$$(\delta/\delta C^*(\boldsymbol{\xi}))\mathbf{d}(\mathbf{x}) = -M_g(\boldsymbol{\xi})\mathbf{G}(\mathbf{x}|\boldsymbol{\xi}) \cdot \begin{pmatrix} 0 & 0\\ 1 & 0 \end{pmatrix} \cdot \mathbf{d}(\boldsymbol{\xi}).$$
(A.5b)

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## Diffraction Streaks Due to Phase Disorder in One-Dimensional **Displacive Modulation**

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#### Abstract

It can be proved mathematically that the initial phase disorder in the wave of a one-dimensional displacive modulation introduces additional variations in the structure of reciprocal space and, as a result, causes characteristic diffraction streaks. Using a simple twodimensional model, the occurrence of such streaks is confirmed in optical diffraction patterns. Electron diffraction streaks from two crystals with incommensurately modulated structures are presented and explained as the diffraction effect of the phase disorder.

#### 1. Introduction

The wave of structural modulation is usually treated as a plane wave except for discommensurate-type modulation (Janssen & Janner, 1987; Steeds, Bird, Eaglesham, McKernan, Vincent & Withers, 1985). However, when the initial phase is not constant but varies depending on position in real space, an additional diffraction effect is expected. Recently, some streaks passing through the satellite spots were found in the electron diffraction patterns of some materials with a displacive modulated structure (Wu, Li & Hashimoto, 1990; Suzuki, Tanaka, Ishigame,

#### Suemoto, Shibata, Onoda & Fujiki, 1986). It was proposed that there is a group of reflection planes in reciprocal space caused by disorder in the initial phase of the modulation wave.

In this paper, starting from the formula given by de Wolff (1974), it is shown that the initial phase disorder (IPD) changes the structure of reciprocal space and hence some extra diffraction streaks arise. This is also confirmed by optical diffraction for simple two-dimensional models. Finally, electron diffraction streaks observed from two kinds of crystals are shown and discussed from the viewpoint of IPD.

#### 2. Reciprocal space of the crystal with IPD

de Wolff (1974) has given a formula to calculate the structure factor in the case of a one-dimensional displacive modulation. If the modulation wave is sinusoidal, atomic positions can be written as

$$\mathbf{r}_{j} = \mathbf{r}_{j0} + u_{j0} \sin 2\pi (\tau - \alpha_{j}), \qquad (1)$$

where  $\mathbf{r}_i$  is the average position of the *j*th atom,  $u_{i0}$ the displacement amplitude,  $\tau$  the atom coordinate in four-dimensional space and  $\alpha_i$  the initial phase. The structure factor is

$$F(hklm) = \sum_{j} f_{j} \exp 2\pi i [hx_{j0} + ky_{j0} + lz_{j0} + m(\alpha_{j} + \frac{1}{2})] J_{m}(2\pi \mathbf{g} \cdot \mathbf{u}), \qquad (2)$$

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where  $f_i$  is the atomic scattering factor of the *j*th atom,  $J_m$  the *m*th-order Bessel function,  $\mathbf{g} = \mathbf{H} + m\mathbf{q} =$  $h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^* + m\mathbf{q}$  the diffraction vector in reciprocal space,  $\mathbf{q} = q_1 \mathbf{a}^* + q_2 \mathbf{b}^* + q_3 \mathbf{c}^*$  the modulation wave vector and m is an integer.

Usually the initial phases  $\alpha_i$  are taken as constants for all atoms located at equivalent positions. In other words, a plane P normal to the modulation direction in real space can be a wavefront of the modulation wave, where all the equivalent atoms are displaced by an equal amount. But after introducing IPD,  $\alpha_i$ varies depending on the position in the plane P and is written as  $\alpha_i(\mathbf{p})$ . Here **p** is a vector in the plane P.

For simplicity, a model with only two atoms in a unit cell, A and B, is used in the following discussion. Their positions are displaced by the modulation wave with the same wave vector. The initial phase is constant for A atoms  $(\alpha_A = \alpha)$  but variable for B atoms  $[\alpha_B = \beta(\mathbf{p})]$ . For this simple model the structure factor (2) can be written as

$$F(uvwm)$$

$$= f_A \exp 2\pi i [\mathbf{H} \cdot \mathbf{r}_{A0} + m(\alpha + \frac{1}{2})] J_m(2\pi \mathbf{g} \cdot \mathbf{u}_{A0})$$

$$+ f_B \exp 2\pi i \{\mathbf{H} \cdot \mathbf{r}_{B0} + m[\beta(\mathbf{p}) + \frac{1}{2}]\}$$

$$\times J_m(2\pi \mathbf{g} \cdot \mathbf{u}_{B0}), \qquad (3)$$

where  $\mathbf{H} = u\mathbf{a}^* + v\mathbf{b}^* + w\mathbf{c}^*$ . u, v and w are used as the indices only to emphasize that after introducing  $\beta(\mathbf{p})$  variation occurs in reciprocal space.

The intensity of the diffraction pattern is:

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$$I(uvwm) \propto \left| \sum_{n}^{N} F(uvwm) \right|^{2}$$
  
=  $(1/N_{1}N_{2}N_{3}^{2})[f_{A}^{2}J_{m}(2\pi\mathbf{g} \cdot \mathbf{u}_{A0})T_{1}$   
+  $2f_{A}f_{B}J_{m}(2\pi\mathbf{g} \cdot \mathbf{u}_{A0})J_{m}(2\pi\mathbf{g} \cdot \mathbf{u}_{B0})$   
× Re  $(T_{2}C) + f_{B}^{2}J_{m}(2\pi\mathbf{g} \cdot \mathbf{u}_{B0})T_{3}],$  (4)

where  $C = \exp 2\pi i [\mathbf{H} \cdot (\mathbf{r}_{A0} - \mathbf{r}_{B0}) + m\alpha]$  represents a constant phase factor,  $N_1$ ,  $N_2$  and  $N_3$  the numbers of unit cells along the three principal axes, Re the real part of  $T_2C$ , and

$$T_{1} = \sum_{n,n'}^{N} \exp 2\pi i \mathbf{H} \cdot (\mathbf{R} - \mathbf{R}')$$
  
=  $\sum \exp 2\pi i [u(n_{1} - n'_{1}) + v(n_{2} - n'_{2}) + w(n_{3} - n'_{3})]$   
=  $(N_{1}N_{2}N_{3})^{2} \delta_{uh} \delta_{vk} \delta_{wl}$  (h, k, l are integers), (5)

$$T_{2} = \sum_{\mathbf{n},\mathbf{n}'}^{N} \exp 2\pi i [\mathbf{H} \cdot (\mathbf{R} - \mathbf{R}') - m\beta(\mathbf{p})]$$
  
=  $\sum \exp 2\pi i [u(n_{1} - n_{1}') + v(n_{2} - n_{2}') + w(n_{3} - n_{3}') - m\beta(\mathbf{p})]$  (6)

$$T_{3} = \sum_{\mathbf{n},\mathbf{n}'}^{N} \exp 2\pi i \{ u(n_{1} - n_{1}') + v(n_{2} - n_{2}') + w(n_{3} - n_{3}') + m[\beta(\mathbf{p}) - \beta(\mathbf{p}')] \},$$
(7)

where  $\mathbf{R} = n_1 \mathbf{a} + n_2 \mathbf{b} + n_3 \mathbf{c}$   $(n_1, n_2, n_3 \text{ are integers})$ ,  $\mathbf{N} = (N_1, N_2, N_3), \mathbf{n} = (n_1, n_2, n_3)$  and  $\delta$  is the delta function.

 $T_1$  is related only to A atoms, whose displacement is defined by a plane modulation wave and gives sharp reflection spots, while  $T_2$  and  $T_3$  contain the information from B atoms. When  $\beta(\mathbf{p})$  is constant (as a special case), we have from (6) and (7)

$$T_2 = T_3 = (N_1 N_2 N_3)^2 \delta_{uh} \delta_{vk} \delta_{wl}$$

This means that all diffraction spots are sharp. This is the condition satisfied in the case of the normal modulated structure.

When  $\beta(\mathbf{p})$  is a function variable on the plane P, the diffraction intensity is no longer as simple and some extra diffraction effects arise. In following discussions  $\beta(\mathbf{p})$  is assumed to be a random function; the initial phase is assumed to be completely disordered. Under this assumption, three different conditions for the plane P should be considered.

#### (i) Plane P coincides with a principal plane (**a**, **b**)

This condition implies that the initial phase function can be expressed as

$$\beta(n_1, n_2). \tag{8}$$

Substituting this function into (6) and (7) gives

$$T_{2} = \sum \exp 2\pi i w (n_{3} - n'_{3})$$

$$\times \sum \exp 2\pi i [u(n_{1} - n'_{2}) + v(n_{2} - n'_{2}) - m\beta(n'_{1}, n'_{2})]$$

$$= N_{3}^{2} \delta_{wl} \sum \exp 2\pi i [u(n_{1} - n'_{1}) + v(n_{2} - n'_{2}) - m\beta(n'_{1}, n'_{2})] \qquad (9)$$

$$T_{3} = N_{3}^{2} \delta_{wl} \sum \exp 2\pi i \{ u(n_{1} - n_{1}') + v(n_{2} - n_{2}') + m[\beta(n_{1}, n_{2}) - \beta(n_{1}', n_{2}')] \}.$$
(10)

Equations (9) and (10) lead to non-zero  $T_2$ ,  $T_3$  on the planes whose index w is an integer. On the other hand, there are no limitations for the other two indices. Any reciprocal lattice planes parallel to  $(\mathbf{a}^*, \mathbf{b}^*)$  satisfy the reflection condition for  $T_2$  and  $T_3$ . When the reciprocal spaces are intersected by the reflection spheres, not only sharp spots but also some streaks are excited.

In the case m = 0, it is found from (9) and (10) that

$$T_2 = T_3 = (N_1 N_2 N_3)^2 \delta_{uh} \delta_{vk} \delta_{wl}.$$

This means that these reflection planes only pass through satellite spots, while all main spots always remain sharp. This is evidently different to the diffraction effect from IPD and from that of conventional crystal defects such as stacking faults.

When A atoms are located strictly at the average positions,  $\mathbf{u}_{A0} = \mathbf{0}$ , it is found from (4) that  $J_m(2\pi \mathbf{g} \cdot \mathbf{u}_{A0}) = 0$  if  $m \neq 0$ . This means that A atoms do not give any contribution to satellite spots, while B atoms contribute to the formation of streaks. Some diffraction patterns obtained under this condition have been reported (Watanabe, Fujiki, Yoshikado & Ohachi, 1988; Terauchi, Futamura, Ishii & Fujiki, 1984; Beyeler, 1976).

# (ii) Plane P coincides with a lattice plane (hkl) (at least two of h, k and l are non-zero)

In this case  $\beta(\mathbf{p})$  cannot be written as (8) directly, however, fortunately, because of the coincidence of the plane P with the (*hkl*) plane, it is possible to select two new unit-cell vectors on the plane P instead of the old ones. By using the new basic vectors, expressions similar to (9) and (10) can be obtained. This means that streaks also occur in diffraction patterns.

#### (iii) Plane P does not coincide with any lattice planes

The function  $\beta(\mathbf{p})$  will depend on three variables,  $n_1$ ,  $n_2$  and  $n_3$ , in this case. No  $\delta$  function is obtained from (6) and (7). The  $T_2$  and  $T_3$  are non-zero at any reciprocal-lattice point and their contribution only broadens diffraction spots.

It should be noted that the incommensurate condition is not used at all in the discussion above. This means that previous conclusions can be used not only for the incommensurately but also for the commensurately modulated structure (superlattices). Since the plane P coincides with a lattice plane in the superlattice structure, the IPD in a displacive superlattice always produces the reflection planes. The high-order satellite spots sometimes coincide with the main spots. Therefore, some streaks pass through the main spots. This apparently shows a difference between the streaks from an incommensurate phase and those from a superlattice phase. In this case it is sometimes difficult to elucidate the origin of the streaks clearly, because similar streaks can be produced by defects such as stacking faults.

All of the previous discussions are based on a very simple model, in which two assumptions are introduced. (1) There are only two atoms in one unit cell. One (A) is displaced with the modulation and another (B) is displaced with the modulation added by IPD. (2) The modulation is described by a sinusoidal wave. In spite of these simple assumptions, this method can be extended to more complicated structures, for example, to a structure composed of many atoms in a unit cell, without difficulties. Furthermore, this method might also be applied for a chimney ladder structure [see section 4(ii)] or a substitutional modulation structure, which would give similar diffraction patterns.

#### 3. Optical diffraction patterns (ODPs)

By an optical diffraction method, the effect of IPD in a modulated structure can be shown directly using a simple two-dimensional model: (1) the unit cell is a simple square, a = b; (2) it is composed of only two atoms, A and B, in a unit cell and their average positions are (0, 0) and  $(\frac{1}{2}, \frac{1}{2})$ , respectively; (3) both of them are modulated by waves with the same wave vector and amplitude; (4) for atoms A the initial phase is a constant, while for B,  $\beta(\mathbf{p})$  is a constant (*i.e.* the whole modulation structure is perfect) in case (a) or  $\beta(\mathbf{p})$  is randomly chosen between 0 and 20° (IPD) in case (b). Under these conditions, the following three types of wave vectors must be considered. (i)  $\mathbf{q} = \mathbf{a}^*/2 \cdot 3$ 

Figs. 1(a) and (c) show the atomic positions for cases (a) and (b), respectively. The arrow **q** shows the propagating direction of the modulation wave. Along another arrow **p**, normal to **q**, the array of atoms with a displacive modulation can be seen clearly in Fig. 1(a) but not in Fig. 1(c). Figs. 1(b)and (d) show the ODPs corresponding to Figs. 1(a)and (c), respectively. In Fig. 1(b) all the diffraction spots are sharp, while in Fig. 1(d) every satellite spot is accompanied by some extra weak spots along **b**<sup>\*</sup>. The extra spots originate owing to the introduction of the IPD. Because of the low intensity of the laser

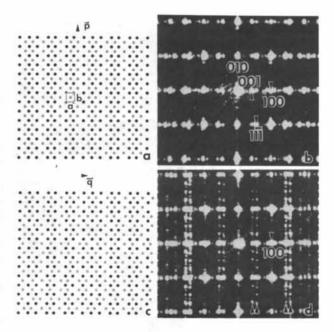


Fig. 1. Two-dimensional crystal models constructed by two atoms A and B, whose positions are indicated by dots and open circles, respectively. (a) The positions of both atoms are modulated by the plane wave propagating along the a axis with  $\mathbf{q} = \mathbf{a}^*/2\cdot 3$ . (b) The ODP corresponding to (a). Indices hkm are indicated. (c) The positions of B atoms are modulated by the wave with random initial phase between 0 and 20°. (d) The ODP corresponding to (c). Diffraction streaks arise passing through satellite spots.

source for this optical diffractometer, only a small area as in Fig. 1(d) could contribute in the optical transformation. If the ODP could be obtained from a much larger area, the extra spots in Fig. 1(d) would become weak streaks.

(ii)  $q = (a^* - b^*)/2 \cdot 3$ 

The perfect modulation structure is shown in Fig. 2(*a*) and the IPD structure is shown in Fig. 2(*c*). If one looks along the arrow **p**, the array of atoms in lines can be seen clearly in Fig. 2(*a*), but not so clearly in Fig. 2(*c*). The ODPs in Figs. 2(*b*) and (*d*) correspond to the structures in Figs. 2(*a*) and (*c*), respectively. The extra spots accompanying every satellite spot are along the  $\mathbf{a^*} - \mathbf{b^*}$  direction. The main spots, (*hk*0), are sharp in Fig. 2(*d*). Except for the direction of the streaks, Fig. 2(*d*) is very similar to Fig. 1(*d*). (iii)  $\mathbf{q} = q_1 \mathbf{a^*} - q_2 \mathbf{b^*}$ ,  $q_2/q_1 = \sqrt{2}$ ,  $|q| = d_{(11)}/2$ 

Figs. 3(a) and (c) show the perfect structure and the IPD structure for the atoms whose positions are modulated by the wave propagating along the direction  $\theta = 54.7^{\circ}$  ( $\theta$  is the angle between **a** and **q**), respectively. The plane *P* does not coincide with any lattice planes. Figs. 3(b) and (d) show the ODPs taken from Figs. 3(a) and (c), respectively. Comparison of Fig. 3(b) with Fig. 3(d) reveals that the main reflection spots do not show any obvious change, but the satellite spots change markedly. Unlike the observation with regard to Figs. 1(d) and 2(d), there are no streaks at all in Fig. 3(d). Some satellite spots, which are indicated by arrows, are broadened. We may conclude that the sharp satellite spots in Fig. 3(d) are due to the contribution from A atoms, while the broadened spots are due to B atoms. As a result, the IPD does not produce streaks, but makes the spots diffuse.

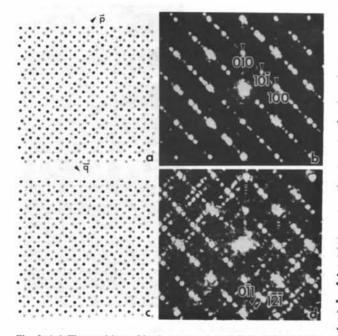
We may say that the ODPs in Figs. 1, 2 and 3 are in good agreement with the conclusion reached in the preceding section. Though these ODPs have been obtained using a two-dimensional model, it is not difficult to extend them to the three-dimensional case.

### Electron diffraction patterns (EDPs) in ankangite and (Sr, Ca)<sub>3</sub>Cu<sub>5+δ</sub>O<sub>y</sub>

#### (i) Ankangite

Ankangite is a new mineral discovered recently in China (Zhou & Ma, 1987). X-ray and electron diffraction studies show that it belongs to the tetragonal system with unit-cell parameters a = 10.2 and c = 2.96 Å (Wu, Li & Hashimoto, 1990; Zhou & Ma, 1987). The incommensurate modulation structure along the c axis is found with  $\mathbf{q} = \mathbf{c}^*/2.30$ . Further analysis has shown the formation of a displacive modulation structure (Wu, Li & Hashimoto, 1990).

Fig. 4 shows the EDPs taken along (a) [100], (b) [101], (c) [102] and (d) [103]. It is interesting that some streaks can be seen at the positions indicated by the arrows S and R, and all satellite spots are located on these streaks as seen in Figs. 4(a) and (c).



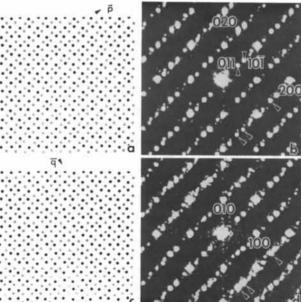


Fig. 2. (a) The position of both atoms, A and B, is defined by a plane modulation wave with vector  $\mathbf{q} = (\mathbf{a}^* - \mathbf{b}^*)/2 \cdot 3$ . (b) The ODP corresponding to (a). (c) The position of B atoms is defined by a modulation wave with random initial phase between 0 and 20°. (d) The ODP corresponding to (c). The array of atoms along the p direction is clearly seen to be roughly in lines in (a) but not so clearly in (c). There are some streaks along  $\mathbf{a}^* + \mathbf{b}^*$  in (d) but no streaks in (b).

Fig. 3. (a) The position of both dots is defined by the plane modulation waves with  $\mathbf{q} = \mathbf{q}_1 \mathbf{a}^* - \mathbf{q}_2 \mathbf{b}^*$ ,  $q_2/q_1 = \sqrt{2}$  and  $|\mathbf{q}| = d_{(11)}/2$ . (b) The ODP corresponding to (a). (c) The positions of B atoms are defined by a modulation wave with IPD. (d) The ODP corresponding to (c). There are no streaks in (b) or (d). The satellite spots are broadened in (d).

The cross section in reciprocal space is presented in Fig. 5. The dots indicate the main spots, the open circles the first-order satellite spots and the dashed lines the reflection planes caused by IPD. Straight lines a, b, c, d and e show the reflection spheres for the incident electron beam parallel to [100], [101], [102], [103] and [104], respectively.  $S_1$  and  $R_1$  indicate the distance from the center spot to the nearest

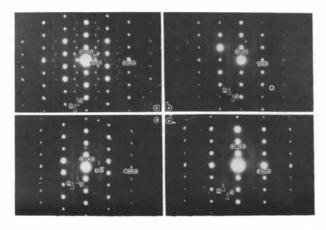


Fig. 4. Electron diffraction patterns taken from ankangite with the incident beam parallel to (a) [100], (b) [101], (c) [102] and (d) [103]. In every EDP the streaks are found at the positions indicated by the arrows. It is known from (a) and (c) that all satellite spots are always located on these streaks. Indices *hklm* are indicated.

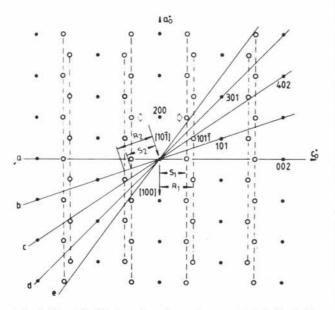


Fig. 5. The  $(\mathbf{a}^*, \mathbf{c}^*)$  plane in reciprocal space. Dots indicate the main spots, the open circles the first-order satellite spots and the dashed lines the reflection plane.  $S_1$  and  $R_1$  show the distance from the center spot to the nearest reflection lines for the incident beam parallel to [100], and  $S_2$  and  $R_2$  are for the incident beam parallel to [101]. The lines a, b, c, d and e show the reflection spheres for the incident beam parallel to  $[10\overline{2}]$ ,  $[10\overline{3}]$  and  $[10\overline{4}]$ , respectively.

reflection planes for the incident beam parallel to [100], and  $S_2$  and  $R_2$  are for the incident beam parallel to [101]. It is seen from this figure that the reflection spheres always interesect the reflection planes except for the case of the incident beam being along [001]. This is in good agreement with the observation that there are always some streaks in the EDPs of Fig. 4.

When the electron beam is incident along [102], on the other hand, the reflection sphere does not cross satellite spots such as  $101\overline{1}$  and 3011. But if we consider the elongation of the spots, which might be caused by a crystal form or defects, it is possible to observe the trace of these spots. Fig. 4(c) must correspond to a case such as this.

In this crystal the origin of the modulation structure must be connected with the array of ions in the channels. The ion radius is a little smaller than that of the channel (Zhou & Ma, 1987) and hence there is freedom of movement along the channel. In a channel, the interaction among ions is strong so that they regularly distribute with a modulation period. On the other hand, the interaction between ions in neighboring channels is weak so that the initial phases in each channel are approximately independent. This must be a major reason for the occurrence of the initial phase disorder.

#### (ii) $(Sr, Ca)_3Cu_{5+\delta}O_y$

 $(Sr, Ca)_3Cu_{5+\delta}O_y$  has been found as a minor phase in bismuth-based high- $T_c$  superconductive oxides (Horiuchi, Shoda, Tsutsumi, Kosuda & Nozaki, 1989). X-ray single-crystal diffraction analysis has revealed that it belongs to the orthorhombic system with unit-cell parameters a = 12.811, b = 11.3446 and  $c = 5 \times 3.9035$  Å (Kato, Muromachi, Kosuda & Uchida, 1988). X-ray and electron diffraction analysis have shown that there are two sets of incommensurate lattices along the c axis with  $c_1 = 3.90$  and  $c_2 = 2.75$  Å, to form a chimney ladder structure (Kato et al., 1988; McCarron, Subramanian, Calabrese & Harlow, 1988; Siegrist, Schneemeyer, Sunshine, Waszczak & Roth, 1988; Wu & Horiuchi, 1991). Fig. 6 shows a series of EDPs taken with the incident electron beam along (a) [110], (b) [120], (c) [130] and (d) [150]. In spite of the modulated mechanisms being different from each other, the EDPs in Figs. 6 and 4 are very similar; the streaks always appear passing through the satellite spots. In Fig. 6, EDPs are taken on tilting the crystal around the  $c^*$  axis and the distances from the center to the streaks are invariable. In other words, the streaks can always be found at the same position in different EDPs, as indicated by the arrows. The weak spots indicated by a small arrow in Fig. 6(b) are produced for the same reason as that mentioned for 1011 in Fig. 4(c).

It can be seen that there is essentially no difference between Figs. 4, 6 and 1(d). All the streaks must therefore be due to the contribution from atoms which are modulated by the displacive wave with disordered initial phases in ankangite. On the other hand, in the crystal (Sr, Ca)<sub>3</sub>Cu<sub>5+ $\delta$ </sub>O<sub> $\nu$ </sub>, the streaks arise from a set of sublattices  $(c_2 = 2.75 \text{ Å})$  with different initial phases (Wu & Horiuchi, 1991). That is to say, some of the equivalent atoms in this sublattice start with different z, but have a strict period along the c direction. In other words, this sublattice is disordered partially in the (a,b) plane. From the viewpoint of mathematics, the initial phase disorder in the displacive modulation structure and the sublattice phase disorder in the chimney ladder structure can be treated almost equally. Therefore, it is not surprising that we obtain similar diffraction patterns from these two crystals. We may thus say that the method proposed in this paper can be used not only for the displacive modulation structure but also for the other modulation structures.

#### 5. Concluding remarks

Mathematical derivation shows that IPD in a onedimensional displacive modulated structure changes the structure of reciprocal space and produces reflec-

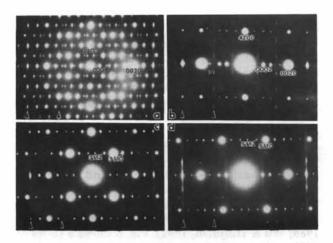


Fig. 6. EDPs taken from (Sr, Ca)<sub>3</sub>Cu<sub>5+8</sub>O, crystal with incident beam along (a) [110], (b) [120], (c) [130] and (d) [150]. The streaks passing through weak spots can be seen.

tion streaks in diffraction patterns. (i) When a modulation wave propagates along a zone [hkl], some reflection planes parallel to the (hkl) plane occur in reciprocal space; all these planes always pass through the satellite spots. (ii) If the plane P at the modulation wavefront does not coincide with any lattice plane, the IPD broadens the satellite spots. ODPs obtained from the simple two-dimensional models show directly the structure of reciprocal space after the change on introducing IPD and the results coincide well with the mathematical conclusion. Using the IPD model, we can also explain the streaks in EDPs obtained from ankangite crystals. It is shown that this method can also be used for other modulation structures, such as the chimney ladder structure in  $(Sr, Ca)_3Cu_{5+\delta}O_{\nu}$ , to explain the similar diffraction patterns.

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