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Surface Superlattice Reflections and Kinematical Approximation in RHEED

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Abstract

The physical nature of the sharpness and weakness of surface superlattice spots in reflection high-energy electron diffraction (RHEED) and the validity of the kinematical approximation for analyzing the intensities of the superlattice spots are examined, using a Bloch-wave formulation of the dynamical theory of RHEED. It is found that although it is adequate to treat surface superlattice diffraction kinematically within the selvedge, a kinematical analysis of RHEED intensities of superlattice spots is not in general valid, unless some criteria are satisfied. These include a projection approximation for the superlattice in the selvedge, a glancing incidence such that no diffracted beams other than the incident and specularly reflected beams are excited in both the selvedge and the underlying bulk crystal.

1. Introduction

In recent years there has been a trend in the structure determination of reconstructed surfaces to use high-energy electron diffraction techniques, and to employ a kinematical approximation in analyzing the intensities of surface superlattice reflections (Takayanagi, Tanishiro, Takahashi & Takahashi, 1985*a*, *b*; Ino, 1977; Wu & Schowalter, 1988; Horio & Ichimiya, 1989). Theoretically this trend results from the intractability of carrying out dynamical calculations for large numbers of possible surface models and experimentally it is motivated by the great success of the determination of the dimer adatom stacking-fault (DAS) structure of the Si(111) 7×7 reconstructed surface using a simple kinematical analysis (Takayanagi *et al.*, 1985*a*, *b*).

While in the case of transmission electron diffraction (TED) the validity of the kinematical approximation has been examined using multislice calculations (Spence, 1983; Tanishiro & Takayanagi, 1989) for Si and certain incident-beam directions and analyzed using Bloch-wave theory (Peng & Whelan, 1991), far less has been done in the reflection high-energy electron diffraction (RHEED) case. Although it has long been realized that in RHEED electrons interact strongly with the atoms and dynamical calculations are needed, the extreme sharpness and weakness of the surface superlattice spots in RHEED patterns have been taken to suggest the validity of a kinematical description, *i.e.* the diffraction processes undergone by electrons associated with superlattice reflections are dominated by single-scattering processes. The intensities of superlattice spots in RHEED have therefore been analyzed kinematically (Ino, 1977; Wu & Schowalter, 1988; Horio & Ichimiya, 1989). It is the purpose of this paper to examine the various diffraction processes involved in both the selvedge and the underlying bulk crystal, within the framework of Bloch-wave dynamical theory, and to set criteria for the validity for the kinematical approximation in analyzing RHEED intensities of superlattice reflections.

2. General description

We shall consider a beam of high-energy electrons (10 keV or more, say) which is incident upon a flat surface at grazing incidence. Here the surface is thought of as consisting of a selvedge, over which there is a reconstruction, and the underlying bulk crystal. There are three distinct regions which need to be treated separately. For the vacuum region above the surface, there exist the incident electron beam and several reflected beams, and the total wavefunction may be written as

$$\psi_v(\mathbf{r}) = \exp(i\boldsymbol{\chi}.\mathbf{r}) + \sum_m R_m \exp(i\mathbf{K}_m.\mathbf{r}), \quad (1)$$

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where $\boldsymbol{\chi}$ is the incident-beam wavevector, and $\mathbf{K}_m$ and R_m are the reflected-beam wavevector and amplitude, respectively, in vacuum. Here we have taken a set of two-dimensional (2D) reciprocal-lattice vectors $\{\mathbf{m}\}$ as the basis of a plane-wave expansion in (1). The reflected wavevectors are given by

$$\mathbf{K}_m = (\boldsymbol{\chi}_\rho + \mathbf{m}) + \mathbf{n}[\chi^2 - (\boldsymbol{\chi}_\rho + \mathbf{m})^2]^{1/2},$$

where the unit vector $\mathbf{n}$ is the outward surface normal of the crystal and the subscript ρ denotes the surface parallel component.

Within the selvedge many diffracted beams are excited. The electron wavefunction can be written as a sum of Bloch waves $b^{(i)}(\mathbf{k}^{(i)}, \mathbf{r})$ with amplitude $\alpha^{(i)}$:

$$\begin{aligned}\psi_s(\mathbf{r}) &= \sum_i \alpha^{(i)} b^{(i)}(\mathbf{k}^{(i)}, \mathbf{r}) \\ &= \sum_i \alpha^{(i)} \sum_g C_g^{(i)} \exp[i(\mathbf{k}^{(i)} + \mathbf{g}).\mathbf{r}], \qquad (2)\end{aligned}$$

in which the wavevector $\mathbf{k}^{(i)}$ and Bloch-wave coefficients $C_g^{(i)}$ satisfy the fundamental equation of electron diffraction

$$[K_0^2 + U_0 - (\mathbf{k}+\mathbf{g})^2]C_g + \sum_{h \neq g} U_{g-h} C_h = 0, \qquad (3)$$

for an electron moving in a periodic potential

$$V(\mathbf{r}) = (2m|e|/\hbar^2) \sum_g U_g \exp(i\mathbf{g}.\mathbf{r}),$$

where m, e and $\hbar$ have their usual physical meaning and the vector $\mathbf{g}$ denotes a reciprocal-lattice vector of the periodic structure. For our purpose of discussing surface superlattice reflections it is appropriate to discriminate in the set of all vectors $\mathbf{g}$ two subsets of reflections $\{\mathbf{b}\}$ and $\{\mathbf{s}\}$ which contain the fundamental bulk (b) and surface (s) superlattice reciprocal-lattice vectors, respectively. If we consider a case where p distinct superlattice reciprocal-lattice rods and n distinct fundamental reciprocal-lattice rods are involved, there are then $2(n+p)$ independent Bloch waves, and (2) then becomes

$$\begin{aligned}\psi_s(\mathbf{r}) = \sum_{i=1}^{2(n+p)} \alpha^{(i)} \Big\{ &\sum_b C_b^{(i)} \exp(i\mathbf{k}_b^{(i)}.\mathbf{r}) \\ &+ \sum_s C_s^{(i)} \exp(i\mathbf{k}_s^{(i)}.\mathbf{r}) \Big\}. \qquad (4)\end{aligned}$$

In the underlying bulk crystal, the counterpart of (3) can be written in matrix notation, noticing that we always have $U_{b_i - s_j} = 0$ in the bulk:

$$\begin{pmatrix} \{M_b\} & \{0\} \\ \{0\} & \{M_s\} \end{pmatrix} \begin{pmatrix} \{C_b\} \\ \{C_s\} \end{pmatrix} = 0, \qquad (5)$$

in which

$$\mathbf{M}_b = \begin{pmatrix} K_0^2 + U_0 - k_0^2 & U_{-b_1} & \dots & U_{-b_n} \\ U_{b_1} & K_0^2 + U_0 - k_{b_1}^2 & \dots & U_{b_1 - b_n} \\ \vdots & \vdots & \vdots & \vdots \\ U_{b_n} & U_{b_n - b_1} & \dots & K_0^2 + U_0 - k_{b_n}^2 \end{pmatrix}$$

$$\mathbf{M}_s = \begin{pmatrix} K_0^2 + U_0 - k_{s_1}^2 & U_{s_1 - s_2} & \dots & U_{s_1 - s_p} \\ U_{s_2} & K_0^2 + U_0 - k_{s_2}^2 & \dots & U_{s_2 - s_p} \\ \vdots & \vdots & \vdots & \vdots \\ U_{s_p} & U_{s_p - s_2} & \dots & K_0^2 + U_0 - k_{s_p}^2 \end{pmatrix}.$$

For a non-trivial solution the determinant of the matrix in (5) must be zero, giving $2(n+p)$ distinct eigenvectors $\mathbf{C}$

$$\mathbf{C}^{(i)} = \begin{pmatrix} \{C_b^{(i)}\}_i \\ \mathbf{0} \end{pmatrix} \quad (i = 1, \dots, 2n)$$

$$\mathbf{C}^{(j)} = \begin{pmatrix} \mathbf{0} \\ \{C_s^{(j)}\} \end{pmatrix} \quad [j = 2n+1, \dots, 2(n+p)].$$

The situation is similar to that in the Laue transmission case, as discussed by Peng & Whelan (1991), *i.e.* within the bulk crystal there exist two groups of Bloch waves. The first group $\{C_b^{(i)}\}$ is associated with bulk reciprocal-lattice vectors $\{\mathbf{b}\}$ only, resulting from the following equation:

$$\mathbf{M}_b \mathbf{C}_b = 0, \qquad (6)$$

and the second group $\{C_s^{(j)}\}$ is associated with surface superlattice reciprocal-lattice vectors $\{\mathbf{s}\}$, satisfying

$$\mathbf{M}_s \mathbf{C}_s = 0. \qquad (7)$$

The total wave function within the bulk is given by

$$\begin{aligned}\psi_b(\mathbf{r}) = &\sum_{i=1}^{2n} \alpha^{(i)} \sum_b C_b'^{(i)} \exp(i\mathbf{k}_b'^{(i)}.\mathbf{r}) \\ &+ \sum_{j=2n+1}^{2(n+p)} \alpha^{(j)} \sum_s C_s'^{(j)} \exp(i\mathbf{k}_s'^{(j)}.\mathbf{r}). \qquad (8)\end{aligned}$$

Here we have used a prime to distinguish bulk quantities from those of the selvedge. Excitations of Bloch waves within the selvedge are determined by the incident beam, as well as by the boundary conditions on the upper face of the selvedge and the selvedge/bulk interface. Cross coupling, *i.e.* coupling between the two sets of reflections $\{\mathbf{b}\}$ and $\{\mathbf{s}\}$, is allowed within the selvedge. Current may therefore be transferred from incident and fundamental beams to the superlattice reflections. Inside the bulk crystal the two groups of Bloch waves associated with $\{\mathbf{b}\}$ and $\{\mathbf{s}\}$ are completely uncoupled, electrons in one diffracted beam may be dynamically scattered into some other beams, but only within their own group, that is either within fundamental beams or within surface superlattice associated beams. Current cannot be transferred from incident and fundamental beams to surface superlattice diffracted beams in the bulk crystal. All exchange processes of current between fundamental reflections and superlattice reflections must occur within the selvedge.

3. Kinematical and dynamical diffraction in the selvedge

Formally, the scattering by the selvedge should be treated using the fundamental equations (3). However, in many cases the experimentally observed superlattice spots in RHEED are very sharp and weak in comparison with the fundamental ones. This is indeed suggestive of weak scattering by the surface superlattice, *i.e.* the current transfer from the incident and strongly diffracted fundamental beams to a surface superlattice beam is small. It is therefore not unreasonable to treat the scattering of the superlattice within the selvedge kinematically, even though it has been commonly believed that, owing to the small glancing angle of incidence, electrons might have travelled far enough within the selvedge to be scattered dynamically.

Take the Si(111) 7×7 reconstructed surface as an example. For a typical experimental arrangement of 30 keV electrons and incident glancing angle (θ) from 1 to 3°, we may estimate the electron path length within a selvedge of the Si(111) surface to be $(a_0/3^{1/2})/\sin\theta = 65\text{–}200$ Å. Since values in this range are comparable to the Bragg two-beam extinction distance of 360 Å (along the incident-beam direction) for a fundamental 111 reflection and 30 keV primary-beam energy (Whelan, 1970), the fundamental lattice diffraction therefore needs to be treated dynamically in the selvedge. On the other hand, since the structure factors of surface superlattice reflections are typically less than 1% of the value for a fundamental reflection (see, for example, Wu & Schowalter, 1988), the effective extinction distance along the incident-beam direction may therefore be as much as 36 000 Å for superlattice reflections. We can then neglect dynamical effects for those superlattice reflections and write the diffracted-beam amplitude associated with superlattice reflection **s** as

$$\begin{aligned}\varphi_s(\mathbf{r}) &\propto \sum_b f_{s-b}\left[\sum_i \alpha^{(i)} C_b^{(i)} \exp(i\mathbf{k}_b^{(i)}.\mathbf{r})\right] \\ &\quad \times \exp[i(\mathbf{s}-\mathbf{b}).\mathbf{r}] \\ &= \sum_b f_{s-b} \exp[i\mathbf{s}.\mathbf{r}] \\ &\quad \times \left[\sum_i \alpha^{(i)} C_b^{(i)} \exp(i\mathbf{k}^{(i)}.\mathbf{r})\right], \qquad (9)\end{aligned}$$

in which f_{s-b} is the kinematical amplitude of superlattice diffraction from a fundamental reflection **b** to a superlattice reflection **s**, and $\sum \alpha^{(i)} C_b^{(i)} \exp(i\mathbf{k}_b^{(i)}.\mathbf{r})$ are the dynamically diffracted fundamental-beam amplitudes. Both the incident and diffracted fundamental beams can contribute to the superlattice reflections {s} *via* kinematical processes as schematically shown in Fig. 1. For a selvedge having moderate thickness (from a few to ten ångströms), the contribution from diffracted fundamental beams may be comparable to that from the incident beam.

4. Dynamical diffraction and reflection in the bulk and at the selvedge/bulk interface

In general, the diffraction processes in the bulk crystal are dynamically dominating, and the total wavefunction is given by (8). The dynamical diffraction processes are, however, completely uncoupled for the two sets of reflections {**b**} and {**s**}, *i.e.* current can be transferred either between the incident and diffracted fundamental beams or between the selvedge superlattice diffracted beams within the bulk crystal, but not from the fundamental beams to the superlattice

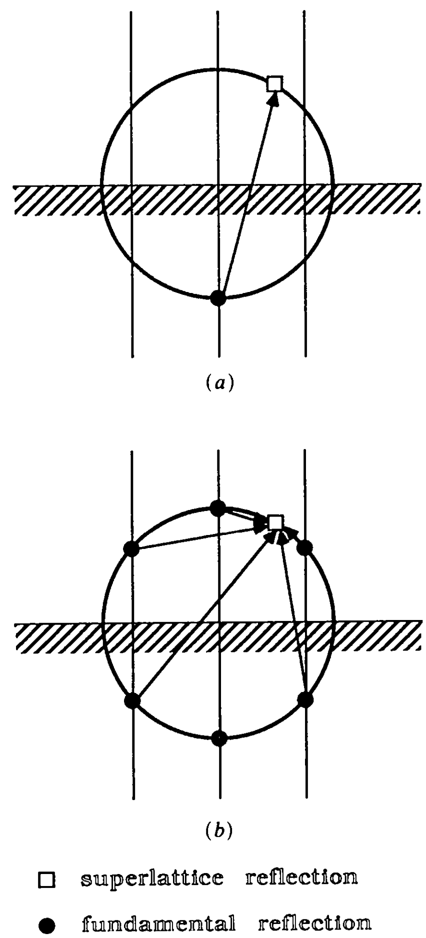

Fig. 1. Schematic diagrams of (*a*) a kinematical diffraction process of the incident beam by the selvedge superlattice; (*b*) dynamical diffraction processes of the incident beam by the fundamental selvedge lattice, followed by kinematical diffraction by the selvedge superlattice. The circle represents the intersection of the Ewald sphere with the zero-order Laue zone and the hatched boundary represents the shadow edge in the RHEED pattern.

reflections or *vice versa*. Using a kinematical approximation to the superlattice diffraction in the selvedge, we may distinguish two distinct contributions to the superlattice reflections in RHEED.

Firstly, the incident and diffracted fundamental beams may undergo dynamical diffraction within the bulk and reflection at the selvedge/bulk interface. The specular and other reflected fundamental beams may then be subject to a kinematical diffraction process in the selvedge and contribute either directly to the superlattice spots in RHEED, or to a secondary incident beam as schematically shown in Fig. 2. Secondly, for the latter case, this secondary incident beam, together with other selvedge superlattice diffracted beams, will then be dynamically diffracted within the bulk and reflected at the selvedge/bulk interface as is the incident or a fundamental diffracted beam, giving contributions to the superlattice reflections in RHEED patterns. The diffraction and reflection processes are sketched in Fig. 3.

5. Kinematical approximation in RHEED analysis of superlattice reflections

We have shown in the previous sections (2–4) that although the surface superlattice diffraction within the selvedge is kinematical, this does not necessarily lead to the validity of the kinematical analysis of the intensities of RHEED superlattice spots. In general, a surface superlattice diffracted beam may be subject to contributions from four types of diffraction processes as schematically shown in Figs. 1 to 3, *i.e.* (1) kinematical diffraction of the incident beam by the superlattice (Fig. 1*a*); (2) dynamical diffraction of the incident beam by the selvedge fundamental periodicity, followed by kinematical diffraction by the selvedge superlattice (Fig. 1*b*); (3) dynamical diffraction and reflection of the incident and dynamical diffracted selvedge fundamental beams in the bulk and at the bulk/selvedge interface, followed by kinematical diffraction by the selvedge superlattice (Figs. 2*a*–*c*); and (4) dynamical diffraction and reflection of the selvedge kinematically diffracted beams and the secondary incident beams resulting from type (3) processes (Figs. 2*d* and 3).

Strictly speaking, a kinematical analysis of RHEED intensities of superlattice spots will be valid only if processes of type (1) were involved. But since in RHEED it is impossible to avoid diffraction processes of types (3) and (4), although type (2) can be avoided, a kinematical approximation to the intensities of RHEED superlattice spots is inadequate in general. However, in cases like those shown in Fig. 4, where there exist no fundamental beams other than the incident and specular reflected beam, we may approximate the superlattice reflected-beam amplitude as (see Appendix)

$$f_{s_1}+\frac{K_{z_1}-k_{z_1}}{K_{z_1}+k_{z_1}}f_{s_2}+\frac{K_{z_2}-k_{z_2}}{K_{z_2}+k_{z_2}}f_{s_3}, \qquad (10)$$

in which K_z are the surface normal components of the wavevectors in vacuum, k_z are those in the crystal, and the subscripts 1 and 2 refer to the reciprocal-

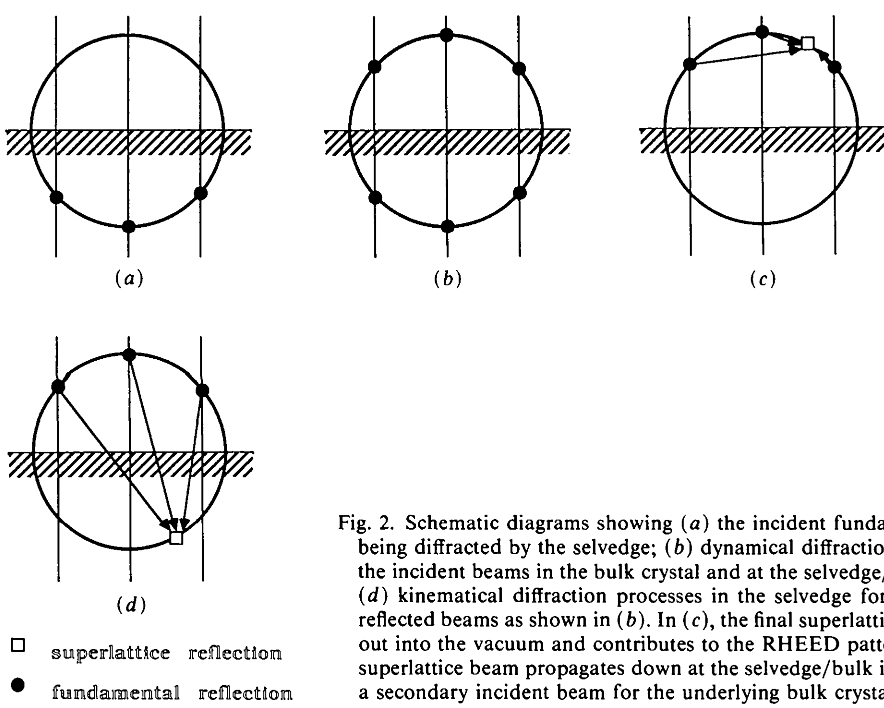

Fig. 2. Schematic diagrams showing (*a*) the incident fundamental beams after being diffracted by the selvedge; (*b*) dynamical diffraction and reflections of the incident beams in the bulk crystal and at the selvedge/bulk interface; (*c*), (*d*) kinematical diffraction processes in the selvedge for those dynamically reflected beams as shown in (*b*). In (*c*), the final superlattice beam propagates out into the vacuum and contributes to the RHEED pattern. In (*d*) the final superlattice beam propagates down at the selvedge/bulk interface and acts as a secondary incident beam for the underlying bulk crystal.

lattice rods containing the specular reflection spot and surface superlattice spot, respectively. In our case we have

$$k_z^2 = \chi_z^2 + U_0. \quad (11)$$

If we go further and use a projection approximation for the selvedge such that $f_{s_1} \simeq f_{s_2} \simeq f_{s_3}$, the superlattice reflected-beam amplitude is therefore directly proportional to the kinematical value. In (11), since the crystal inner potential U_0 is typically 0·1–0·2 Å^{-2} (10–20 eV), and for high-energy electrons (for example 30 keV) which are incident on the surface at less than 20 mrad, K_z is usually less than 0·07 Å^{-1} and k_z is larger than 0·4 Å^{-1}, the reflectivities are near unity for all reflections. When account is taken of the dynamical interactions with other beams, detailed numerical calculations (Peng & Whelan, 1990) show that for most semiconductor surfaces the reflectivity is still nearly unity for glancing incident angles less than about 20 mrad. This is not true in general, however. For example, for the Au(001) surface, the reflectivity decreases rapidly as the incident glancing angle increases, even though the angle is only about a few mrad. A kinematical analysis can then be performed for the RHEED superlattice spot intensities. It is worth mentioning that the criteria just outlined were not satisfied in Wu & Schowalter's (1988) analysis. It is therefore not surprising that they encountered some difficulties in fitting their results to those determined experimentally and the model given by the energy-minimization calculation of Qian & Chadi (1987). On the other hand, in the analysis of Horio & Ichimiya (1989), our criteria are met quite well from their Figs. 1(a) to (e). In their Fig. 1(f), since two fundamental reflections $(1, \bar{1})$ and $(\bar{1}, 1)$ are just emerging from the surface shadow edge, some errors might have been introduced. Some fundamental reflections are also seen in higher-order Laue zones (HOLZ) of the RHEED patterns of Horio & Ichimiya. Since for HOLZ reflections large diffraction vectors $\{\mathbf{s}-\mathbf{b}\}$ are involved, the corresponding structure factors $\{f_{s-b}\}$ are therefore very small. Furthermore, the fundamental HOLZ reflections are much weaker in comparison with the incident and specular reflected beams than is the case for the zero-order Laue zone (ZOLZ), so that the HOLZ effects are negligible for the analysis of low-order RHEED superlattice spots in the ZOLZ. It should be noted that, in the HOLZ rings, important bulk reflections may be excited and coupled with surface superlattice

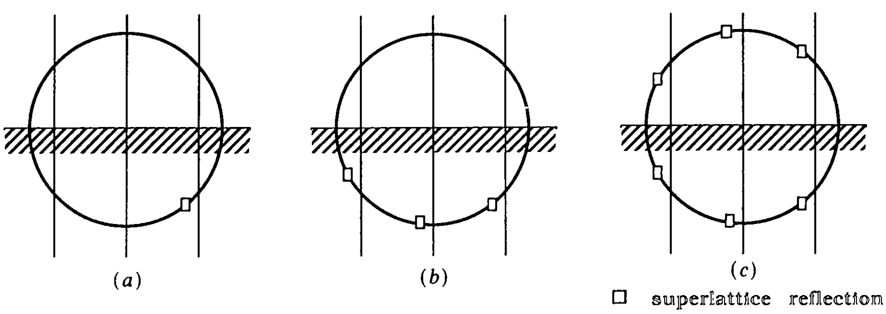

Fig. 3. Diffraction diagrams showing (a) a secondary incident beam resulting from the diffraction processes as shown in Fig. 2(d); (b) kinematically diffracted superlattice reflections after transmission through the selvedge; (c) diffracted and reflected beams after the incident beams shown in (a) and (b) have been dynamically diffracted and reflected in the bulk and at the selvedge/bulk interface.

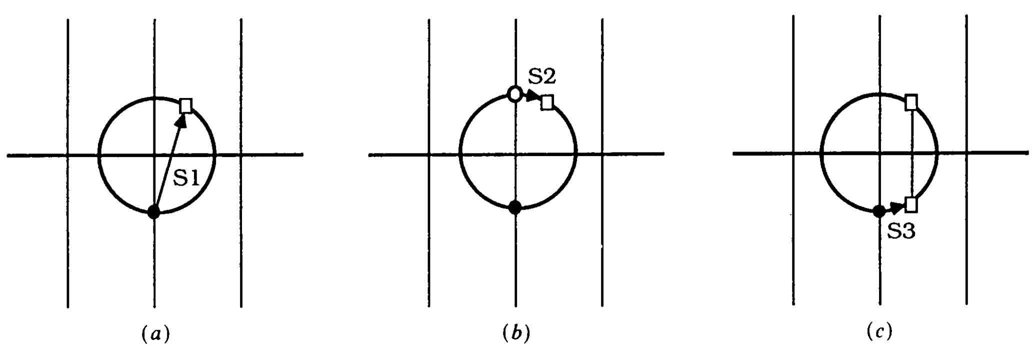

Fig. 4. Diffraction and reflection diagrams for a one-beam case showing different contributions to the superlattice reflection in RHEED patterns. In (a) the incident beam is kinematically diffracted by the selvedge superlattice; (b) the incident beam is dynamically reflected at the selvedge/bulk interface, followed by a kinematical diffraction process by the selvedge superlattice; (c) a kinematically diffracted superlattice beam is dynamically reflected by the selvedge/bulk interface.

reflections in the HOLZ rings. The kinematic approximation cannot therefore be applied in general to those surface superlattice reflections in the HOLZ rings.

6. Concluding remarks

In summary, a Bloch-wave analysis has been made of the dynamical and kinematical diffraction processes from the selvedge fundamental lattice and superlattice, respectively, and of the dynamical diffraction and reflection processes in the underlying bulk crystal and at the selvedge/bulk interface. We have shown that although a kinematical approximation is adequate for treating the superlattice diffraction within the selvedge, a kinematical approximation to the intensities of RHEED superlattice spots does not hold in general. However, for cases where a projection approximation can be made to the selvedge and where the incident-beam direction has been selected so that no strong fundamental reflections other than the specular reflection are strongly excited, we have also shown that the superlattice reflected-beam amplitudes are directly proportional to their corresponding kinematical values. Furthermore, since for glancing incidence the proportionality factors for all low-order superlattice reflections are approximately the same, an analysis of RHEED intensities of superlattice spots based on their kinematical values of diffracted-beam amplitudes is valid. To a first-order approximation, the fundamental HOLZ effects are negligible for analyzing low-order superlattice spot intensities.

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APPENDIX

We consider here a case where high-energy electrons are incident on the crystal surface at such a small glancing angle that beside the incident and specular reflected beam there exist no other beams in the vacuum region above the surface. The total wave function then takes the form

$$\psi_v(\mathbf{r}) = \exp(i\boldsymbol{\chi}.\mathbf{r}) + R\exp(i\mathbf{K}.\mathbf{r}),$$

where $\boldsymbol{\chi}$ and $\mathbf{K}$ are the incident and specular reflected wavevectors. In general we have $\boldsymbol{\chi}_\rho = \mathbf{K}_\rho$ and $\chi_z = -K_z$ and R is the specular reflected-beam amplitude. Within the crystal, to a first-order approximation (by neglecting the dynamical effects resulting from crystal periodicity), we may write the total wavefunction as

$$\psi_c(\mathbf{r}) = t\exp(i\mathbf{k}.\mathbf{r}) + r\exp(i\mathbf{k}'.\mathbf{r}),$$

in which t and r are constants, denoting the transmitted- and reflected-beam amplitudes, respectively, and the surface parallel and normal components of the wavevectors satisfy

$$\mathbf{k}_\rho = \mathbf{k}'_\rho = \boldsymbol{\chi}_\rho$$

$$k_z = -k'_z$$

and

$$k_z^2 = k_z'^2 = \chi_z^2 + U_0.$$

In the above expression of the wavefunction within the crystal, the first term is a continuation of the incident vacuum wave in the crystal and the second term results from reflection from the bottom face of a crystal slab. In RHEED, since we only consider a semi-infinite crystal, the bottom face is a trivial one and the reflected wave from the face can therefore be discarded. By applying the boundary conditions to the electron wave function and its surface normal derivative on the surface

$$1 + R = t$$

$$K_z(1 - R) = tk_z$$

and eliminating t, we obtain the reflected-beam amplitude

$$R = (K_z - k_z)/(K_z + k_z).$$

We now consider the case shown in Fig. 4. The surface superlattice spot in the RHEED pattern is subject to three major contributions: (a) $f_{s_1}\exp(i\mathbf{K}.\mathbf{r})$, kinematic reflection diffraction of the incident beam by a selvedge superlattice reflection; (b) $f_{s_2}[R_1\exp(i\mathbf{K}.\mathbf{r})]$, dynamical reflection diffraction of the incident beam, followed by kinematic diffraction by a selvedge superlattice reflection; and (c) $R_2[f_{s_3}\exp(i\mathbf{K}.\mathbf{r})]$, dynamical reflection diffraction at the surface, following kinematic forward diffraction within the selvedge by a superlattice reflection. The total superlattice reflected-beam amplitude is therefore proportional to

$$f_{s_1} + R_1 f_{s_2} + R_2 f_{s_3}$$

or

$$f_{s_1} + \frac{K_{z_1} - k_{z_1}}{K_{z_1} + k_{z_1}} f_{s_2} + \frac{K_{z_2} - k_{z_2}}{K_{z_2} + k_{z_2}} f_{s_3}.$$

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On the Uncoupling of Surface Superlattice Reflections in TED Analysis of Reconstructed Surfaces

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Abstract

A Bloch-wave analysis is made of the problem of uncoupling surface superlattice reflections from fundamental reflections in transmission electron diffraction (TED) analysis of reconstructed surfaces. This uncoupling problem is proved to be of crucial importance in determining the structure of reconstructed surfaces, for example the Si(111) 7×7 surface [Takayanagi, Tanishiro, Takahashi & Takahashi (1985). *Vac. Sci. Technol.* A3, 1502-1506; (1985). *Surf. Sci.* **164**, 367-392]. It is found that a complete uncoupling, weak coupling and sometimes strong coupling between the bulk scattering and surface superlattice scattering are all possible depending on the diffraction conditions. For a kinematical analysis of reconstructed surfaces to be valid, a weak coupling or a complete uncoupling condition must be realized. General rules for choosing the appropriate diffraction conditions are given.

1. Introduction

Transmission electron diffraction (TED) has proved to be one of the most powerful techniques for structure determination of reconstructed surfaces. One outstanding example of its great power is the derivation of the dimer adatom stacking-fault (DAS) structure of the Si(111) 7×7 reconstructed surface by Takayanagi and his associates (Takayanagi *et al.*, 1985*a*, *b*). In their experiments, several TED patterns were taken from a thin area of the specimen (about 30 nm or less in thickness). The TED intensities were then averaged among the equivalent reflections related by hexagonal symmetry. It was then postulated that, after averaging, the intensities of the surface superlattice reflections will retain their kinematical values after passing through the underlying bulk crystal. The intensities of the superlattice reflections were thus analyzed using kinematical (single-scattering) theory.

The validity of the kinematical approximation has been examined theoretically using dynamical multislice calculations (Spence, 1983; Tanishiro & Takayanagi, 1989). A reliability factor of 10% was declared, given that the incident beam is properly tilted so that a minimum number of strong bulk reflections are excited (Tanishiro & Takayanagi, 1989). However, the examinations of the validity of the kinematical approximation using multislice calculations were made only for Si and for some specific incident-beam conditions. Detailed considerations of the coupling and uncoupling of surface-superlattice-related scattering with bulk scattering still remain to be given.

In principle, a more convenient theoretical framework for analyzing the problem of the coupling and uncoupling of the strong dynamical bulk scattering and the weaker surface superlattice scattering is the Bloch-wave method of Bethe (1928), because the scattering by either the surface layer or the underlying bulk crystal can be represented by a characteristic scattering matrix. The coupling between the surface-layer scattering and bulk scattering can then be understood by analyzing only the structure of the scattering matrices. It is the purpose of this paper to present such an analysis. It will be shown that uncoupling or weaker coupling of the surface superlattice diffracted beams and the underlying bulk-crystal scattering cannot be taken for granted, even for cases in which only superlattice reflections are excited. General rules for choosing the incident-beam conditions to avoid strong coupling are given.

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