_u|^2\cdot|L_u|^2+2|T_k|\cdot|L_k|\cdot|T_u|\cdot|L_u|\\&\times\cos\left[(\psi_k-\psi_u)+(2\pi z^* A_{k,u})\right]\ .\end{aligned}\tag{9}$$

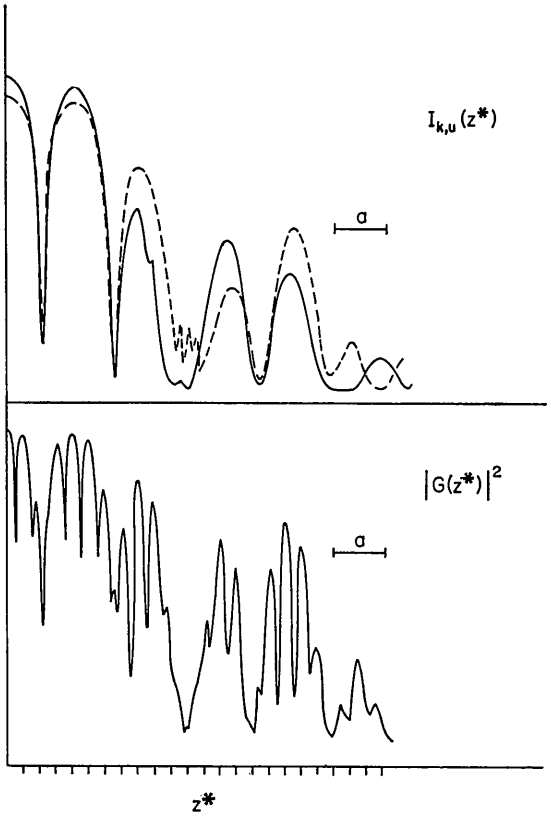

Fig. 3. Densitometer tracings of a light diffraction experiment; $t_k(z)$ and $t_u(z)$ are slits of nearly equal width; $N_k=N_u=2$. $I_{k,u}(z^*)$ correspond to the first and second terms of equation (10). $|G(z^*)|^2$ shows the interference fringes of the third term of equation (10). Intervals of $h/A_{k,u}$ are marked on the z^* axis. In the region of z^* indicated by a, T_k and T_u are out of phase for the first time. The fringes in $|G(z^*)|^2$ are shifted by $1/2\cdot A_{k,u}$ relative to those outside a.

A computed example is shown in Fig. 2. $|G|^2$ is calculated for a composite multilayer structure with five unit cells of either barium palmitate of barium myristate as spacer structure. The known and unknown structures were identical for this calculation, and comprised two unit cells of barium stearate each. With myristate as spacer structure the approximations for equation (9) are satisfied in the intervals $1/d_s \leq z^* \leq 3/d_s$ and $4/d_s \leq z^* \leq 7/d_s$, and with palmitate as spacer in $2/d_s \leq z^* \leq 6/d_s$. The fringes seen in these intervals arise from the third term in equation (9). Using fatty acids of proper chain length for the spacer and reference structures, we may choose the z^* regions in which these fringes can be seen in an unperturbed condition.

$t_u(z)$ centrosymmetric

Since $t_k(z)$ is also centrosymmetric, equation (9) becomes for the appropriate conditions mentioned above:

$$\begin{aligned}|G|^2=&|T_k|^2\cdot|L_k|^2+|T_u|^2\cdot|L_u|^2\\&\pm 2\cdot|T_k|\cdot|L_k|\cdot|T_u|\cdot|L_u|\cdot\cos(2\pi z^* A_{k,u})\end{aligned}\tag{10}$$

for reasons identical to those mentioned in § 2. The only difference between equations (10) and (6) is that the separation $A_{k,u}$ includes now the spacer structure. This results in a frequency for $\cos(2\pi z^* A_{k,u})$ which is considerably greater than the fluctuations in $|T_k|$, $|L_k|$, $|T_u|$ and $|L_u|$. The fringes introduced in $|G|^2$ by this cosine term are readily recognizable. The position of these fringes relative to the origin $z^*=0$ depends on the sign of the third term.

For $\psi_u=\psi_k$, the sign is positive and the fringes occur at $h/A_{k,u}$, where h is an integer, relative to the origin. For $\psi_u \neq \psi_k$, *i.e.* $(\psi_k-\psi_u)=\pi$, the fringes are displaced by $1/2\cdot A_{k,u}$ from the positions for $\psi_u=\psi_k$.

Hence, by determining the positions of the fringes relative to the origin in $I(z^*)$ from such a composite multilayer structure, the relative 'total structure phase' $(\psi_k-\psi_u)$ may be determined over a large range of z^* using different spacer structures. Then the phase of T_u and hence $t_u(z)$ may be determined in the fashion discussed in § 2.

Such fringe shifts are demonstrated in the following optical analogue experiment (Fig. 3). $t_k(z)$ and $t_u(z)$ were slits of finite height and of nearly equal width. $N_k=N_u=2$. No spacer structure as such was used; the two double slits $[t_k(z)*l_k(z)]$ and $[t_u(z)*l_u(z)]$ were merely physically separated on the mask. Hence, equation (10) was satisfied for all z^*. The optical transforms $(T_k\cdot L_k)$ and $(T_u\cdot L_u)$ are shown. The fringes arising from $\cos(2\pi z^* A_{k,u})$ are easily recognizable. The appropriate shift in the position of these fringes for $\psi_k \neq \psi_u$ relative to those for $\psi_k = \psi_u$ is clearly shown.

$t_u(z)$ *asymmetric*

The method we propose here applies as well for $t_u(z)$ centrosymmetric. Equation (9) may be obtained in a different fashion. $|G|^2$ and independently the second, fourth, and fifth terms in equation (8) can be determined experimentally. If we choose N_k, N_u and N_s all small (*e.g.* 2–3) and $d_k \simeq d_u \neq d_s$ then $|G|^2$ is sufficiently slowly varying that the scaled second, fourth, and fifth terms can be subtracted from $|G|^2$ to give an expression similar to equation (9)

$$\begin{aligned}|G'|^2 = |T_k|^2 \cdot |L_k|^2 + |T_u|^2 \cdot |L_u|^2 \\ + 2 \cdot |T_k| \cdot |L_k| \cdot |T_u| \cdot |L_u| \cdot \\ \times \cos\left[(\psi_k - \psi_u) + (2\pi z^* A_{k,u})\right] \qquad (11)\end{aligned}$$

The second term of equation (8), $|T_s|^2 \cdot |L_s|^2$ is obtained from diffraction by the spacer structure alone, while the fourth and fifth terms may be determined experimentally from two-structure composite multilayers with just the known plus spacer and just the spacer plus unknown structures. We may then take the inverse integral Fourier transform of $|G'|^2$ to give the generalized Patterson function $P(z)$ for $0 \leq z^* \leq z_0^*$ and N_s not too large that the true linewidth of cos $(2\pi z^* A_{k,u})$ and sin $(2\pi z^* A_{k,u})$ can be observed.

$$\begin{aligned}P(z) = [t_k(z) * t_k(-z)] * [l_{N,k}(z) * l_{N,k}(-z)] * \delta(z) \\ + [t_u(z) * t_u(-z)] * [l_{N,u}(z) * l_{N,u}(-z)] * \delta(z) \\ + [t_k(-z) * l_{N,k}(-z)] * [t_u(z) * l_{N,u}(z)] * \delta(z - A_{k,u}) \\ + [t_k(z) * l_{N,k}(z)] * [t_u(-z) * l_{N,u}(-z)] * \delta(z + A_{k,u}) . \\ (12)\end{aligned}$$

For $N_{k.u}$ small (*e.g.*, $N_k = N_u = 2$) and $N_s < 5$ (*e.g.*, $N_s = 2$) and $d_k \simeq d_u \neq d_s$ the first two terms in equation (11) are zero for $|z| > 2d_{k,u}$. Thus, for $|z| \geq A_{k,u} > 2d_{k,u}$ the inverse Fourier transform of $|G'|^2$ is just the convolution of the known structure with the unknown structure. This second Fourier transformation has 'reconstructed' the convolution of the unknown structure with the known structure instead of just the unknown structure itself as in off-axis Fourier transform holography (Smith, 1969). For the latter, we would require a narrow slit as our known structure whose width is small relative to d_u analogous to the pinhole suggested by Winthrop & Worthington (1965). The unknown structure $t_u(z)$ may then be separated from this convolution *via* a suitable recursion process (Hosemann & Bagchi, 1962) for $t_u(z)$ either centrosymmetric or asymmetric.

4. X-ray diffraction experiments

Multilayer systems of fatty acids of phospholipids can be formed by passing a clean metal surface repeatedly through a monomolecular layer of the desired molecule(s) spread on the surface of an electrolyte solution (Blodgett, 1935). The planes of the bimolecular leaflets are parallel to the plane of the metal surface. The z axis is normal to the metal surface. The number of unit cells is controlled by the number of passes through the monolayer. Composite systems can be formed if N_u layers of molecules of the sort u are deposited on top of N_k layers of molecules of the sort k. In this way diffracting systems composed of different structures with a defined geometric relation to each other are obtained.

A precise knowledge of the electron density distribution of the reference structure is essential to apply the methods outlined above to determine the phase angles of an unknown structure. Fatty acid multilayers are suitable reference structures for the systems under discussion. Diffraction data have been obtained from as few as two unit cells of barium stearate and a direct determination of its structure has been performed (Lesslauer & Blasie, 1971). The projected electron density distribution $t_{\text{u.c.}}(z)$ was determined using 9 lamellar reflections of a 47 Å repeat. The layer of Ba^{2+} ions, the methylene chains, and the terminal methyl groups were easily recognizable in $t_{\text{u.c.}}(z)$. This provides a reference structure and shows that it is possible to observe diffraction from systems with a controlled and small number of unit cells, which is important both for the case of systems with two structures (*viz.* the reference structure) and for that of 3 structures (*viz.* spacer and reference structure).

Fig. 4 illustrates in an X-ray diffraction experiment the interference fringes predicted by the third term in equation (10) for a composite structure of 2 unit cells of barium stearate (reference structure), 5 unit cells of barium myristate (spacer) and 2 unit cells of barium stearate (unknown structure). This corresponds to the computed example in Fig. 2. The fringes can be seen most clearly in the range $2/d_s < z^* < 3/d_s$. The fringes occur at the appropriate z^* positions of $h/A_{k,u}$ as predicted by equation (9). In this example the reference and the unknown structure are identical and their transforms are necessarily everywhere in phase. It is, therefore, impossible to observe the displacement of the fringes by $1/2 \cdot A_{k,u}$ for the z^* regions where T_k and T_u are out of phase. The conclusion $\psi_u = \psi_k$ is trivial, because $t_u(z) = t_k(z)$. Despite this, the experiment in Fig. 4 demonstrates that the interference term in equations (5) and (9) can be experimentally observed in X-ray diffraction. The relative magnitude of these fringes is in qualitative agreement with Fig. 2. A precise determination of the intensities is more difficult as background scattering extends into this region.

The X-ray diffraction pattern in Fig. 4 contains phase information out to the 5th lamellar reflection. It is expected that the phase angles may be determined by this method in the whole z^* range where diffraction from systems with only a few unit cells is observed.

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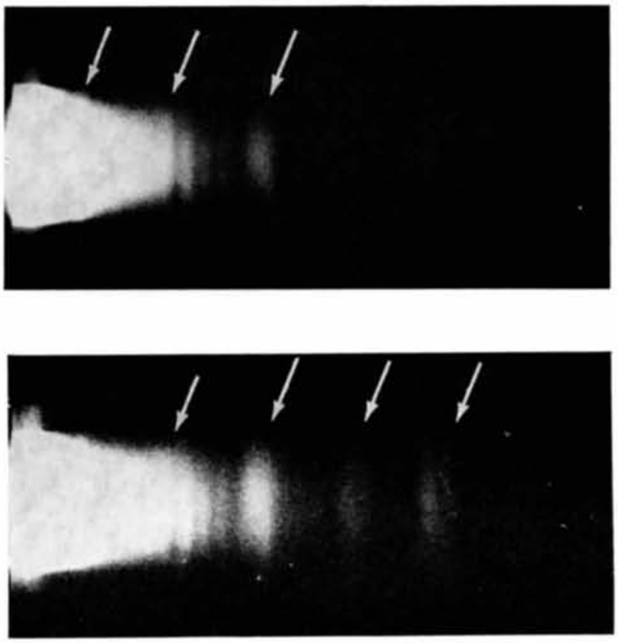

Fig. 4. X-ray diffraction from a composite multilayer consisting of 2 unit cells of barium stearate (known structure), 15 unit cells of barium myristate (spacer), and 2 unit cells of barium stearate (unknown structure). 3 fringes can be seen between $2/D_s$ and $3/D_s$. They arise from the third term in equation (10) as seen in Fig. 2. The observed intensity in the regions $3/D_s$, $4/D_s$, and $5/D_s$ can be interpreted in terms of Fig. 2 if correction factors are allowed for. (Arrows mark diffraction peaks from the spacer structure at h/D_s.)

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The Resolution Function of a Slow Neutron Rotating Crystal Time-of-Flight Spectrometer I. Application to Phonon Measurements

By A. Furrer

Delegation für Ausbildung und Hochschulforschung am Eidg. Institut für Reaktorforschung, Wuerenlingen, Switzerland

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The resolution function of a slow neutron rotating crystal time-of-flight spectrometer applied to phonon measurements is treated analytically. It is demonstrated that every component of the instrument may contribute to the uncertainty of the time-of-flight measurement. Focusing conditions are derived leading to the concept of removable and irremovable time-of-flight spreads. No approximations are made concerning the phonon dispersion surface. Experimental evidence is presented to support the resolution functions calculated on the basis of this theory.

1. Introduction

In the course of the past decade the scattering of thermal neutrons has proved to be one of the most versatile experimental techniques for studying dynamics and structure of solids and liquids. Fig. 1 shows a schematic sketch of a slow neutron spectrometer. The essential parts are the neutron source, the monochromator system, the sample, and the analyser system, elements of which are denoted by the indices 0, 1, 2, 3 according to the order in which they are located along the neutron flight-path. Intermediate elements such as collimators C, flight-paths L, and neutron wave-vectors $\mathbf{k}$ have double indices. The experimental spectrum $I(\mathbf{Q},\omega)$ observed by the analyzer is given by the convolution integral

$$I(\mathbf{Q},\omega)=\iint R(\mathbf{Q}'-\mathbf{Q},\omega'-\omega)\sigma(\mathbf{Q}',\omega')\mathrm{d}\mathbf{Q}'\mathrm{d}\omega'\ , \quad (1\cdot1)$$

where $R(\mathbf{Q},\omega)$ is the instrumental resolution function and $\sigma(\mathbf{Q},\omega)$ is the unknown scattering cross section. The variables $\mathbf{Q},\omega$ are defined by the momentum transfer

$$\hbar\mathbf{Q}=\hbar(\mathbf{k}_{12}-\mathbf{k}_{23}) \quad (1\cdot2)$$

and by the energy transfer

$$\hbar\omega=\frac{\hbar^2}{2m}(k_{12}^2-k_{23}^2)\ , \quad (1\cdot3)$$

where m denotes the neutron mass. The problem is to evaluate $\sigma(\mathbf{Q},\omega)$ by an unfolding procedure, provided $R(\mathbf{Q},\omega)$ is known. The most accurate method of determining $R(\mathbf{Q},\omega)$ would be a direct measurement requiring no prior knowledge of instrumental parameters. This, however, is usually impossible except for zero energy transfer, so that in general $R(\mathbf{Q},\omega)$ will have to be calculated. This was done analytically by Collins (1963), Peckham, Saunderson & Sharp (1967),

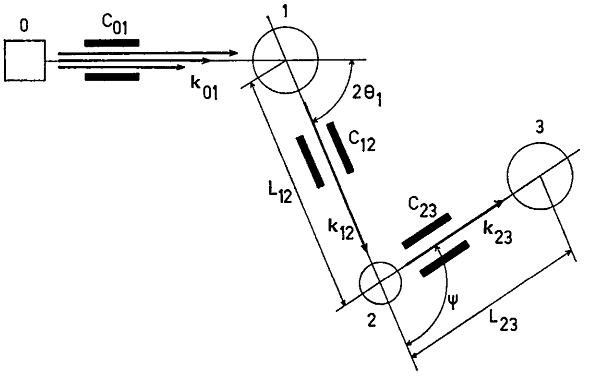

Fig. 1. Schematic sketch of a slow neutron spectrometer. 0: neutron source, 1: monochromator system, 2: sample, 3: analyser system.