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Electron Diffraction Symmetries in Reflections in Higher-order Laue Zones

Simetrías de Difracción Electrónica en Reflecciones de Zonas Superiores de Laue

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Abstract: The standard symmetry tables for convergent-beam diffraction are for the case of reflections in the zero-order Laue zone. However, the symmetries of reflections in the higher-order Laue zones are in general different from the symmetries of the reflections in the zero-order Laue zone. Four symmetry operations produce symmetry relations for reflections in the zero-order Laue zone only, the corresponding symmetry operations are missing in reflections in higher-order Laue zones. Twenty-one of the 31 diffraction groups have one or more of these four operations (alone or in combination with other symmetry operations). The four operations are those operations which exchange "up" and "down"; they are an inversion center, a horizontal mirror, a horizontal two-fold axis and a four-fold inversion axis. Despite the difference between the symmetries of zero-layer and higher-order-Laue-zone reflections, the pattern symmetries of on-axis convergent beam patterns are unmodified.

Key words: Convergent-beam diffraction, diffraction groups, symmetry, Higher-order Laue Zones (HOLZ), zone-axis patterns.

Resumen: Las tablas de simetría standard para difracción convergente del rayo se utilizan para el caso de reflecciones en el orden-cero de la zona de Laue. Sin embargo, las simetrías de reflecciones en las zonas de Laue de alto orden son en general diferentes de las simetrías de las reflexiones en el orden-cero de la zona de Laue. Cuatro operaciones de simetría producen relaciones de simetría para las reflexiones en el orden-cero de Laue únicamente, las operaciones de simetría correspondientes están ausentes en las reflexiones en el orden superior de las zonas de Laue. Veintiuno de los 31 grupos de la difracción tienen uno o más de estas cuatro operaciones (sólo o en la combinación con otros funcionamientos de simetría). Las cuatro operaciones son aquellas que intercambian "arriba" y "abajo"; ellas están en un centro de inversión, un espejo horizontal y un eje horizontal de dos pliegues y un eje de inversión de cuatro pliegues. A pesar de la diferencia entre las simetrías de capa-cero y reflexiones de orden superior de la zona de Laue, los patrones de simetrías de rayos convergentes no están modificados.

Palabras clave: Difracción convergente, difracción de grupos, simetría, zonas de Laue de alto orden, patrones de zonas ejes.

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Introduction

Convergent-beam diffraction patterns taken at (or near) a zone axis have symmetries which are related to the crystal symmetry of the sample. Thus, the symmetry of convergent-beam patterns is used to characterize the symmetry of the sample. This is done using tables which relate the diffraction symmetry to the crystal symmetry. These tables were first given by Buxton et al (1). The tables give the symmetry within each reflection and the symmetry between the reflections, in the diffraction pattern are quite general, except that without explicitly saying so, the results are given for the reflections in the zero-order Laue zone (referred to for convenience and brevity as the zero layer).

Tanaka et al. (2), pointed out that the symmetries in and between the reflections in higher-order Laue zones (HOLZ) are different from those in the zero layer. The paper of Buxton et al. (1), is incomplete in this sense. The present paper completes the work of Tanaka et al. (2), who did not describe all such examples in a systematic way.

The principle used to derive the symmetry in a convergent-beam pattern is this: two "experiments" are found, involving two different directions of the incident beam, such that the two experiments are related by a symmetry operation, and with the use of the reciprocity principle (3), if needed. Then the intensities of those two orientations in a convergent beam pattern will be the same and the pattern develops a corresponding symmetry.

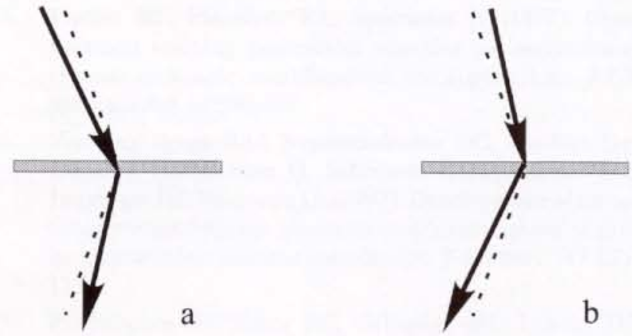

Figure 1. Schematic diagram showing the incident beam and one diffracted beam in two different orientations close to but not at the Bragg angle for the reflection. The dashed line represents the orientation of corresponding to the Bragg angle.

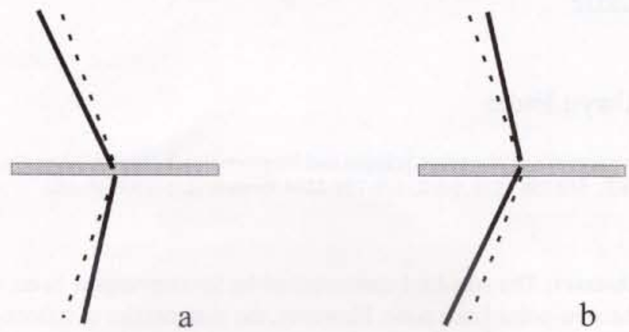

Figure 2. Schematic diagram as for Figure 1 except that the arrowheads have been removed. The reciprocity theorem states that the rays can be reversed, so the arrowheads are not relevant. Now it can be seen that the two diagrams are related by a reflection across a horizontal line through the sample.

An example: a horizontal mirror

Consider the case of a sample in which the crystal structure contains a mirror plane which is horizontal (i.e. parallel to the foil and normal to the beam). We can choose two directions of the incident beam and show that they must give rise to two equal intensities in the diffraction pattern because they are related by the mirror operation. In a dark field disc, these directions are on opposite sides of the center of the pattern. Therefore, each dark field disc has a two fold axis as a result of the presence of the mirror plane.

Fig. 1a shows an incident beam and a diffracted beam. The dashed line indicates the orientation of the incident beam that would be at the exact Bragg angle for this reflection. Fig. 1b shows a different orientation of the incident beam and the corresponding diffracted beam. These two diagrams are the same, if one of them is reflected about a horizontal line through the sample. Actually, they are not the same but they would be the same if the arrows were reversed. Since the reciprocity theorem (3) tells us that we are allowed to reverse the direction of the arrows, we can take off the arrow heads (Fig. 2) and the two diagrams are indeed the same if one pattern is reflected about a horizontal line. Therefore, the two directions in the diffracted beam represented by the two outgoing rays in Figs. 1a and 1b must have the same intensity. These two rays are at equal angles from the position of the exact Bragg angle and the same relation will exist between all pairs of rays that are at opposite positions with respect to the Bragg point. The diffracted beam must contain a two-fold symmetry axis. Indeed all diffracted discs for a sample with a horizontal mirror must have two-fold symmetry.

A point which may need clarification arises from the fact that Figs. 1 and 2 are two-dimensional and represent a section through a situation which is essentially three-dimensional. In two dimensions, a two-fold axis and a mirror are the same. To establish that in the case of the horizontal mirror it is a two-fold axis and not a mirror, it is necessary to consider what happens if the ingoing ray in figure 1a, for example, is not in the plane of the paper but slightly above it. This will reveal that the result is indeed a two-fold rotation. In the work of Buxton et al. (1), the diagrams were drawn using the stereographic projection to make the understanding of this three-dimensional aspect of the analysis easier.

The analysis given above, is carried out on the assumption that the two diffracted directions are direc-

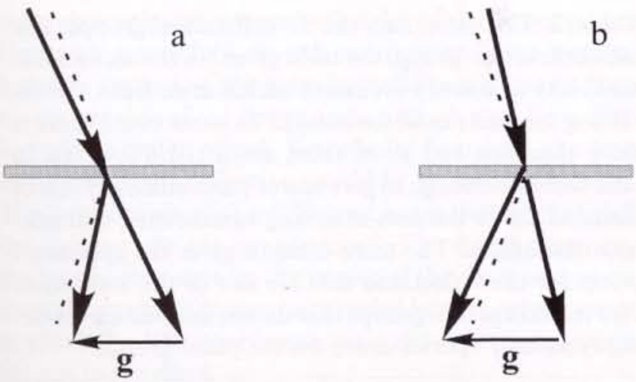

Figure 3. In this diagram the direct (or transmitted) beam is added to the figure so that the diffraction vector g can be shown. Since the vector is the same in the two diagrams, the diffracted beams of Figures 2a and 2b correspond to different points for the same reflection.

tions within the same diffracted beam. This is the case only if the diffracted beam is in the zero-order Laue zone. Fig. 3 shows the direct beam as well as the incident and diffracted beams. From this, we can see that the **g** vector is the same for the two cases and that it is parallel to the sample (and therefore to the mirror plane).

If instead we look at a reflection which is in the first-order Laue zone, the **g** vector is not parallel to the mirror, Fig. 4. This is similar to Figs. 1 to 3 except that the dashed line representing the exact Bragg condition for the diffracted beam is not symmetric with respect to the mirror. Now, it is clear that the two directions in the diffracted beam (represented in Figs. 4a and 4b) are not related by a mirror operation. If instead we look to see which directions are mirror related (Fig. 5), we see that the two directions that give the same diffracted intensity are in different reflections. One is in the first-order Laue zone while the other is in the Laue zone of order minus one. This implies that there is a symmetry between reflections in Laue zones of opposite sign. In normal convergent beam diffraction, of course, Laue zones with negative order are not seen. In the conical scan method of Tanaka et al. (1), they are seen and the fact that the patterns are related to the Laue zones of positive order is put to good use there.

The general result

We can generalize the above result and see that any diffraction symmetry produced by a symmetry operation that exchanges the top and bottom of the sample will apply to reflections in the zero layer only. We note that many of the symmetries in diffracted beams are produced by symmetry operations which do not exchange the top and bottom of the sample. These include rotations that have the rotation axis perpendicular to the sample and mirrors that are perpendicular to the sample. Crystal symmetries of this kind produce symmetries in the diffracted beams that affect the zero-layer and higher-order reflections equally.

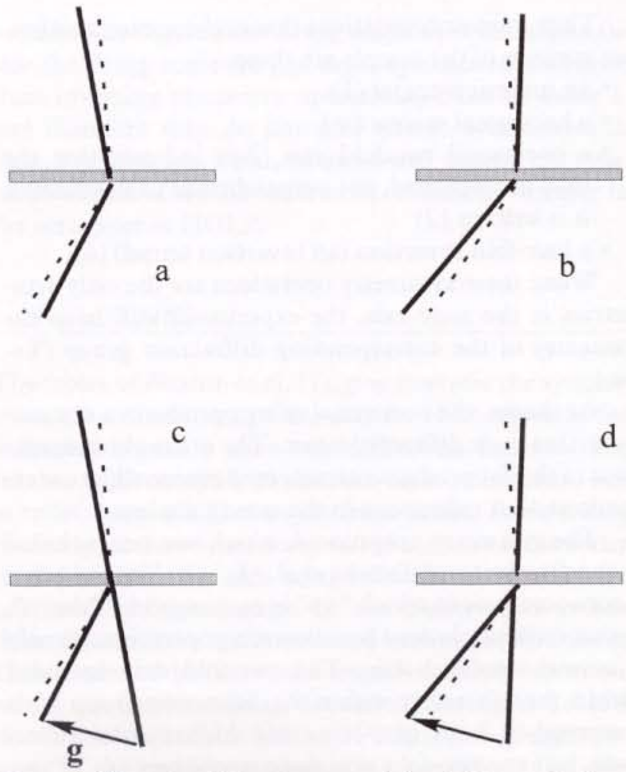

Figure 4. When the diffraction vector is in a higher-order Laue zone, the dashed line (corresponding to the Bragg condition) is not symmetric about the sample, the g vector is not parallel to the sample, and the two situations represented by Figures a and b are not related by a mirror operation.

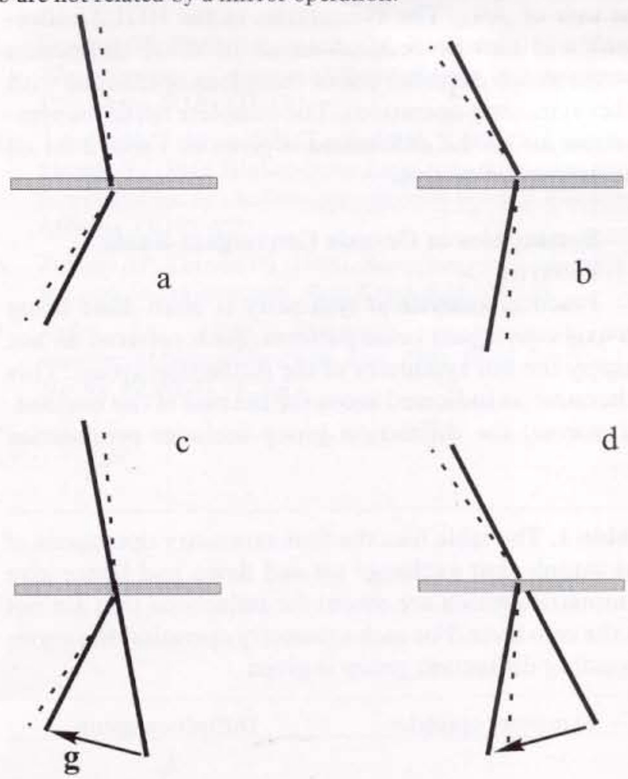

Figure 5. The two orientations represented by Figures a and b are now chosen to be related by the mirror operation, but the completion of the diagrams in Figures c and d shows that the two conditions do not correspond to reflections in the same Laue zone.

The symmetry operations that exchange top and bottom surfaces of the sample are these:

- an inversion center ($\bar{1}$)
- a horizontal mirror (m)
- a horizontal two-fold axis (2-to indicate that the diad is horizontal, not perpendicular to the surface, it is written 12)
- a four-fold inversion (an inversion tetrad) ($\bar{4}$)

When these symmetry operations are the only symmetries at the zone axis, the experiment will have the symmetry of the corresponding diffraction group (Table 1).

As shown, the horizontal mirror produces a symmetry within each diffracted beam. The other three operations in the list produce a symmetry between different reflections - but reflections in the same Laue zone.

The symmetry operation $\bar{4}$, which was not included in the discussion of Tanaka et al. (2), is unlike the other three operations in which "up" is exchanged for "down", in that it also includes a non-inverting operation, namely a normal two-fold axis. The two-fold axis included within (and therefore within the diffraction group 4_R) is preserved in both zero-layer and higher-order reflections, just as a two-fold axis alone would be.

The symmetries described in Buxton et al. (1) as occurring in the case of these diffraction groups will not occur in the reflections in the higher-order Laue zone reflections, with the exception of the two-fold symmetry in the case of $\bar{4}/4_R$. The symmetries of the HOLZ reflections will also be reduced in all of those diffraction groups which combine one of these four operations with other symmetry operations. The complete list of the symmetries for HOLZ reflections is given in Table 2 for all the diffraction groups.

Symmetries in On-axis Convergent-Beam Patterns

Practical analysis of symmetry is often done using on-axis convergent-beam patterns. Such patterns do not display the full symmetry of the diffraction group. This is because (as indicated above for the case of the horizontal mirror) the diffraction group includes symmetries

Table 1. The table lists the four symmetry operations of the sample that exchange up and down and hence give symmetries which are absent for reflections that are not in the zero layer. For each symmetry operation the corresponding diffraction group is given

Symmetry operation	Diffraction group
$\bar{1}$	2_R
m	1_R
12	m_R
$\bar{4}$	4_R

Table 2. The table lists the 31 diffraction groups. For each diffraction group, the table gives (in the second column) any symmetry elements which arise from the inverting symmetries of the sample. In some cases, there is more than one and all of them are listed. However, in each case it is enough to give one of them since any one of them, added to the non-inverting symmetries, will generate the others. The third column gives the symmetry group for the reflections that are not in the zero layer. The ten diffraction groups that do not include an inverting symmetry operation are the ten plane groups

Diffraction Group	Inversion Operations	Symmetry group for HOLZ reflections
1	-	
1_R	1_R	1
2	-	
2_R	2_R	1
21_R	$1_R(2_R)$	2
m_R	m_R	1
m	-	
$m1_R$	$1_R(m_R)$	m
$2m_Rm_R$	m_R	2
2mm	-	
2_Rmm_R	$2_R(m_R)$	m
$2mm1_R$	$1_R(2_Rm_R)$	2mm
4	-	
4_R	4_R	2
41_R	$1_R(4_R2_R)$	4
$4m_Rm_R$	m_R	4
4mm	-	
4_Rmm_R	$4_R(m_R)$	2mm
$4mm1_R$	$1_R(4_R2_Rm_R)$	4mm
3	-	
31_R	1_R	3
$3m_R$	m_R	3
3m	-	
$3m1_R$	$1_R(m_R)$	3m
6	-	
6_R	2_R	3
61_R	$1_R(2_R)$	6
$6m_Rm_R$	m_R	6
6mm	-	
6_Rmm_R	$2_R(m_R)$	3m
$6mm1_R$	1_R	6mm

that relate orientations on both sides of the Bragg point. However, a standard on-axis convergent-beam pattern includes regions of the zero-layer diffracted beams that are on one side of the Bragg point only. A zero-layer disc includes a region which lies wholly "outside" the line along which the reflection would be at the Bragg angle. The orientations of the diffracted beam that lie "inside" the Bragg angle are not observed. The direct beam includes the zone axis (at the center of the disc) so that its internal symmetries may be revealed but the orientations at which the diffracted beams are at the exact Bragg angle are not in the discs.

In using the symmetry of convergent-beam patterns to characterize crystal symmetry therefore, the full symmetry of the diffraction group is generally not found. Only those symmetries which can be seen in the on-axis pattern are used. These are the symmetries called the "whole pattern" symmetry and the "bright field" symmetry; there are two possibilities for each them, the full or three-dimensional symmetry and the "projection symmetry", the symmetry which is given if the effects of HOLZ diffraction are invisible or ignored (1, 4). These four symmetries represent the maximum information which may be obtained from an on-axis pattern.

The four inverting symmetries are those symmetries which relate orientations of the beam outside of the Bragg point to orientations inside the Bragg point (this can be seen from Table 1 of Buxton et al. (1). This is precisely the information that is lost in the on-axis pattern. In short, the symmetries of the on-axis pattern are just the symmetries predicted in the tables of Buxton et al. (1) - even when the modified symmetries of the HOLZ reflections are taken into account.

We can elaborate this point a bit further. As stated above, a disc in the zero layer covers a range of angles that lie outside the Bragg angle. In contrast, discs corresponding to reflections in higher-order Laue zones include the Bragg angle for the reflection. Therefore such reflections should display the full symmetry of the diffraction group for the reflection. However the symmetries that relate orientations outside the Bragg angle to orientations inside the Bragg angle are just those symmetries that arise from inverting symmetry operations, those of Table 1, and therefore they do not give rise to symmetries in HOLZ reflections. Thus, in an *on-axis pattern*, the symmetries *observed* are the same in all reflections whether in the zero layer or HOLZ.

Conclusions

The tables of Buxton et al. (1), give *inter alia* the symmetries, about the Bragg point, that are to be expected for diffracted beams in zone-axis diffraction patterns. The results, as given, apply to the zero layer reflections - not to reflections in higher-order Laue zones. Table 1 here list the symmetry operations that give rise to symmetries in and between the zero layer reflections but which do not introduce symmetries in the higher-order reflections. (Symmetries are introduced between reflections in Laue zones of positive and negative order, but these would not be observed in normal microscopy.) Table 2 shows how this affects all the 31 diffraction groups. Because of the geometry of on-axis convergent-beam patterns, the symmetries of on-axis patterns are not affected.

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