and

$$J_{ij}(12)=(\nabla_1-\nabla\ )_i(\nabla_2-\nabla_2')_j[\langle\psi^+(1')\psi(1)\times\psi^+(2)\psi(2')\rangle-\langle\psi^+(1')\psi(1)\rangle\times\langle\psi^+(2)\psi(2')\rangle]$$
$$=(-1)\left(\frac{2m}{ie}\right)^2[\langle j_i^{(0)}(1)j_j^{(0)}(2)\rangle-\langle j_i^{(0)}(1)\rangle\langle j_j^{(0)}(2)\rangle], \quad \text{(III-8)}$$

where we introduced the density operator $\varrho(1)=\psi^+(1)\psi(1)$ and the current operator $j^{(0)}$ defined by equation (IIA-7). From equation (III-7), it is obvious that $iS(12)$ is related to the generalized dielectic function (including the core electrons).

References

ASHKIN, M. & KURIYAMA, M. (1966). *J. Phys. Soc. Japan*, **21**, 1549.
BARTON, G. (1963). *Introduction to Advanced Field Theory*, Ch. 4. New York: Interscience.
BJORKEN, J. D. & DRELL, S. D. (1965). *Relativistic Quantum Fields*, Ch. 16. New York: McGraw-Hill.
DUMOND, J. W. M. (1929). *Phys. Rev.* **33**, 643.
HEDIN, L. (1965). *Phys. Rev.* **139**, A796.
HEISENBERG, W. (1942). *Z. Phys.* **120**, 513, 678.
KUBO, R. (1959). *Lectures in Theoretical Physics*, Vol. 1, p. 120. Edited by W. BRITTIN, New York: Interscience.
KURIYAMA, M. (1967). *J. Phys. Soc. Japan*, **23**, 1369.
KURIYAMA, M. (1968). *J. Phys. Soc. Japan*, **25**, 846.
KURIYAMA, M. (1970). *Acta Cryst.* A**26**, 56.
KURIYAMA, M. & ALEXANDROPOULOS, N. G. (1971). *J. Phys. Soc. Japan*, **31**, .
MIZUNO, Y. & OHMURA, Y. (1967). *J. Phys. Soc. Japan*, **22**, 445.
LEHMAN, H., SYMANZIK, K. & ZIMMERMANN, W. (1955). *Nuovo Cim.* **1**, 1425.
LEHMAN, H., SYMANZIK, K. & ZIMMERMAN, W. (1957). *Nuovo Cim.* **6**, 319.
LOW, F. (1955). *Phys. Rev.* **97**, 1392.
SCHWEBER, S. S. (1962). *An Introduction to Relativistic Quantum Field Theory*, Chs. 17*d* and 18*d*. New York: Harper & Row.
SCHWINGER, J. (1951*a*). *Proc. Nat. Acad. Sci. U.S.A.* **37**, 452.
SCHWINGER, J. (1951*b*). *Proc. Nat .Acad. Sci. U.S.A.* **37**, 455.
SUZUKI, T. (1967). *J. Phys. Soc. Japan*. **22**, 1139.
SUZUKI, T., KISHIMOTO, T., KAJI, T. & SUZUKI, T. (1970). *J. Phys. Soc. Japan*, **29**, 730.
ZIMAN, J. M. (1969). *Elements of Advanced Quantum Theory*, Ch. 4.4. Cambridge Univ. Press.

Acta Cryst. (1971). A**27**, 647

The One-Dimensional Anti-Phase Domain Structures. I. A Classification of Structure and the Patterson Method Applied to the Layer Sequence Determination

BY JIRO KAKINOKI AND TERUAKI MINAGAWA†

Department of Physics, Faculty of Science, Osaka City University, 459 *Sugimoto-cho, Sumiyoshi-ku, Osaka, Japan*

(*Received* 5 *February* 1971 *and in revised form* 6 *May* 1971)

The one-dimensional anti-phase domain structures with an out-of-step vector $\mathbf{u}=(\mathbf{a}+\mathbf{b})/2$ are classified into the following three kinds: (1) The complex out-of-step structure, (2) the complex *APD* (antiphase domain) structure, (3) the simple *APD* structure. These structures are characterized by the use of the similar symbols to the Zhdanov symbol. Intensity formulae are derived for some typical cases. The application of the Patterson method gives some useful relations between the symbol adopted and a quantity which is obtained by Fourier cosine transformation of the unitary intensities. Since this quantity is any one of a set of integers of the form (P^2-4qP) (P: period, q: integer), the correct layer sequence may be obtained even if the observed intensities are not so accurate. Applications for some ideal and real cases are shown.

1. The unitary intensity

An example of the one-dimensional anti-phase domain structures of A_3B-type with an out-of-step vector,

$$\mathbf{u}=\frac{(\mathbf{a}+\mathbf{b})}{2}, \quad (1)$$

is shown in Fig. 1, where the out-of-steps occur along the **c** direction at every four unit cells, and the structure consists of two kinds of unit cells as shown in Fig. 2. The structure factor of the unit cell shown in Fig. 2(a), which is denoted by V_0, is expressed as

$$V_0=f_B+f_A[\exp\{\pi i(\xi+\eta)\}+\exp\{\pi i(\eta+\zeta)\}+\exp\{\pi i(\zeta+\xi)\}]$$

where f_A and f_B are the atomic scattering factors of A and B atoms, respectively, and ξ, η and ζ are the par-

† Present address: Department of Physics, Osaka Kyoiku University, Tennoji, Osaka, Japan.

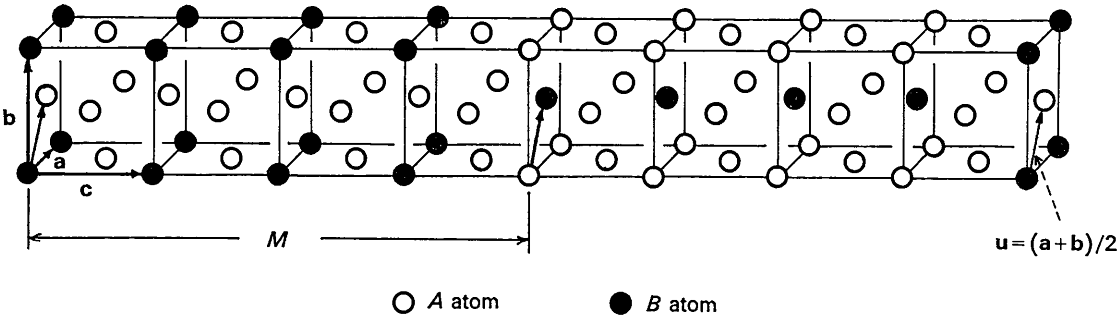


Fig. 1. An example of the one-dimensional anti-phase domain structure of A_3B-type with an out-of-step vector $\mathbf{u}=(\mathbf{a}+\mathbf{b})/2$ and with half-period $M=4$.

ameters along $\mathbf{a}^*$, $\mathbf{b}^*$ and $\mathbf{c}^*$, respectively; $\mathbf{a}^*$, $\mathbf{b}^*$ and $\mathbf{c}^*$ are the reciprocal vectors of $\mathbf{a}$, $\mathbf{b}$ and $\mathbf{c}$, respectively. On the other hand, the structure factor of the unit cell shown in Fig. 2(*b*), which is obtained from V_0 by displacing the structure by the out-of-step vector, $\mathbf{u}$, is given by

$$V_0'=\varepsilon V_0$$

where ε is the phase factor corresponding to the out-of-step vector, $\mathbf{u}$, and is expressed as

$$\varepsilon=\exp\{\pi i(\xi+\eta)\}\,.$$

By the use of V_0 and V_0', the intensity of X-rays diffracted by the crystal can be expressed in electron units as

$$I^{(0)}=V_0V_0^*G_1^2(\xi)G_2^2(\eta)I(\varphi) \tag{2}$$

with

$$G_1(\xi)=\frac{\sin \pi K\xi}{\sin \pi\xi} \quad \text{and} \quad G_2(\eta)=\frac{\sin \pi L\eta}{\sin \pi\eta}$$

where we assume that there are K and L unit cells along the $\mathbf{a}$ and $\mathbf{b}$ directions, respectively. The last factor in equation (2), $I(\varphi)$, is called the unitary intensity and expressed as

$$I(\varphi)=\Psi(\varphi)\Psi^*(\varphi)=\left|\sum_{n=0}^{N-1}\varepsilon_n \exp(in\varphi)\right|^2, \quad \varphi=2\pi\zeta \tag{3}$$

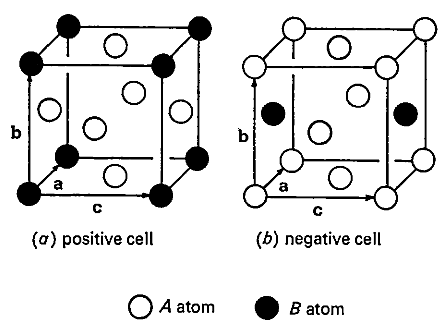


Fig. 2. Two kinds of unit cells in the one-dimensional anti-phase domain structure of A_3B-type shown in Fig. 1.

where

$$\varepsilon_n=\begin{cases}1 & \text{when the } n\text{th cell is } V_0\\ \varepsilon & \text{when the } n\text{th cell is } V_0'\end{cases}$$

and we assume that there are N unit cells along the $\mathbf{c}$ direction.

Since we have two Laue functions, $G_1^2(\xi)$ and $G_2^2(\eta)$, in equation (2), the crystal can be considered to have a layer structure and hence we have only to pay attention to the cases corresponding to

$$\begin{cases}\xi=h & h\colon 0,\ \pm1,\ \pm2,\ \pm3,\ \ldots\\ \eta=k & k\colon 0,\ \pm1,\ \pm2,\ \pm3,\ \ldots.\end{cases}$$

As a result, ε_n in equation (3) becomes

$$\varepsilon_n=\begin{cases}1 & \text{when the } n\text{th layer is the positive one}\\ (-1)^{h+k} & \text{when the } n\text{th layer is the negative one}\end{cases} \tag{4}$$

where *positive* and *negative* correspond to V_0 and V_0', respectively.

2. A classification of the structures

Similarly to the Zhdanov symbol for the close-packed structures, a layer sequence symbol to specify the structure of a one-dimensional anti-phase domain structure is conveniently defined by a set of positive and negative numbers such as

$$(a_1\bar{b}_1a_2\bar{b}_2a_3\bar{b}_3\,\ldots\,a_t\bar{b}_t)$$

with a period

$$P=\sum_{i=1}^{t}(a_i+b_i)$$

where a_i and b_i are the numbers of successive positive and negative layers, respectively. The corresponding structure will be hereafter called a *complex out-of-step structure.*

If P is even *i.e.* $P=2M$ and if the sign of each layer in the last M layers is opposite to that of the corresponding layer in the first M layers, the structure is called a *complex APD* (*anti-phase domain*) *structure* and denoted as $([M] \mid [\bar{M}])$ with $[M]=(a_1\bar{b}_1a_2\bar{b}_2\ldots a_s\bar{b}_sa_{s+1})$.* The vertical line in $([M] \mid [\bar{M}])$ means that the last $[\bar{M}]$ is obtained from the first $[M]$ only by changing all the signs; corresponding to this change in signs a horizontal bar is put on as $[\bar{M}]$. Half a period M is given by

$$M=\sum_{i=1}^{s+1} a_i+\sum_{i=1}^{s} b_i \quad \text{with} \quad P=2M\,.$$

By comparing the symbol with that of the complex out-of-step structure, we have the relation

$$2s+1=t. \tag{5}$$

The simplest structure of the complex *APD* structure is obtained by putting $s=0$ and $a_1=M$. This structure is called a *simple APD structure* and denoted as $(M \mid \bar{M})$ with $P=2M$.

According to the above classification of the structures, the structure shown in Fig. 1 is a simple *APD* structure, $(4 \mid \bar{4})$ with $P=8$. A standard structure, which Fujiwara (1957) adopted when he analyzed the structure with $M=1{\cdot}8$,† is given by $((2\bar{2}2\bar{2}1) \mid (\bar{2}2\bar{2}2\bar{1}))$ *i.e.* $([9] \mid [\bar{9}])$ with $[9]=(2\bar{2}2\bar{2}1)$ and $P=18$ and hence belongs to the complex *APD* structure. Fujiwara also considered other structures slightly deviated from the standard structure. One of them is $(2\bar{2}2\bar{2}1\bar{2}2\bar{2}1\bar{2})$ which belongs to the complex out-of-step structure with $P=18$.

3. Calculations of the unitary intensity

If a complex out-of-step structure has a period P, the unitary intensity defined by equation (3) with equation (4) is rewritten as

$$I(\varphi)=I_lG^2(\varphi) \tag{6}$$

where

$$G^2(\varphi)=\frac{\sin^2 N_0\dfrac{P\varphi}{2}}{\sin^2\dfrac{P\varphi}{2}}=\frac{\sin^2 \pi N_0P\zeta}{\sin^2\pi P\zeta}$$

$$I_l=\psi_l\psi_l^*=\left|\sum_{n=0}^{P-1}\varepsilon_n \exp(in\varphi)\right|^2=\left|\sum_{n=0}^{P-1}\varepsilon_n \exp(2\pi in\zeta)\right|^2$$
$$=\left|\sum_{n=0}^{P-1}\varepsilon_n \exp(inl\theta)\right|^2 \tag{7}$$

with

$$\theta=2\pi/P \quad \text{and} \quad N=PN_0 \tag{8}$$

and the suffix l in I_l comes from the fact that $I(\varphi)$ has sharp maxima at

$$\zeta=\frac{l}{P} \quad l\colon 0,\ \pm1,\ \pm2,\ \pm3,\ \ldots \tag{9}$$

because of the Laue function, $G^2(\varphi)$. Therefore, omitting the Laue function, we may call I_l the unitary intensity.

When $h+k$ is even, we have $\varepsilon_n=1$ and hence we get at once

$$I_l=\frac{\sin^2\left(\dfrac{P\varphi}{2}\right)}{\sin^2\left(\dfrac{\varphi}{2}\right)}\,.$$

Therefore

$$I(\varphi)=\frac{\sin^2\left(N\dfrac{\varphi}{2}\right)}{\sin^2\left(\dfrac{\varphi}{2}\right)} \tag{10}$$

from which we obtain

$$I_l=P^2\delta_{l,nP} \quad \text{and} \quad I(2n\pi)=P^2N_0^2=N^2 \tag{11}$$

where $\delta_{l,nP}$ is Kronecker's delta with $n=0,\ \pm1,\ \pm2,\ \ldots$. This equation gives the unitary intensities of the fundamental reflexions, and holds in general even if there is any disordering in the stacking of layers.

The intensities of superlattice reflexions can be calculated as shown below for some typical cases.

(i) $(M\,\bar{M}')$

The unitary intensity for the simplest structure of the complex out-of-step structures, which is obtained by $t=1$, $a_1=M$ and $b_1=M'$, *i.e.* $(M\bar{M}')$ with $P=M+M'$, is readily calculated, and we obtain

$$I_l=\left|\sum_{n=0}^{M-1}\exp(in\varphi)+\varepsilon\exp(iM\varphi)\sum_{n=0}^{M'-1}\exp(in\varphi)\right|^2$$
$$=[1-\varepsilon\cos^2(P\varphi/2)-(1-\varepsilon)\cos\{(M-M')\varphi/2\}$$
$$\times\cos(P\varphi/2)]/\sin^2(\varphi/2)\,. \tag{12}$$

Since we have $\varepsilon=-1$ for superlattice reflexions, equation (12) with equation (9) turns to

$$I_l=\frac{2}{\sin^2(\pi l/P)}\left\{1-(-1)^l\cos\frac{\pi l}{P}(M-M')\right\}.$$

Therefore

$$\begin{cases} I_l=\dfrac{4\sin^2\dfrac{\pi l}{2P}(M-M')}{\sin^2\dfrac{\pi l}{P}} & \text{for} \quad l\colon \text{even} \\[3ex] I_l=\dfrac{4\cos^2\dfrac{\pi l}{2P}(M-M')}{\sin^2\dfrac{\pi l}{P}} & \text{for} \quad l\colon \text{odd,} \end{cases} \tag{13}$$

from which we have

$$I_0=(M-M')^2. \tag{14}$$

(ii) $(M\,\overline{M+1})$

When $M'=M+1$ in case(i), we have $(M\,\overline{M+1})$ with

* Another form $(a_1\bar{b}_1a_2\bar{b}_2\ldots a_s\bar{b}_sa_{s+1}\bar{b}_{s+1})$ can be transformed into the above form by displacing $\bar{b}_{s+1}$, for example, $(4\bar{3}1\bar{2}|\bar{4}3\bar{1}2)\rightarrow(6\bar{3}1|\bar{6}3\bar{1})$.

† The case of a non-integral value of M, which was treated by Fujiwara (1957), will be treated in the forthcoming papers as part II and part III of this series.

$P=2M+1$ and hence from equations (13) and (14) we have

$$\begin{cases} I_l=\dfrac{1}{\cos^2\dfrac{\pi l}{2P}} & \text{for } l\text{: even} \\ I_l=\dfrac{1}{\sin^2\dfrac{\pi l}{2P}} & \text{for } l\text{: odd}\,. \end{cases} \tag{15}$$

(iii) $(M \mid \bar{M})$

When $M'=M$ in case (i), we have $(M \mid \bar{M})$ with $P=2M$ *i.e.* the simple *APD* structure, and equation (13) gives

$$\begin{cases} I_l=0 & \text{for } l\text{: even} \\ I_l=\dfrac{4}{\sin^2\dfrac{\pi l}{P}} & \text{for } l\text{: odd}\,. \end{cases} \tag{16}$$

(iv) $([M] \mid [\bar{M}])$

The unitary intensity in the case of the complex *APD* structure, $([M] \mid [\bar{M}])$ with $P=2M$, is given by

$$I_l=\left|\{1+\varepsilon \exp(iM\varphi)\}\sum_{n=0}^{M-1}\varepsilon_n \exp(in\varphi)\right|^2$$
$$=2(1+\varepsilon\cos M\varphi)I_l^* \tag{17}$$

with

$$I_l^*=\left|\sum_{n=0}^{M-1}\varepsilon_n \exp(in\varphi)\right|^2. \tag{18}$$

This form is different from I_l in equation (7) because in I_l the summation is carried out over n from 0 to $P-1$ while in I_l^* from 0 to $M-1$ *i.e.* over half a period. As a result, for the superlattice reflexions, we have

$$\begin{cases} I_l=0 & \text{for } l\text{: even} \\ I_l=4I_l^* & \text{for } l\text{: odd}\,. \end{cases} \tag{19}$$

The first relation in equation (19) gives the extinction rule in the case of the complex *APD* structure [see equation (41)]. Equation (19) includes equation (16).

4. The Patterson method

Because, as mentioned in the previous section, the unitary intensity of the fundamental reflexion is usually given by either equation (10) or equation (11), the information with respect to the layer sequence is included only in the superlattice reflexions. Therefore, in this section we are concerned only with the superlattice reflexions *i.e.* the case of $\varepsilon=-1$. In this case, equation (7) is rewritten as

$$I_l=\sum_{n=0}^{P-1}\varepsilon_n^2+\sum_{m=1}^{P-1}\left(\sum_{n=0}^{P-1-m}\varepsilon_n\varepsilon_{n+m}\right)\exp(-iml\theta)+\text{conj.}$$
$$=P+\sum_{m=1}^{P-1}(A_m-B_m)\exp(-iml\theta)+\text{conj.}\,,\quad \theta=\frac{2\pi}{P} \tag{20}$$

where conj. means the complex conjugate of the foregoing term. A_m and B_m are the numbers of pairs separated by m layers with the same and different signs respectively when the mth layer is limited within one period as shown in Fig. 3(*a*) and (*b*), where examples of $(3\bar{2}3\bar{2}1\bar{1})$ with $P=12$ are shown for the cases of $m=5$ and 7.

Putting

$$N_m^+=A_m+A_{P-m}(1-\delta_{m,0})\quad N_m^-=B_m+B_{P-m}$$
$$m\text{: } 0, 1, 2, \ldots, P-1$$
$$D_m=N_m^+-N_m^-=\sum_{n=0}^{P-1}\varepsilon_n\varepsilon_{n+m}\,, \tag{21}$$

we can rewrite equation (20) as

$$I_l=\sum_{m=0}^{P-1}D_m\cos ml\theta \quad\text{with}\quad \theta=\frac{2\pi}{P}\,. \tag{22}$$

N_m^+ and N_m^- in equation (21) are found to be respectively the numbers of pairs separated by m layers with the same and different signs, when the mth layer is allowed to go into the next period, as can be seen in Fig. 3(*c*).

From the definition of N_m^+ and N_m^- in equation (21), we have the relations

$$N_{P-m}^+=N_m^+\,,\quad N_{P-m}^-=N_m^-\,,\quad N_m^++N_m^-=P\,. \tag{23}$$

Therefore

$$D_m=2N_m^+-P=P-2N_m^-=D_{P-m}$$
$$(\text{therefore } D_0=P)\,. \tag{24}$$

In the case of complex *APD* structure, we have

$$\varepsilon_{n+M}=-\varepsilon_n \tag{25}$$

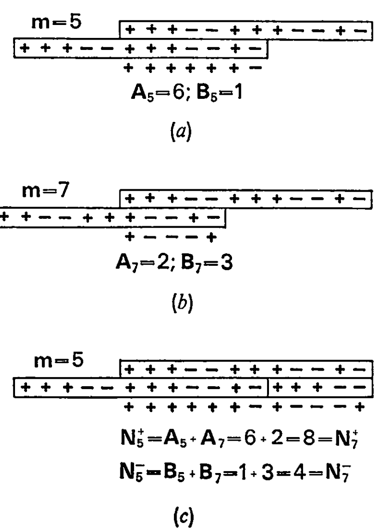


Fig. 3. A_m, B_m, N_m^+ and N_m^- with $m=5$ and 7 in the case of $(3\bar{2}3\bar{2}1\bar{1})$ with $P=12$.

and hence we get $D_{m+M}=-D_m$, which, with equation (24), gives the relation

$$D_{M-m}=D_{m+M}=-D_m\,.$$

Therefore

$$D_M=-D_0=-P \quad \text{and} \quad D_{\frac{M}{2}}=0 \quad (M:\text{even}). \quad (26)$$

From equation (22), we obtain at once two general relations

$$I_{P-l}=I_l \quad \text{and} \quad \sum_{l=0}^{P-1} I_l=PD_0=P^2. \quad (27)$$

The latter relation gives the normalization condition which is necessary when the observed intensities measured in an arbitrary unit are converted into the unitary intensities.

By the Fourier cosine transformation of equation (22), we obtain

$$C_m=\sum_{l=0}^{P-1} I_l \cos ml\theta=D_mP \quad (28)$$

in which C_0 gives the same relation as equation (27).

Since C_m is an integral multiple of P, we may determine the correct layer sequence even if the observed intensities are not so accurately measured, as will be seen in § 7.

The relation between C_m and the usual Patterson function

$$P(uvw)=\frac{1}{v_0}\sum_h\sum_k\sum_l I_{hkl}\cos 2\pi(hu+kv+lw)$$

is as follows:

Values of (uvw) are limited to the following two cases as

$$\left(0\,0\,\frac{m}{P}\right) \quad \text{and} \quad \left(\tfrac{1}{2}\,\tfrac{1}{2}\,\frac{m}{P}\right)$$

and

$$\begin{cases} v_0P\left(0\,0\,\frac{m}{P}\right)=\frac{1}{2P}(P^2+C_m)=N_m^+ \\ v_0P\left(\frac{1}{2}\,\frac{1}{2}\,\frac{m}{P}\right)=\frac{1}{2P}(P^2-C_m)=N_m^- \end{cases}$$

(see Appendix I), where v_0 is the volume of the unit cell with a height Pc.

5. The relation between C_m and the layer sequence symbol

In the case of the close-packed structures, one of us and others (Kakinoki, Kodera & Aikami, 1969) obtained useful relations between the letter sequences in Zhdanov symbols and a set of C_m and S_m, where C_m and S_m are respectively the Fourier cosine and sine transformations of the unitary intensity. Similarly in the present case, we can obtain useful relations between the layer sequence symbol and C_m. From equation (27), we have $S_m=0$. In order to derive the relations, it is useful to consider N_m^+ which is related to C_m and D_m by the relation

$$D_m=C_m/P=2N_m^+-P=D_{P-m} \quad (29)$$

[equations (24) and (28)].

If $\omega_{p_1\bar{n}_1\ldots p_g\bar{n}_g}$ is defined as the occurring frequency of a $2g$ letter sequence such as $p_1\bar{n}_1p_2\bar{n}_2\ldots p_g\bar{n}_g$ with $g\le t$ in the layer sequence symbol $(a_1\bar{b}_1a_2\bar{b}_2\ldots a_t\bar{b}_t)$, then we have from the definition the following relations:

$$\sum_{p=1}\omega_p=\sum_{n=1}\omega_{\bar{n}}=t\,,\quad \sum_{p=1}p\omega_p+\sum_{n=1}n\omega_{\bar{n}}=P \quad (30)$$

$$\left.\begin{array}{l} \sum_{p_1=1}\omega_{p_1\bar{n}_1\ldots p_g\bar{n}_g}=\omega_{\bar{n}_1p_2\bar{n}_2\ldots p_g\bar{n}_g}\,, \\ \sum_{n_g=1}\omega_{p_1\bar{n}_1\ldots p_g\bar{n}_g}=\omega_{p_1\bar{n}_1\ldots p_g}\,,\ \ldots\,, \\ \sum_{p=1}\omega_{p\bar{n}}=\omega_{\bar{n}}\,,\quad \sum_{n=1}\omega_{p\bar{n}}=\omega_p\,. \end{array}\right\} \quad (31)$$

Examples of the notation $\omega_{p_1\bar{n}_1\ldots p_g\bar{n}_g}$ are as follows: In a layer sequence symbol $(3\bar{1}2\bar{2}3\bar{1})$, they are

$$\omega_2=\omega_{\bar{2}}=1,\quad \omega_3=\omega_{\bar{1}}=2$$
$$\omega_{3\bar{1}}=2,\quad \omega_{\bar{1}2}=\omega_{2\bar{2}}=\omega_{\bar{2}3}=\omega_{\bar{1}3}=1$$
$$\omega_{3\bar{1}2}=\omega_{\bar{1}2\bar{2}}=\omega_{2\bar{2}3}=\omega_{\bar{2}3\bar{1}}=\omega_{3\bar{1}3}=\omega_{\bar{1}3\bar{1}}=1$$
$$\omega_{3\bar{1}2\bar{2}}=\omega_{\bar{1}2\bar{2}3}=\omega_{2\bar{2}3\bar{1}}=\omega_{\bar{2}3\bar{1}3}=\omega_{3\bar{1}3\bar{1}}=\omega_{\bar{1}3\bar{1}2}=1$$

and so on.

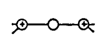
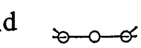
$m=1$ If we have a letter p in the layer sequence symbol, the number of pairs contributing to N_1^+ is $p-1$ and if we have a letter $\bar{n}$ in the symbol, the number is $n-1$, and N_1^+ can be calculated as

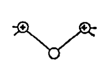
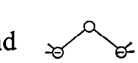
$$N_1^+=\sum_{p=1}(p-1)\omega_p+\sum_{n=1}(n-1)\omega_{\bar{n}}=P-2t$$

by the use of equation (30). As a result, we obtain from equation (29)

$$D_1=P-4t=C_1/P\,.$$

$m=2$ The routes contributing to N_2^+ are shown in Fig. 4, where the upper row indicates the positive layer and the lower one the negative layer. There are two sets of routes *i.e.*

the first set:  and 

the second set:  and  .

If we have two letters, p and $\bar{n}$, in the symbol, the number contributing to the first set of routes is $[(p-2)+(n-2)]$ so long as p, $n\ge 2$, and hence the total number contributing to the first set, $N_2^{+(1)}$, is calculated as

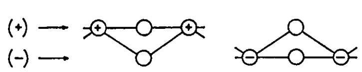


Fig. 4. Routes contributing to N_2^+.

$$N_2^{+(1)} = \sum_{p=2}(p-2)\omega_p + \sum_{n=2}(n-2)\omega_{\bar{n}}$$
$$= \sum_{p=1}(p-2)\omega_p + \omega_1 + \sum_{n=1}(n-2)\omega_{\bar{n}} + \omega_{\bar{1}}$$
$$= P - 4t + \{\Omega_1\}$$

by the use of equation (30), and $\{\Omega_1\}$ in the above equation is

$$\{\Omega_1\} = \omega_1 + \omega_{\bar{1}} \, .$$

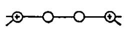
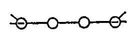
If 1 and $\bar{1}$ are in the symbol, we have the second set of routes, and by the use of equation (31) the number contributing to the second set, $N_2^{+(2)}$, is calculated as

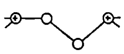
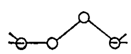
$$N_2^{+(2)} = \sum_{n_1=1}\sum_{n_2=1}\omega_{\bar{n}_1 1 \bar{n}_2} + \sum_{p_1=1}\sum_{p_2=1}\omega_{p_1\bar{1}p_2} = \omega_1 + \omega_{\bar{1}} = \{\Omega_1\} \, .$$

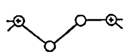
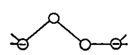
Finally, we have

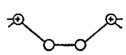
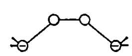
$$N_2^+ = N_2^{+(1)} + N_2^{+(2)} = P - 4t + 2\{\Omega_1\} \, ,$$
$$\therefore D_2 = P - 8t + 4\{\Omega_1\} \, .$$

$\boxed{m=3}$ The routes contributing to N_3^+ are shown in Fig. 5 and there are four sets of routes *i.e.*

the first set:  and 

the second set:  and 

the third set:  and 

the fourth set:  and 

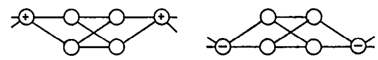


Fig. 5. Routes contributing to N_3^+.

By a similar consideration using equations (30) and (31), the number contributing to the ith set of routes, $N_3^{+(i)}$, is calculated as follows:

$$N_3^{+(1)} = \sum_{p=3}(p-3)\omega_p + \sum_{n=3}(n-3)\omega_{\bar{n}}$$
$$= \sum_{p=1}(p-3)\omega_p + \omega_2 + 2\omega_1$$
$$+ \sum_{n=1}(n-3)\omega_{\bar{n}} + \omega_{\bar{2}} + 2\omega_{\bar{1}}$$
$$= P - 6t + 2\{\Omega_1\} + (\omega_2 + \omega_{\bar{2}})$$
$$N_3^{+(2)} = \sum_{p=2}\omega_{p\bar{1}} + \sum_{n=2}\omega_{\bar{n}1}$$
$$= \sum_{p=1}\omega_{p\bar{1}} - \omega_{1\bar{1}} + \sum_{n=1}\omega_{\bar{n}1} - \omega_{\bar{1}1}$$
$$= \{\Omega_1\} - (\omega_{1\bar{1}} + \omega_{\bar{1}1})$$
$$N_3^{+(3)} = \sum_{p=2}\omega_{\bar{1}p} + \sum_{n=2}\omega_{1\bar{n}}$$
$$= \{\Omega_1\} - (\omega_{1\bar{1}} + \omega_{\bar{1}1})$$
$$N_3^{+(4)} = \omega_2 + \omega_{\bar{2}} \, .$$

Thus, we have

$$N_3^+ = \sum_{i=1}^{4} N_3^{+(i)} = P - 6t + 2(2\{\Omega_1\} + \{\Omega_2\}) \, .$$
$$\therefore D_3 = P - 12t + 4(2\{\Omega_1\} + \{\Omega_2\})$$

with

$$\{\Omega_2\} = \omega_2 + \omega_{\bar{2}} - (\omega_{1\bar{1}} + \omega_{\bar{1}1}) \, .$$

Table 1. *Lists of* N_m^+, D_m, N_m^{+*} *and* D_m^* *for some values of m*

	Complex out-of-step structure, $(a_1b_1a_2b_2 \ldots a_tb_t)$	Complex *APD* structure, $([M]\|[\bar{M}])$ with $[M]=(a_1b_1 \ldots a_sb_sa_{s+1})$
m	N_m^+	$N_m^{+*} = N_m^+/2,\ 2s+1=t$
0	P	M
1	$P-2t$	$M-1-2s$
2	$P-4t+2\{\Omega_1\}$	$M-2-4s+2\{\Omega_1^*\}$
3	$P-6t+4\{\Omega_1\}+2\{\Omega_2\}$	$M-3-6s+4\{\Omega_1^*\}+2\{\Omega_2^*\}$
4	$P-8t+6\{\Omega_1\}+4\{\Omega_2\}+2\{\Omega_3\}$	$M-4-8s+6\{\Omega_1^*\}+4\{\Omega_2^*\}+2\{\Omega_3^*\}$
	$N_m^+ = P-2mt+2\sum_{r=1}^{m-1}(m-r)\{\Omega_r\}$	$N_m^{+*} = M-m-2ms+2\sum_{r=1}^{m-1}(m-r)\{\Omega_r^*\}$
m	$D_m = C_m/P$	$D_m^* = C_m^*/M = D_m/2$
0	P	M
1	$P-4t$	$M-2-4s$
2	$P-8t+4\{\Omega_1\}$	$M-4-8s+4\{\Omega_1^*\}$
3	$P-12t+8\{\Omega_1\}+4\{\Omega_2\}$	$M-6-12s+8\{\Omega_1^*\}+4\{\Omega_2^*\}$
4	$P-16t+12\{\Omega_1\}+8\{\Omega_2\}+4\{\Omega_3\}$	$M-8-16s+12\{\Omega_1^*\}+8\{\Omega_2^*\}+4\{\Omega_3^*\}$
	$D_m = P-4mt+4\sum_{r=1}^{m-1}(m-r)\{\Omega_r\}$	$D_m^* = M-2m-4ms+4\sum_{r=1}^{m-1}(m-r)\{\Omega_r^*\}$

Continuing the similar process, we get the results listed in Table 1. Furthermore, we can show that the following relations hold in general:

$$N_m^+ = P - 2mt + 2\sum_{r=1}^{m-1}(m-r)\{\Omega_r\}$$

$$\text{with} \quad N_0^+ = P \quad \text{and} \quad N_1^+ = P - 2t \tag{32}$$

and hence

$$D_m = P - 4mt + 4\sum_{r=1}^{m-1}(m-r)\{\Omega_r\}$$

$$\text{with} \quad D_0 = P \quad \text{and} \quad D_1 = P - 4t \tag{33}$$

(see Appendix II), where $\{\Omega_r\}$ is the symbol implying, *e.g.*

$$\{\Omega_5\} = \{\Omega_5\}_{\text{odd}} - \{\Omega_5\}_{\text{even}} \tag{34}$$

with

$$\begin{aligned}\{\Omega_5\}_{\text{odd}} &= \{\Omega_5\}_1 + \{\Omega_5\}_3 + \{\Omega_5\}_5 \\ &= \omega_5 + \omega_5 \\ &+ \omega_{3\bar{1}1} + \omega_{1\bar{3}1} + \omega_{1\bar{1}3} + \omega_{2\bar{2}1} + \omega_{2\bar{1}2} + \omega_{1\bar{2}2} \\ &+ \omega_{\bar{3}1\bar{1}} + \omega_{\bar{1}3\bar{1}} + \omega_{\bar{1}1\bar{3}} + \omega_{\bar{2}2\bar{1}} + \omega_{\bar{2}1\bar{2}} + \omega_{\bar{1}2\bar{2}} \\ &+ \omega_{1\bar{1}1\bar{1}1} + \omega_{\bar{1}1\bar{1}1\bar{1}}\end{aligned} \tag{35}$$

$$\begin{aligned}\{\Omega_5\}_{\text{even}} &= \{\Omega_5\}_2 + \{\Omega_5\}_4 \\ &= \omega_{4\bar{1}} + \omega_{3\bar{2}} + \omega_{2\bar{3}} + \omega_{1\bar{4}} \\ &+ \omega_{\bar{4}1} + \omega_{\bar{3}2} + \omega_{\bar{2}3} + \omega_{\bar{1}4} \\ &+ \omega_{2\bar{1}1\bar{1}} + \omega_{1\bar{2}1\bar{1}} + \omega_{1\bar{1}2\bar{1}} + \omega_{1\bar{1}1\bar{2}} \\ &+ \omega_{\bar{2}1\bar{1}1} + \omega_{\bar{1}2\bar{1}1} + \omega_{\bar{1}1\bar{2}1} + \omega_{\bar{1}1\bar{1}2}\,.\end{aligned} \tag{36}$$

Namely, for example, $\{\Omega_r\}_{\text{odd}}$ is the sum of occurring frequencies of the types of $\omega_{p_1\bar{n}_1\ldots p_g\bar{n}_g p_{g+1}}$ and $\omega_{\bar{p}_1 n_1\ldots\bar{p}_g n_g\bar{p}_{g+1}}$ with

$$\sum_{i=1}^{g+1} p_i + \sum_{i=1}^{g} n_i = r\,,$$

in the layer sequence symbol, *i.e.* those of all combinations of the odd partition of a given r. Some examples of $\{\Omega_r\}$ are listed in Table 2.

From the general expressions of N_m^+ and D_m, equations (32) and (33), we can derive the following relations:

$$\begin{aligned}\{\Omega_r\} &= (N_{r-1}^+ - 2N_r^+ + N_{r+1}^+)/2 \\ &= (D_{r-1} - 2D_r + D_{r+1})/4 \\ &= (C_{r-1} - 2C_r + C_{r+1})/4P = \{\Omega_{P-r}\}\end{aligned} \tag{37}$$

$$t = (P - D_1)/4 = (P^2 - C_1)/4P\,. \tag{38}$$

Furthermore, from the general expression of D_m, we get

$$C_m = PD_m = P^2 - 4Pq_m \tag{39}$$

with

$$q_m = mt - \sum_{r=1}^{m-1}(m-r)\{\Omega_r\}\,. \tag{40}$$

Thus, C_m obtained from the observed intensities with the normalization condition given by equation (27) should be an integer which satisfies equation (39), *i.e.* any one of a set of integers which start from P^2 at intervals, $4P$. As a result, even if the observed intensities are not so accurately measured, we may obtain the correct layer sequence, as will be seen in § 7.

6. The case of the complex *APD* structure

If a period P is even, *i.e.* $P = 2M$ and if all I_l's with l even vanish, we obtain, by the use of equation (22), the relation

$$0 = \sum_{l'=0}^{M-1} I_{2l'} = \sum_{m=0}^{P-1} D_m \sum_{l'=0}^{M-1} \cos 2\pi\frac{ml'}{M} = M(D_0 + D_M)$$

and hence

$$D_M = -P \quad i.e. \quad N_M^- = P\,. \tag{41}$$

This equation indicates that the structure should be the complex *APD* structure, ($[M] \mid [\bar{M}]$) with $P = 2M$, in accordance with equation (19).

Table 2. *Lists of* $\{\Omega_r\}$ *and* $\{\Omega_r^*\}$ *for some values of* r

r	$\{\Omega_r\} = \{\Omega_{P-r}\}$	$\{\Omega_r^*\} = \{\Omega_r\}/2 = -\{\Omega_{M-r}^*\}$
(0)	$t = (P - D_1)/4$	$s = (M - 2 - D_1^*)/4$
1	$\omega_1 + \omega_{\bar{1}}$	
	$= (P - 2D_1 + D_2)/4$	$= (M - 2D_1^* + D_2^*)/4$
2	$\omega_2 + \omega_{\bar{2}} - (\omega_{1\bar{1}} + \omega_{\bar{1}1})$	
	$= (D_1 - 2D_2 + D_3)/4$	$= (D_1^* - 2D_2^* + D_3^*)/4$
3	$\omega_3 + \omega_{\bar{3}} + \omega_{1\bar{1}1} + \omega_{\bar{1}1\bar{1}}$	
	$-(\omega_{2\bar{1}} + \omega_{1\bar{2}} + \omega_{\bar{2}1} + \omega_{\bar{1}2})$	
	$= (D_2 - 2D_3 + D_4)/4$	$= (D_2^* - 2D_3^* + D_4^*)/4$
4	$\omega_4 + \omega_{\bar{4}} + \omega_{2\bar{1}1} + \omega_{1\bar{2}1} + \omega_{1\bar{1}2} + \omega_{\bar{2}1\bar{1}} + \omega_{\bar{1}2\bar{1}} + \omega_{\bar{1}1\bar{2}}$	
	$-(\omega_{3\bar{1}} + \omega_{2\bar{2}} + \omega_{1\bar{3}} + \omega_{\bar{3}1} + \omega_{\bar{2}2} + \omega_{\bar{1}3} + \omega_{1\bar{1}1\bar{1}} + \omega_{\bar{1}1\bar{1}1})$	
	$\{\Omega_r\} = (N_{r-1}^+ - 2N_r^+ + N_{r+1}^+)/2$	$\{\Omega_r^*\} = (N_{r-1}^{+*} - 2N_r^{+*} + N_{r+1}^{+*})/2$
	$= (D_{r-1} - 2D_r + D_{r+1})/4$	$= (D_{r-1}^* - 2D_r^* + D_{r+1}^*)/4$
	$= (C_{r-1} - 2C_r + C_{r+1})/4P$	$= (C_{r-1}^* - 2C_r^* + C_{r+1}^*)/4M$

In the case of complex *APD* structure, the unitary intensities of the superlattice reflexions are given by equation (19), *i.e.*

$$\begin{cases} I_l=0 & \text{for } l\text{: even} \\ I_l=4I_l^* & \text{for } l\text{: odd} \end{cases}$$

where

$$I_l^*=\left|\sum_{n=0}^{M-1}\varepsilon_n \exp(inl\theta)\right|^2 \quad \text{and} \quad \theta=\frac{\pi}{M} \tag{42}$$

with the normalization condition

$$\sum_{l\text{odd}=1}^{2M-1} I_l^*=M^2 \tag{43}$$

which is obtained from equation (27).

If the corresponding quantities to N_m^+, N_m^-, D_m and $\{\Omega_r\}$ are counted not over P but over M and denoted respectively by N_m^{+*}, N_m^{-*}, D_m^* and $\{\Omega_r^*\}$, then we obtain from their definitions the following relations:

$$N_m^+ \;=2N_m^{+*}, \quad N_m^-=2N_m^{-*},$$

and therefore,

$$D_m \;=2(N_m^{+*}-N_m^{-*})=2D_m^* \quad \text{with} \quad D_0^*=M;$$

$$N_{m+M}^{+*}=N_m^{-*}, \quad N_{m+M}^{-*}=N_m^{+*}$$

and therefore,

$$D_{m+M}^*=-D_m^*;$$

$$N_{M-m}^{+*}=N_m^{-*}, \quad N_{M-m}^{-*}=N_m^{+*}$$

and therefore,

$$D_{M-m}^*=-D_m^*;$$

$$N_m^{+*}+N_m^{-*}=M \quad \therefore \quad D_m^*=2N_m^{+*}-M=M-2N_m^{-*} \tag{44}$$

$\{\Omega_r\}=2\{\Omega_r^*\}$ (see Appendix III).

Using these relations, we can derive from the corresponding equations in the previous section the following relations:

$$I_l^*=\sum_{m=0}^{M-1} D_m^* \cos ml\theta \quad \text{for} \quad l \text{ odd with } \theta=\frac{\pi}{M} \tag{45}$$

$$C_m^*=\sum_{l\text{odd}=1}^{P-1} I_l^* \cos ml\theta=MD_m^* \tag{46}$$

$$N_m^{+*}=M-m-2ms+2\sum_{r=1}^{m-1}(m-r)\{\Omega_r^*\}$$
$$\text{with} \quad N_0^{+*}=M \quad \text{and} \quad N_1^{+*}=M-1-2s \tag{47}$$

$$D_m^*=M-2m-4ms+4\sum_{r=1}^{m-1}(m-r)\{\Omega_r^*\}$$
$$\text{with} \quad D_0^*=M \quad \text{and} \quad D_1^*=M-2-4s \tag{48}$$

$$\begin{aligned}\{\Omega_r^*\}&=(N_{r-1}^{+*}-2N_r^{+*}+N_{r+1}^{+*})/2\\ &=(D_{r-1}^*-2D_r^*+D_{r+1}^*)/4\\ &=(C_{r-1}^*-2C_r^*+C_{r+1}^*)/4M=-\{\Omega_{M-r}^*\}\end{aligned} \tag{49}$$

$$s=(M-2-D_1^*)/4=(M^2-2M-C_1^*)/4M\,. \tag{50}$$

Some examples are listed in Tables 1 and 2.

7. Examples

Ideal cases

There are 21 independent structures in the case of the complex *APD* structure with $M=9$ *i.e.* ([9] | [$\bar{9}$]). They are listed in Table 3 together with D_m^* obtained by the way shown in Fig. 3(*c*) and I_l^* calculated from equation (45).

Table 3. 21 *independent structures of* ([M] | [$\bar{M}$]) *with* $M=9$ *and the values of* D_m^* *and* I_l^*

s	[9]	D_1^*	D_2^*	D_3^*	D_4^*	I_1^*	I_3^*	I_5^*	I_7^*	I_9^*
0	(9)	7	5	3	1	33·163	4	1·704	1·133	1
	$(7\bar{1}1)$	3	5	3	1	25·645	0	3·094	7·261	9
	$(6\bar{2}1)$	3	1	3	1	19·517	4	10·612	5·871	1
1	$(5\bar{3}1)$	3	1	−1	1	15·517	12	6·612	1·871	9
	$(5\bar{2}2)$	3	−3	−1	1	9·389	16	14·130	0·481	1
	$(4\bar{4}1)$	3	1	−1	−3	14·127	16	0·484	9·389	1
	$(4\bar{3}2)$	3	−3	−5	−3	3·999	28	4·002	3·999	1
	$(5\bar{1}1\bar{1}1)$	−1	5	−1	1	14·127	4	0·484	9·389	25
	$(4\bar{2}1\bar{1}1)$	−1	1	−1	−3	6·609	12	1·874	15·517	9
	$(4\bar{1}2\bar{1}1)$	−1	1	3	−3	10·609	4	5·874	19·517	1
	$(3\bar{3}1\bar{1}1)$	−1	1	−5	1	3·999	16	4·002	3·999	25
2	$(3\bar{1}3\bar{1}1)$	−1	1	−1	5	9·389	4	14·130	0·481	25
	$(3\bar{2}2\bar{1}1)$	−1	−3	−1	1	1·871	12	15·520	6·609	9
	$(3\bar{2}1\bar{2}1)$	−1	−3	3	1	5·871	4	19·520	10·609	1
	$(3\bar{2}1\bar{1}2)$	−1	−3	−1	−3	0·481	16	9·392	14·127	1
	$(3\bar{1}2\bar{2}1)$	−1	−3	3	5	7·261	0	25·648	3·091	9
	$(2\bar{2}2\bar{2}1)$	−1	−7	3	5	1·133	4	33·166	1·701	1
	$(3\bar{1}1\bar{1}1\bar{1}1)$	−5	5	−5	5	3·999	4	4·002	3·999	49
3	$(2\bar{2}1\bar{1}1\bar{1}1)$	−5	1	−1	1	0·481	4	9·392	14·127	25
	$(2\bar{1}2\bar{1}1\bar{1}1)$	−5	1	3	−3	3·091	0	7·264	25·645	9
	$(2\bar{1}1\bar{2}1\bar{1}1)$	−5	1	3	−7	1·701	4	1·136	33·163	1

Some examples of the Patterson method in the ideal cases are shown below:

$[9]=(9)$ with $(7, 5, 3, 1)$ Here, $(7, 5, 3, 1)$ shows the values of $(D_1^*, D_2^*, D_3^*, D_4^*)$ obtained from Table 3. From equation (50), we have $s=(9-2-7)/4=0$ and hence we have at once $[9]=(9)$.

$[9]=(7\bar{1}1)$ with $(3, 5, 3, 1)$ From equation (50), we have $s=(7-3)/4=1$ and hence we have $[9]=(a_1\bar{b}_1a_2)$ with $a_1+b_1+a_2=9$. From equation (49), we have $\omega_1+\omega_{\bar{1}}=(9-6+5)/4=2$ and hence the third number should be 7 or -7. However, we can assume without loss of generality that the first number a_1 is the greatest of all and we have at once $(7\bar{1}1)$.

$[9]=(4\bar{4}1)$ with $(3, 1, -1, -3)$ From equation (50), we have $s=(7-3)/4=1$ and hence we have $[9]=(a_1\bar{b}_1a_2)$ with $a_1+b_1+a_2=9$. From equation (49), we have $\omega_1+\omega_{\bar{1}}=(9-6+1)/4=1$ and hence we can put $a_2=1$ without loss of generality, and we obtain $[9]=(a_1\bar{b}_11)$ with $a_1+b_1=8$ and $a_1, b_1\geq 2$. From equation (49) or Table 2, we have $\{\Omega_2^*\}=\omega_2+\omega_{\bar{2}}-(\omega_{1\bar{1}}+\omega_{\bar{1}1})=(3-2-1)/4=0$. Since $\{\Omega_1^*\}=1$, we have $\omega_{1\bar{1}}+\omega_{\bar{1}1}=0$ and hence $\omega_2+\omega_{\bar{2}}=0$ and $a_1, b_1\geq 3$. As a result, there are two choices $[9]=(5\bar{3}1)$ and $(4\bar{4}1)$. From Table 2, we have $\{\Omega_3^*\}=(1+2-3)/4=0$ and hence the case $(5\bar{3}1)$ can be excluded and we obtain $(4\bar{4}1)$.

$[9]=(2\bar{1}1\bar{2}1\bar{1}1)$ with $(-5, 1, 3, -7)$ From equation (50), we have $s=(7+5)/4=3$ and hence we have $[9]=(a_1\bar{b}_1a_2\bar{b}_2a_3\bar{b}_3a_4)$ with $a_1+b_1+a_2+b_2+a_3+b_3+a_4=9$. From Table 2, we have $\omega_1+\omega_{\bar{1}}=(9+10+1)/4=5$ and hence we have three choices such as $[9]=(2\bar{2}1\bar{1}1\bar{1}1)$, $(2\bar{1}2\bar{1}1\bar{1}1)$ and $(2\bar{1}1\bar{2}1\bar{1}1)$. From Table 2, we have $\omega_2+\omega_{\bar{2}}-(\omega_{1\bar{1}}+\omega_{\bar{1}1})=(-5-2+3)/4=-1$. Since we have $\omega_2+\omega_{\bar{2}}=2$, we get $\omega_{1\bar{1}}+\omega_{\bar{1}1}=3$ from which we can exclude the first choice. From Table 2, we have $\{\Omega_3^*\}=\omega_3+\omega_{\bar{3}}+\omega_{1\bar{1}1}+\omega_{\bar{1}1\bar{1}}-(\omega_{2\bar{1}}+\omega_{1\bar{2}}+\omega_{\bar{2}1}+\omega_{\bar{1}2})=(1-6-7)/4=-3$. This value is compatible only with the third choice *i.e.* $[9]=(2\bar{1}1\bar{2}1\bar{1}1)$.

Real case

It was reported by one of us (Kakinoki, 1962) that the use of the unitary intensity, I_l^*, was useful in determining the layer sequence of a complex *APD* structure of $([9]\,|\,[\bar{9}])$ with $[9]=(2\bar{2}2\bar{2}1)$ and $P=18$, which had been adopted by Fujiwara (1957) as a starting structure in the analysis of the structure with $M=1\cdot 8$, and that the correct structure was obtained only by assuming the relations $I_5^*>4I_3^*$ and $I_3^*>I_1^*, I_7^*, I_9^*$.

Table 4. *Unitary intensities, I_l^*, for the structure* $([9]\,|\,[\bar{9}])$ *with* $[9]=(2\bar{2}2\bar{2}1)$ *and* $P=18$

l	Calculated	A rough estimation	The roughest estimation
1, 17	1·1		0
3, 15	4·0	$>I_1^*, I_7^*, I_9^*$	0
5, 13	33·2	$>4I_3^*$	40·5
7, 11	1·7		0
9	1·0		0

It is shown below that by the use of D_m^* the correct structure can be obtained even by a rough estimation of the observed intensities as given in the last column in Table 4. By the use of these values, D_m^*'s in equation (46) are calculated as listed in Table 5 together with the correct values of them. By the use of $D_m^*=-1\cdot 57$ and equation (50) with $M=9$, s is calculated as $s=(M-2-D_1^*)/4=2\cdot 14$. Therefore, we may put $s=2$ and the sequence should be

$$[9]=(a_1\bar{b}_1a_2\bar{b}_2a_3)$$

with

$$a_1+b_1+a_2+b_2+a_3=9.$$

Table 5. *Values of* D_m^*

m	D_m^* calculated from the roughest estimation of intensities	Correct values
0	9	9
1	$-1\cdot 57$	-1
2	$-8\cdot 46$	-7
3	4·50	3
4	6·89	5

By the use of $D_0^*=9$, $D_1^*=-1\cdot 57$, $D_2^*=-8\cdot 46$ and Table 2, $\{\Omega_1^*\}$ is calculated as

$$\{\Omega_1^*\}=\omega_1+\omega_{\bar{1}}=(D_0^*-2D_1^*+D_2^*)/4=0\cdot 92$$

where we may put $\omega_1=1$ and, as a result, the sum of the remaining four letters is 8 and each of them is larger than or equal to 2. Hence we have $\omega_2+\omega_{\bar{2}}=4$ and finally, we get the correct layer sequence $[9]=(2\bar{2}2\bar{2}1)$.

Thus, even with such a roughest estimation of intensities as shown in the last column of Table 4, we can obtain the correct layer sequence by the use of Patterson method.

Generally speaking, the value of D_m obtained from the observed intensities is not an integer because of experimental errors. If, however, we use two successive integral values between which D_m lies, the incorrect one will be excluded in the course of calculating $\{\Omega_r\}$ as r increases, or, at least, we can limit the number of models to be examined.

APPENDIX I

The relation between C_m and the usual Patterson function

The usual Patterson function is defined as

$$P(uvw)=\frac{1}{v_0}\sum_h\sum_k\sum_l I_{hkl}\cos 2\pi(hu+kv+lw) \quad \text{(A1)}$$

where v_0 is the volume of the unit cell with a height Pc. In order to get the relation between C_m defined by equation (28) and $P(uvw)$ defined above, we have only to substitute the unitary intensity, I_l, defined by equation (7) into I_{hkl} in the above equation. From the properties of the unitary intensity which are given by equations (7), (11) and (22), I_{hkl} has such properties as

$$\begin{cases} I_{hkl+nP}=I_{hkl} \quad l=0,1,2,\ldots,P-1 \\ \qquad n=0,\pm1,\pm2,\ldots & \text{(A2)} \\ I_{hkl} \quad =P^2\delta_{l,nP} \quad \text{for} \quad h+k=2g & \text{(A3)} \\ I_{hkl} \quad =I_l \text{ in equation (22)} \\ \qquad \text{for } h+k=2g+1 & \text{(A4)} \end{cases}$$

where g is an integer including 0.

Using equation (A2), we can calculate equation (A1) as follows:

$$v_0P(uvw)=\sum_h\sum_k\sum_{l=0}^{P-1}I_{hkl}$$
$$\times\sum_{n=-\infty}^{\infty}\cos 2\pi\{hu+kv+(l+nP)w\}$$
$$=\sum_{l=0}^{P-1}\sum_h\sum_k I_{hkl}\cos 2\pi(hu+kv+lw)$$
$$\times\sum_{m=-\infty}^{\infty}\delta(Pw-m) \qquad \text{(A5)}$$

by the use of the well-known relations

$$\begin{cases}\sum_{n=-\infty}^{\infty}\cos 2\pi nax=\frac{1}{|a|}\sum_{m=-\infty}^{\infty}\delta\left(x-\frac{m}{a}\right) \\ \sum_{n=-\infty}^{\infty}\sin 2\pi nax=0 & \text{(A6)}\end{cases}$$

where $\delta(x)$ is Dirac's delta function. Since we have $\delta(Pw-m)$ in equation (A5), w may be limited to m/P and hence we rewrite equation (A5) as

$$v_0P\left(uv\frac{m}{P}\right)=\frac{1}{P}\sum_{l=0}^{P-1}\sum_h\sum_k I_{hkl}\cos 2\pi\left(hu+kv+\frac{ml}{P}\right) \qquad \text{(A7)}$$

where we can limit the values of m as

$$m=0,1,2,\ldots,P\text{–}1$$

since we may put $0\le w<1$.

From equations (A3) and (A4), the calculation of equation (A7) results in

$$v_0P\left(uv\frac{m}{P}\right)=\frac{1}{P}\sum_{l=0}^{P-1}\sum_{h=-\infty}^{\infty}\sum_{g=-\infty}^{\infty}\left[P^2\delta_{l,nP}\right.$$
$$\times\cos 2\pi\left\{hu+(2g-h)v+\frac{ml}{P}\right\}$$
$$\left.+I_l\cos 2\pi\left\{hu+(2g+1-h)v+\frac{ml}{P}\right\}\right]$$
$$=\frac{1}{P}\left[P^2\sum_{h=-\infty}^{\infty}\cos 2\pi h(u-v)\right.$$
$$\left.+\sum_{l=0}^{P-1}I_l\sum_{h=-\infty}^{\infty}\cos 2\pi\left\{h(u-v)+v+\frac{ml}{P}\right\}\right]$$
$$\times\sum_{p=-\infty}^{\infty}\delta(2v-p) \qquad \text{(A8)}$$

by the use of equation (A6). Since we have $\delta(2v-p)$ in equation (A8), v can be limited to $p/2$ and hence we can rewrite equation (A8) as

$$v_0P\left(u\frac{p}{2}\frac{m}{P}\right)=\frac{1}{2P}\left[P^2\sum_{h=-\infty}^{\infty}\cos 2\pi h\left(u-\frac{p}{2}\right)\right.$$
$$\left.+\sum_{l=0}^{P-1}I_l\sum_{h=-\infty}^{\infty}\cos 2\pi\left\{h\left(u-\frac{p}{2}\right)+\frac{p}{2}+\frac{ml}{P}\right\}\right] \qquad \text{(A9)}$$

where we can limit the values of p as $p=0$ and 1 since we may put $0\le v<1$.

For $p=0$, equation (A9) is calculated as

$$v_0P\left(u0\frac{m}{P}\right)=\frac{1}{2P}\left\{P^2\sum_{h=-\infty}^{\infty}\cos 2\pi hu\right.$$
$$\left.+\sum_{l=0}^{P-1}I_l\sum_{h=-\infty}^{\infty}\cos 2\pi\left(hu+\frac{ml}{P}\right)\right\}$$
$$=\frac{1}{2P}\left(P^2+\sum_{l=0}^{P-1}I_l\cos 2\pi\frac{ml}{P}\right)\sum_{q=-\infty}^{\infty}\delta(u-q)\,. \qquad \text{(A10)}$$

Since we may put $0\le u<1$, we have $q=0$ and therefore, $u=0$. Using C_m defined by equation (28), and equation (24), we can rewrite equation (A10) as

$$v_0P\left(0\,0\frac{m}{P}\right)=\frac{1}{2P}(P^2+C_m)=N_m^+. \qquad \text{(A11)}$$

Similarly, for $p=1$ we obtain

$$v_0P\left(\tfrac{1}{2}\,\tfrac{1}{2}\frac{m}{P}\right)=\frac{1}{2P}(P^2-C_m)=N_m^-\,. \qquad \text{(A12)}$$

APPENDIX II

The derivation of the general forms of N_m^+ and D_m

From equation (30), we have

$$\sum_{p=1}\omega_p=\sum_{n=1}\omega_{\bar{n}}=t\,,\quad \sum_{p=1}p\omega_p+\sum_{n=1}n\omega_{\bar{n}}=P\,. \qquad \text{(A13)}$$

In order to derive the general form of N_m^+, it is convenient to divide the forms of routes contributing to N_m^+ into two parts; one is the linear form as shown in Fig. 6 and the other the zigzag form as shown in Fig. 7.

Linear form If we have two letters p and $\bar{n}$ in the layer sequence symbol

$$(a_1\bar{b}_1a_2\bar{b}_2\ldots a_t\bar{b}_t)\,,$$

then the number of pairs contributing to N_m^+ is given by $(p-m)+(n-m)$ so long as $p,n\ge m$. Therefore, the

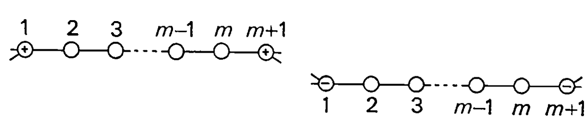


Fig. 6. The linear form of routes contributing to N_m^+.

total number of pairs contributing to N_m^+ from the linear form, $N_m^{+(l)}$, is calculated as follows:

$$N_m^{+(l)}=\sum_{p=m}(p-m)\omega_p+\sum_{n=m}(n-m)\omega_{\bar{n}}$$

$$=\sum_{p=1}(p-m)\omega_p-\sum_{p=1}^{m-1}(p-m)\omega_p$$

$$+\sum_{n=1}(n-m)\omega_{\bar{n}}-\sum_{n=1}^{m-1}(n-m)\omega_{\bar{n}}$$

and hence, by the use of equation (A 13), we get

$$N_m^{+(l)}=P-2mt+\sum_{r=1}^{m-1}(m-r)(\omega_r+\omega_{\bar{r}})\,. \quad \text{(A14)}$$

Zigzag form For simplicity, we consider here only the zigzag form for which both ends are positive. This form, as can be seen in Fig. 7, consists of three parts; the first part is the first u positive layers, the second the middle S layers and the third the last v positive layers. Thus, the zigzag form is expressed in a general form $(u\bar{n}_1p_1\bar{n}_2p_2\ldots\bar{n}_{v-1}p_{v-1}\bar{n}_vv)$ with

$$\sum_{i=1}^{v-1}p_i+\sum_{i=1}^{v}n_i=S \quad\text{and}\quad u+S+v=m+1\,.$$

Therefore, if in the layer sequence symbol we have a $2v+1$ letter sequence such as $(p_0S_{2v-1}^{(--)}p_v)$ with $(S_{2v-1}^{(--)})=(\bar{n}_1p_1\bar{n}_2p_2\ldots\bar{n}_{v-1}p_{v-1}\bar{n}_v)$, $p_0\geq u$ and $p_v\geq v$, then it contributes to $N_m^{+(z+)}$; here the superscript $(z+)$ means the zigzag form for which both ends are positive. $S_{2v-1}^{(--)}$ represents one configuration of $2v-1$ letter sequence for which both ends are negative and with fixed values of S and v.

Therefore, $N_m^{+(z+)}$ is given by

$$N_m^{+(z+)}=\sum_{S=1}^{m-1}\sum_{v}\sum_{S_{2v-1}^{(--)}}N_m^+(S_{2v-1}^{(--)}) \quad \text{(A15)}$$

with

$$N_m^+(S_{2v-1}^{(--)})=\sum_{u=1}^{m-S}\sum_{p_0=u}\sum_{p_v=v}\omega_{p_0S_{2v-1}^{(--)}p_v} \quad \text{(A16)}$$

where the summation over $S_{2v-1}^{(--)}$ means that the summation is carried out over all possible configurations of $2v-1$ letters when S and v are fixed. The calculation of equation (A16) is performed, by the use of equation (31), as follows:

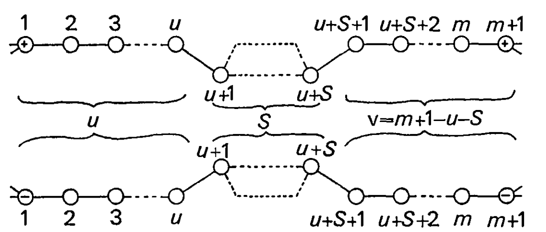


Fig. 7. The zigzag form of routes contributing to N_m^+.

$$N_m^+(S_{2v-1}^{(--)})=\sum_{u=1}^{m-S}\sum_{p_0=u}\sum_{p_v=v=m+1-S-u}\omega_{p_0S_{2v-1}^{(--)}p_v}$$

$$=\sum_{u=1}^{m-S}\sum_{p_0=u}\Big\{\sum_{p_v=1}\omega_{p_0S_{2v-1}^{(--)}p_v}-\sum_{p_v=1}^{m-S-u}\omega_{p_0S_{2v-1}^{(--)}p_v}$$

$$\times(1-\delta_{u,m-S})(1-\delta_{S,m-1})\Big\}=\sum_{u=1}^{m-S}\sum_{p_0=u}\omega_{p_0S_{2v-1}^{(--)}}$$

$$-\sum_{u=1}^{m-S-1}\sum_{p_0=u}\sum_{p_v=1}^{m-S-u}\omega_{p_0S_{2v-1}^{(--)}p_v}(1-\delta_{S,m-1})$$

$$=\sum_{u=1}^{m-S}\Big\{\sum_{p_0=1}\omega_{p_0S_{2v-1}^{(--)}}$$

$$-\sum_{p_0=1}^{u-1}\omega_{p_0S_{2v-1}^{(--)}}(1-\delta_{u,1})(1-\delta_{S,m-1})\Big\}$$

$$-\sum_{u=1}^{m-S-1}\sum_{p_v=1}^{m-S-u}\Big\{\sum_{p_0=1}\omega_{p_0S_{2v-1}^{(--)}p_v}$$

$$-\sum_{p_0=1}^{u-1}\omega_{p_0S_{2v-1}^{(--)}p_v}(1-\delta_{u,1})(1-\delta_{S,m-2})\Big\}(1-\delta_{S,m-1})$$

$$=\sum_{u=1}^{m-S}\omega_{S_{2v-1}^{(--)}}-\Big\{\sum_{u=2}^{m-S}\sum_{p_0=1}^{u-1}\omega_{p_0S_{2v-1}^{(--)}}$$

$$+\sum_{u=1}^{m-S-1}\sum_{p_v=1}^{m-S-u}\omega_{S_{2v-1}^{(--)}p_v}\Big\}(1-\delta_{S,m-1})$$

$$+\sum_{u=2}^{m-S-1}\sum_{p_0=1}^{u-1}\sum_{p_v=1}^{m-S-u}\omega_{p_0S_{2v-1}^{(--)}p_v}$$

$$\times(1-\delta_{S,m-2})(1-\delta_{S,m-1})\,.$$

Finally, changing the order of summations, we have

$$N_m^+(S_{2v-1}^{(--)})=(m-S)\omega_{S_{2v-1}^{(--)}}$$

$$-\sum_{p=1}^{m-S-1}(m-S-p)\{\omega_{pS_{2v-1}^{(--)}}+\omega_{S_{2v-1}^{(--)}p}\}(1-\delta_{S,m-1})$$

$$+\sum_{p_0=1}^{m-S-2}\sum_{p_v=1}^{m-S-p_0-1}(m-S-p_0-p_v)\omega_{p_0S_{2v-1}^{(--)}p_v}$$

$$\times(1-\delta_{S,m-1})(1-\delta_{S,m-2})\,. \quad \text{(A17)}$$

Similarly, corresponding to equations (A15) and (A17) we obtain

$$N_m^{+(z-)}=\sum_{S=1}^{m-1}\sum_{v}\sum_{S_{2v-1}^{(++)}}N_m^+(S_{2v-1}^{(++)}) \quad \text{(A18)}$$

with

$$N_m^+(S_{2v-1}^{(++)})=(m-S)\omega_{S_{2v-1}^{(++)}}$$

$$-\sum_{n=1}^{m-S-1}(m-S-n)\{\omega_{\bar{n}S_{2v-1}^{(++)}}+\omega_{S_{2v-1}^{(++)}\bar{n}}\}(1-\delta_{S,m-1})$$

$$+\sum_{n_0=1}^{m-S-2}\sum_{n_v=1}^{m-S-n_0-1}(m-S-n_0-n_v)\omega_{\bar{n}_0S_{2v-1}^{(++)}\bar{n}_v}$$

$$\times(1-\delta_{S,m-1})(1-\delta_{S,m-2})\,. \quad \text{(A19)}$$

As a result, N_m^+ is expressed as

$$\begin{aligned} N_m^+ &= N_m^{+(l)} + N_m^{+(z+)} + N_m^{+(z-)} \\ &= P - 2mt + N_m^{+(1)} - N_m^{+(2)} + N_m^{+(3)} \end{aligned} \tag{A20}$$

with

$$\begin{aligned} N_m^{+(1)} &= \sum_{r=1}^{m-1} (m-r)(\omega_r + \omega_{\bar r}) \\ &+ \sum_{S=1}^{m-1} (m-S) \left[\sum_v \left\{ \sum_{S_{2v-1}^{(--)}} \omega_{S_{2v-1}^{(--)}} + \sum_{S_{2v-1}^{(++)}} \omega_{S_{2v-1}^{(++)}} \right\} \right] \end{aligned} \tag{A21}$$

$$\begin{aligned} N_m^{+(2)} &= \sum_{S=1}^{m-2} \sum_{p=1}^{m-S-1} (m-S-p) \\ &\times \sum_v \left[\sum_{S_{2v-1}^{(--)}} \{ \omega_{pS_{2v-1}^{(--)}} + \omega_{S_{2v-1}^{(--)}p} \} \right. \\ &+ \left. \sum_{S_{2v-1}^{(++)}} \{ \omega_{\bar p S_{2v-1}^{(++)}} + \omega_{S_{2v-1}^{(++)}\bar p} \} \right] \end{aligned} \tag{A22}$$

$$\begin{aligned} N_m^{+(3)} &= \sum_{S=1}^{m-3} \sum_{p_0=1}^{m-S-2} \sum_{p_v=1}^{m-S-p_0-1} (m-S-p_0-p_v) \\ &\times \left[\sum_v \left\{ \sum_{S_{2v-1}^{(--)}} \omega_{p_0 S_{2v-1}^{(--)} p_v} + \sum_{S_{2v-1}^{(++)}} \omega_{\bar p_0 S_{2v-1}^{(++)} \bar p_v} \right\} \right]. \end{aligned} \tag{A23}$$

The summations with respect to v, $S_{2v-1}^{(--)}$ and $S_{2v-1}^{(++)}$ in the square brackets in equation (A21) mean taking all configurations of odd number of letters whose sum is fixed to S. Hence similarly to equation (35), this term may be expressed as $\{\Omega_S\}_{\text{odd}}$ and $(\omega_r + \omega_{\bar r})$ may be expressed as $\{\Omega_r\}_1$. Using these notations, we can rewrite equation (A21) as

$$N_m^{+(1)} = \sum_{r=1}^{m-1} (m-r)[\{\Omega_r\}_1 + \{\Omega_r\}_{\text{odd}}]. \tag{A24}$$

With respect to $N_m^{+(2)}$ and $N_m^{+(3)}$, the variables in summations are changed so that $p+S$ in $N_m^{+(2)}$ and $p_0 + S + p_v$ in $N_m^{+(3)}$ may be equal to r, as follows:

$$\begin{aligned} N_m^{+(2)} &= \sum_{r=2}^{m-1} (m-r) \left[\sum_{S=1}^{r-1} \sum_v \left\{ \sum_{S_{2v-1}^{(--)}} (\omega_{r-sS_{2v-1}^{(--)}} \right. \right. \\ &+ \left. \left. \omega_{S_{2v-1}^{(--)}r-s}) + \sum_{S_{2v-1}^{(++)}} (\omega_{\overline{r-s}S_{2v-1}^{(++)}} + \omega_{S_{2v-1}^{(++)}\overline{r-s}}) \right\} \right] \end{aligned} \tag{A25}$$

and

$$\begin{aligned} N_m^{+(3)} &= \sum_{r=3}^{m-1} (m-r) \sum_{S=1}^{r-2} \sum_{p_0=1}^{r-S-1} \sum_v \left\{ \sum_{S_{2v-1}^{(--)}} \omega_{p_0 S_{2v-1}^{(--)} r-S-p_0} \right. \\ &+ \left. \sum_{S_{2v-1}^{(++)}} \omega_{\bar p_0 S_{2v-1}^{(++)} \overline{r-S-p_0}} \right\}. \end{aligned} \tag{A26}$$

Similarly to the case of $N_m^{+(1)}$, the summations with respect to S, v, $S_{2v-1}^{(--)}$ and $S_{2v-1}^{(++)}$ in the square brackets in equation (A25) mean to take all configurations of even number of letters whose sum is fixed to r and hence equation (A25) can be rewritten as

$$\begin{aligned} N_m^{+(2)} &= 2 \sum_{r=2}^{m-1} (m-r) \{\Omega_r\}_{\text{even}} \\ &= 2 \sum_{r=1}^{m-1} (m-r) \{\Omega_r\}_{\text{even}}. \end{aligned} \tag{A27}$$

Using a similar notation, we can rewrite equation (A26) as

$$\begin{aligned} N_m^{+(3)} &= \sum_{r=3}^{m-1} (m-r) \{\Omega_r\}_{\text{odd} \geq 3} \\ &= \sum_{r=1}^{m-1} (m-r) \{\Omega_r\}_{\text{odd}} - \sum_{r=1}^{m-1} (m-r) \{\Omega_r\}_1. \end{aligned} \tag{A28}$$

Finally, substitution of equations (A24), (A27) and (A28) into equation (A20) gives

$$N_m^+ = P - 2mt + 2 \sum_{r=1}^{m-1} (m-r) \{\Omega_r\} \tag{A29}$$

because of equation (34). As a result, we have

$$D_m = 2N_m^+ - P = P - 4mt + 4 \sum_{r=1}^{m-1} (m-r) \{\Omega_r\}. \tag{A30}$$

APPENDIX III

Verification of the relation $\{\Omega_r\} = 2\{\Omega_r^*\}$

Let a letter sequence be (S) and let its occurring frequency in the symbol $([M] \mid [\bar M])$ be $\omega_{(S)}$ of which $\omega_{(S)}^{(1)}$ are found in the first $[M]$ and $\omega_{(S)}^{(2)}$ in the last $[\bar M]$. For example, in a layer sequence symbol such as $([10] \mid [\overline{10}]) = ((\underline{2\bar 1}\underline{2\bar 1}\ 1\bar 2 1) \mid (\bar 2 1 \bar 2 1 \bar 1 \underline{2 \bar 1}))$ with $P = 20$, there are three $(S) = 2\bar 1$, two in the first [10] and one in the last $[\overline{10}]$, as shown by the underlining. Thus, we have

$$\omega_{2\bar 1} = 3 \quad \text{with} \quad \omega_{2\bar 1}^{(1)} = 2 \quad \text{and} \quad \omega_{2\bar 1}^{(2)} = 1.$$

Generally, we have

$$\omega_{(S)} = \omega_{(S)}^{(1)} + \omega_{(S)}^{(2)}. \tag{A31}$$

Let a letter sequence which is obtained from (S) by changing all signs be $(\bar S)$ and let its occurring frequency in the symbol be $\omega_{\overline{(S)}}$ of which $\omega_{\overline{(S)}}^{(1)}$ are found in the first $[M]$ and $\omega_{\overline{(S)}}^{(2)}$ in the last $[\bar M]$. In the case of the example shown above, they are

$$(\bar S) = \bar 2 1, \ \omega_{\bar 2 1} = 3 \quad \text{with} \quad \omega_{\bar 2 1}^{(1)} = 1 \quad \text{and} \quad \omega_{\bar 2 1}^{(2)} = 2.$$

Generally, we have

$$\omega_{\overline{(S)}} = \omega_{\overline{(S)}}^{(1)} + \omega_{\overline{(S)}}^{(2)}. \tag{A32}$$

Since the structure is the complex *APD* structure of the type ($[M] \mid [\bar{M}]$), we should have

$$\omega^{(1)}_{(S)} = \omega^{(2)}_{(S)} \quad \text{and} \quad \omega^{(2)}_{(\bar{S})} = \omega^{(1)}_{(S)} \,. \tag{A33}$$

As a result, we have

$$\omega_{(S)} = \omega^{(1)}_{(S)} + \omega^{(1)}_{(\bar{S})} = \omega_{(\bar{S})} \tag{A34}$$

and hence

$$\omega_{(S)} + \omega_{(\bar{S})} = 2\{\omega^{(1)}_{(S)} + \omega^{(1)}_{(\bar{S})}\} \,, \tag{A35}$$

this meaning that the following relation holds in general:

$$\{\Omega_r\} = 2\{\Omega^*_r\} \,. \tag{A36}$$

References

Fujiwara, K. (1957). *J. Phys. Soc. Japan*, **12**, 7.
Kakinoki, J. (1962). *J. Phys. Soc. Japan*, **17**, Supplement B-II, 267.
Kakinoki, J., Kodera, E. & Aikami, T. (1969). *Acta Cryst.* A**25**, 385.

Acta Cryst. (1971). A**27**, 659

Study of Microstructure of Chrysotile Asbestos by High Resolution Electron Microscopy

By Keiji Yada
Research Institute for Scientific Measurements, Tohoku University, Sendai, Japan

(*Received* 3 *February* 1971 *and in revised form* 20 *July* 1971)
(*Dedicated to Professor Tadatosi Hibi in honour o, his* 61*st birthday*)

Several samples of chrysotile asbestos from different localities, including a synthetic sample, were electron-microscopically observed by the lattice imaging method along two directions parallel and perpendicular to the fibre axis. The results are as follows: (*a*) The lattice fringes of 4·5 Å corresponding to 020 were often tilted to the edge of the fibrils with an angular distribution ranging up to about 10° with a peak value at a few degrees, depending on the sample. (*b*) Most of the fibrils examined were hollow cylinders and their circumferential lattice layers form spiral or multi-spiral layers. The perfectly concentric cylindrical layers were also found with a frequency depending on the sample. (*c*) Unusual growth patterns which cannot be explained by Jagodzinski and Kunze's model were observed. (*d*) The lattice images of the conical fibrils (cone-in-cone shape) were observed in the synthetic sample. (*e*) Most fibrils greater than about 350 Å in diameter showed traces of discontinuous growth in two or three steps, depending on the growth conditions, and this gave rise to various distributions of the fibril diameters.

Introduction

From studies using the methods of X-ray diffraction, electron microscopy and electron diffraction, it has been pointed out that there are morphological and structural variations in chrysotile asbestos (Whittaker, 1951; Jagodzinski & Kunze, 1954; Whittaker, 1955, 1956*a*,*b*,*c*, 1957; Whittaker & Zussman; 1956). In earlier X-ray studies, however, single crystals were not available, while in subsequent studies made on single crystals (individual fibrils), by means of electron microscopy combined with selected area electron diffraction, the instrumental resolution was not high enough to resolve the fine structures (Honjo & Mihama, 1954; Zussman, Brindley & Comer, 1957; Bates & Comer, 1957).

Recently, it has become possible to observe lattice planes in the individual fibrils of chrysotile (Fernández-Morán, 1966; Yada, 1967), and it has been found, for example, that the circumferential lattice images observed in the cross-section of a fibril show a spiral or multi-spiral structure. The previous work by the present author, however, was done for a sample from only one source (Quebec, Canada,) and, moreover, the observation of the detailed structure at the inter- and intra-fibril sites was hindered by the damage due to irradiation by the electron beam required for high magnification electron microscopy. Therefore, in order to obtain a comprehensive understanding of the microstructures and growth mechanism of chrysotile, it seemed desirable to study samples from different localities by the use of a improved technique by which the radiation damage was minimized.

The specimens and experimental technique

Table 1 shows a list of the samples examined. Most of these samples are chrysotile ores, except the last one which is powder Mg-chrysotile synthesized under controlled conditions (Noll, Kircher & Sybertz, 1958, 1960).

A small quantity of chrysotile was torn off with tweezers, and as in the previous work (Yada, 1967) observations were made from two directions, parallel and perpendicular to the fibre axis, employing the sectioning technique by ultramicrotomy as well as the