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One Dimensionally Disordered Crystal with a Special Reference to the Anti-Phase Domain Structures

Jiro Kakinoki
Faculty of Science, Osaka City University
Osaka, Japan

The intensity equation and three dimensional Patterson function were calculated for the case of the anti-phase domain structure and some examples are shown.

The intensity of the beam diffracted by a one dimensionally disordered crystal was given by Wilson[1] as

$$I(hk\varphi)=NJ_0+\sum_{m=1}^{N-1}(N-m)J_m e^{-im\varphi}+conj.,$$
$$\varphi=2\pi\zeta\,, \qquad (1)$$

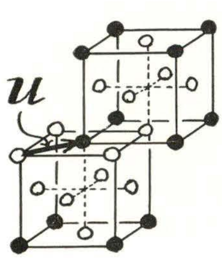
where N is the number of layers, $\varphi=2\pi\zeta$ and ζ the parameter along $\boldsymbol{c}^*$. J_m is the mean of products of two layer form factors separated by m layers and is expressed as[2,3]

$$J_m=\text{spur}\,\boldsymbol{VFP}^m,\quad (\boldsymbol{VF})_{ts}=w_s V_s V_t^*,$$
$$(\boldsymbol{P})_{st}=P_{st}\,, \qquad (2)$$

where V_s is the layer form factor of the layer of s-kind, w_s the existence probability of finding V_s at any position and P_{st} the continuing probability of finding V_t after V_s.

One of the important examples is the displacement stacking fault to which the stacking fault in the close packed structures and the anti-phase domain structures belong. In the latter case we have two kinds of layer form factors as

$$V_1 \text{ and } V_2=\varepsilon V_1 \text{ where } \varepsilon=(-1)^{h+k} \qquad (3)$$



○ : Cu ● : Au

Fig. 1.

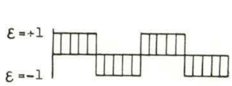


Fig. 2.

corresponding to the out-of-step indicated by $\boldsymbol{u}$ shown in Fig. 1.

In the case where the out-of-step is found at every M layers as shown in Fig. 2 the intensity is expressed by

$$I(\zeta)=V_1V_1^*\frac{\sin^2\pi M\zeta}{\sin^2\pi\zeta}\frac{\sin^2\pi N'(M\zeta-\frac{1}{2})}{\sin^2\pi(M\zeta-\frac{1}{2})}\,,$$
$$N'=\frac{N}{M}\,. \qquad (4)$$

From this equation we can find that the peaks are found at

$$\zeta_n=\frac{2n+1}{2M} \qquad (5)$$

with intensities proportional to

$$\frac{V_1V_1^*}{\sin^2\pi\zeta_n}\,. \qquad (6)$$

Most of examples show such a normal intensity distribution, but some exceptional cases show abnormal intensity distributions. For example, Fujiwara[4] found such an abnormal intensity distribution as shown in Fig. 3 which was explained by him by a model as shown in Fig. 4.

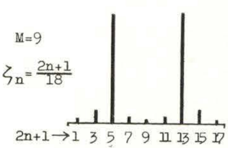


Fig. 3.

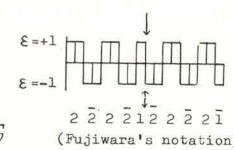


Fig. 4.

In the case when the period is $2M$ and the even order spectra disappear the intensity is generally expressed as

$$I(\zeta)=V_1V_1^*|1-e^{2\pi iM\zeta}|^2SS^*\frac{\sin^2\pi N''2M\zeta}{\sin^2\pi 2M\zeta}\,,$$
$$N''=\frac{N}{2M}=\tfrac{1}{2}N' \qquad (7)$$

which has its maxima at

$$\zeta_l=\frac{l}{2M} \qquad l\colon 0, 1, 2, \cdots, 2M-1 . \quad (8)$$

Here S is a kind of structure factor corresponding to the distribution of the phase factors $\varepsilon=\pm 1$ and is expressed as

$$I_l=|S_l|^2=\left|\sum_{p=0}^{M-1}\varepsilon_p \exp(2\pi i p\zeta_l)\right|^2$$

$$=\sum_{p=0}^{M-1}\sum_{q=0}^{M-1}\varepsilon_p\varepsilon_q \exp\left\{\pi i\frac{(p-q)l}{M}\right\}$$

$$=M+\sum_{m=1}^{M-1}(a_m-b_m)\exp\left(-\pi i\frac{ml}{M}\right)+conj., \quad (9)$$

where $q-p=m$ and a_m and b_m are the numbers of pairs separated by m layers in which $\varepsilon_p\varepsilon_q=+1$ and -1, respectively. So $a_m+b_m=M-m$, hence eq. (9) turns to

$$I_l=\frac{1}{\sin^2\pi\zeta_l}-4\sum_{m=1}^{M-1}b_m\cos\left(\pi\frac{ml}{M}\right),$$

$$\zeta_l=\frac{l}{2M}, \quad l\colon \text{odd} \qquad (10)$$

of which the first term shows the normal intensity distribution and the second one contributes to the abnormal one.

For the Fujiwara's example eq. (10) is written as

$$I_l=I_{18-l}=\frac{1}{\sin^2 l\cdot 10^\circ}-4\sum_{m=1}^{4}(b_m-b_{9-m})\cos(ml\cdot 20^\circ) \qquad (11)$$

and four (b_m-b_{9-m})'s are

$$b_1-b_8=4, \quad b_2-b_7=6, \quad b_3-b_6=0, \quad \text{and} \quad b_4-b_5=-2 . \qquad (12)$$

Substituting eq. (12) into eq. (11) I_l's are calculated as

$$\begin{cases} I_1=I_{17}=1.1, \quad I_3=I_{15}=4.0, \quad I_5=I_{13}=33.2 \\ I_7=I_{11}=1.7, \text{ and } I_9=1.0 \end{cases} \qquad (13)$$

which are the same as the intensities shown in Fig. 3, respectively.

It is desirable, however, to get the model directly from the observed data. I_l's given by eq. (11) are

$$\begin{cases} I_1=33.16-3.76(b_1-b_8)-3.06(b_2-b_7)-2(b_3-b_6)-0.70(b_4-b_5) \\ I_3=\quad 4-2\ (\ \prime\prime\ )+\ 2\ (\ \prime\prime\ )+4(\ \prime\prime\ )+\ 2\ (\ \prime\prime\ ) \\ I_5=\ 1.70+0.70(\ \prime\prime\ )+3.76(\ \prime\prime\ )-2(\ \prime\prime\ )-3.06(\ \prime\prime\ ) \\ I_7=\ 1.13+3.06(\ \prime\prime\ )-0.70(\ \prime\prime\ )-2(\ \prime\prime\ )+3.76(\ \prime\prime\ ) \\ I_9=\quad 1+4\ (\ \prime\prime\ )-\ 4\ (\ \prime\prime\ )+4(\ \prime\prime\ )-\ 4\ (\ \prime\prime\ ) \end{cases} \qquad (14)$$

On the other hand the intensities on the electron diffraction photographs may be estimated, even in the worst case, as follows:

$$I_5>4I_3 \text{ and } I_3>I_1, I_7 \text{ and } I_9 . \qquad (15)$$

In addition to this there are some restrictions which are

$$\begin{cases} 2(I_1+I_3+I_5+I_7)+I_9=M^2=81 \\ I_3 \text{ and } I_9 \text{ are integers} \\ -m\leq(b_m-b_{9-m})\leq 9-m \end{cases} \qquad (16)$$

From eqs. (14) and (16) following table can be obtained. The last condition is found to be satisfied only when

b_3-b_6	$2I_3+I_9$	$I_1+I_3+I_5=\frac{1}{2}(81-2I_3-I_9)$	condition, $I_5>4I_3$
0	9	36	satisfied
1	21	30	not
2	33	24	not

$$b_3-b_6=0 \text{ hence } 2I_3+I_9=9 . \qquad (17)$$

Since both I_3 and I_9 are integers and $I_3>I_9$ we obtain

$$I_3=4 \text{ and } I_9=1 . \qquad (18)$$

Finally following inequalities are obtained from eq. (14)

$$\begin{cases} 29.16<6.82(b_2-b_7)+4.45(b_4-b_5)\leq 33.16 \\ -1.13\leq 2.37(b_2-b_7)+6.82(b_4-b_5)<\ 2.87 , \end{cases} \qquad (19)$$

the unique solution of which is found to be

$$b_2-b_7=6 \text{ and } b_4-b_5=-2 \text{ giving } b_1-b_8=4 . \qquad (20)$$

The final structure $2\bar{2}2\bar{2}1$ is derived even from $b_2-b_7=6$ only, because this value is near its maximum value 7.

When the intensity shows a diffuse scattering and is given by

$$I(h, h+2n+1)=2\pi NV_1V_1^*I_l(\varphi) , \qquad (21)$$

a probability B_m of finding the out-of-step for a pair separated by m layers is given from Zachariasen's W_m[5] by

$$B_m=\frac{1}{2}(1-T_m)\,, \qquad \text{where}$$

$$T_m\left(1-\frac{m}{N}\right)=\int_0^{2\pi} I_1 e^{im\varphi} d\varphi (T_0=1)\,. \qquad (22)$$

References

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3 J. Kakinoki and Y. Komura: J. Phys. Soc. Japan **7** (1952) 30; **9** (1954) 169, 177.
4 K. Fujiwara: J. Phys. Soc. Japan **12** (1957) 7.
5 W.H. Zachariasen: Phys. Rev. **71** (1947) 715.

DISCUSSION

S. OGAWA: I wish to give some comments about the non-integral values of domain size, say $M'=1.8$, talked by Prof. Kakinoki just now. Even if the structure with non-integral domain size has not a strictly regular period, the diffraction maxima will not be largely changed in their intensity and diffuse scattering produced will be vanishingly weak. These effects may be difficult to be detected experimentally. I must say that Dr. Watanabe found a very weak diffraction maximum at one of $n/2\nu M'$ positions in Au_3Mn. Here $2\nu M'=2M=18$ in Prof. Kakinoki's talk. This fact means that the anti-phase structure with non-integral domain size has a period $2\nu M'$, though this may not be strictly regular.

J. KAKINOKI: I agree with your opinion that the intensity can not be measured much accurately in electron diffraction so that neither the weak spots nor the diffuse scattering may be observed. But the method I mentioned may limit, without any missing, the number of structures by which the observed intensities are well explained.