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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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2. General theory

For convenience, it is assumed that the specimen is composed of plane parallel crystal slabs with the z axis normal to the surface. When the usual projected potential approximation is applied we can use the general matrix formulations given by Howie & Whelan (1961) and Sturkey (1962). Following Bethe (1928) we write the electron wavefunction within the crystal as

$$\begin{aligned}\psi(\mathbf{r}) &= \sum_j \alpha^{(j)} b^{(j)}(\mathbf{k}^{(j)}, \mathbf{r}) \\ &= \sum_j \alpha^{(j)} \sum_g C_g^{(j)} \exp(i\mathbf{k}_g^{(j)} \cdot \mathbf{r}),\end{aligned} \quad (1)$$

where $b^{(j)}(\mathbf{k}^{(j)}, \mathbf{r})$ is the jth Bloch wave, $\alpha^{(j)}$ is the excitation amplitude, $\mathbf{g}$ denotes a reciprocal-lattice vector, $\mathbf{k}_g^{(j)}$ is the wavevector ($=\mathbf{k}^{(j)}+\mathbf{g}$) and $C_g^{(j)}$ the associated plane-wave coefficients. The values of $\mathbf{k}_g^{(j)}$ and $C_g^{(j)}$ are determined by solving the fundamental equation

$$(K^2 - k_g^2)C_g + \sum_{h \neq g} U_{g-h} C_h = 0, \quad (2)$$

or the equivalent eigenvalue equation (valid at high energy for small-angle forward scattering)

$$2K(s_g - \gamma)C_g + \sum_{h \neq g} U_{g-h} C_h = 0, \quad (3)$$

in which U_g are structure factors (in Å^{-2}), s_g are excitation errors, $\mathbf{K}$ is the incident wavevector after correction for the mean inner potential and γ the conventional *Anpassung* defined as

$$\mathbf{k}_g^{(j)} = \mathbf{K} + \gamma^{(j)}\mathbf{n} + \mathbf{g}, \quad (4)$$

where $\mathbf{n}$ has been chosen as pointing into the crystal along the surface-normal direction.

According to Howie & Whelan (1961), the amplitude vectors $\mathbf{u}(0) = \{\varphi_g(0)\}$ and $\mathbf{u}(t_s) = \{\varphi_g(t_s)\}$ on the upper and lower surfaces of a crystal slab of thickness t_s are related by a scattering matrix $\mathbf{P}$:

$$\mathbf{u}(t_s) = \mathbf{P}(t_s)\mathbf{u}(0), \quad (5)$$

where

$$\mathbf{P}(t) = \mathbf{C}\mathbf{Y}\mathbf{C}^{-1} \quad (6)$$

and

$$\mathbf{C} = \begin{pmatrix} C_{g_1}^{(1)} & C_{g_1}^{(2)} & \dots & C_{g_1}^{(N)} \\ C_{g_2}^{(1)} & C_{g_2}^{(2)} & \dots & C_{g_2}^{(N)} \\ \dots & \dots & \dots & \dots \\ C_{g_N}^{(1)} & C_{g_N}^{(2)} & \dots & C_{g_N}^{(N)} \end{pmatrix},$$

$$\mathbf{Y} = \begin{pmatrix} \exp(i\gamma^{(1)}t) & 0 & \dots & 0 \\ 0 & \exp(i\gamma^{(2)}t) & \dots & 0 \\ \dots & \dots & \dots & \dots \\ 0 & 0 & \dots & \exp(i\gamma^{(N)}t) \end{pmatrix},$$

$$C^{-1} = \begin{pmatrix} C_{g_1}^{*(1)} & C_{g_2}^{*(1)} & \dots & C_{g_N}^{*(1)} \\ C_{g_1}^{*(2)} & C_{g_2}^{*(2)} & \dots & C_{g_N}^{*(2)} \\ \dots & \dots & \dots & \dots \\ C_{g_1}^{*(N)} & C_{g_2}^{*(N)} & \dots & C_{g_N}^{*(N)} \end{pmatrix}.$$

The scattering matrix $\mathbf{P}$ for transmission through an assembly of crystal slabs is given by the product of the scattering matrices for each slab

$$\mathbf{P} = \mathbf{P}_N \dots \mathbf{P}_2\mathbf{P}_1. \quad (7)$$

Extension has also been made to include higher-order Laue-zone (HOLZ) effects. For details see Peng & Whelan (1990).

It should be noted that the application of the general equation (7) is not limited by the thicknesses of the crystal slabs involved. For example, although the surface layers of a real crystal are typically a monolayer or two in thickness, giving reciprocal-lattice rods rather than points under the kinematical approximation, the excitations of Bloch waves and diffracted beams within the surface layers as well as in the underlying bulk crystal are not affected by the finite thickness of the surface layers. It is an important corollary of this fact that although all reflections leaving the surface layers will have their beam amplitudes not very different from the corresponding kinematic values, regardless of whether or not the corresponding reciprocal-lattice points are on or near the Ewald sphere, only those reflections which are strongly excited (with g points on or near the Ewald sphere) can couple with each other in passing through the bulk crystal and need to be treated as 'strong beams', despite the fact that they may be as weak as other surface superlattice reflections which have not been strongly excited.

3. Some special cases

3.1. *Three-beam uncoupling case*

We now consider a slab system consisting of a reconstructed surface layer on the upper face of the underlying bulk-crystal slab. The surface layer is assumed to be of thickness t_s within which two diffracted beams, associated with fundamental reflection $\mathbf{g}$ and surface superlattice reflection $\mathbf{h}$, respectively, are strongly excited as in Fig. 1. There exist three distinct eigenvalues $\gamma^{(j)}$ ($j = 1, 2, 3$) and thus three eigenvectors within the surface layer:

$$\mathbf{C}^{(j)} = (C_0^{(j)}, C_g^{(j)}, C_h^{(j)}) \; (j = 1, 2, 3).$$

Omitting the brackets from superscripts, one obtains the scattering matrix associated with this surface layer as

$$\mathbf{P}_s=\begin{pmatrix} C_0^1 & C_0^2 & C_0^3 \\ C_g^1 & C_g^2 & C_g^3 \\ C_h^1 & C_h^2 & C_h^3 \end{pmatrix}\begin{pmatrix} \exp(i\gamma^1 t_s) & 0 & 0 \\ 0 & \exp(i\gamma^2 t_s) & 0 \\ 0 & 0 & \exp(i\gamma^2 t_s) \end{pmatrix}\times\begin{pmatrix} C_0^{*1} & C_g^{*1} & C_h^{*1} \\ C_0^{*2} & C_g^{*2} & C_h^{*2} \\ C_0^{*3} & C_g^{*3} & C_h^{*3} \end{pmatrix}$$

$$=\begin{pmatrix} \sum_{j=1}^{3} C_0^j C_0^{*j}\exp(i\gamma^j t_s) & \sum_{j=1}^{3} C_0^j C_g^{*j}\exp(i\gamma^j t_s) & \sum_{j=1}^{3} C_0^j C_h^{*j}\exp(i\gamma^j t_s) \\ \sum_{j=1}^{3} C_g^j C_0^{*j}\exp(i\gamma^j t_s) & \sum_{j=1}^{3} C_g^j C_g^{*j}\exp(i\gamma^j t_s) & \sum_{j=1}^{3} C_g^j C_h^{*j}\exp(i\gamma^j t_s) \\ \sum_{j=1}^{3} C_h^j C_0^{*j}\exp(i\gamma^j t_s) & \sum_{j=1}^{3} C_h^j C_g^{*j}\exp(i\gamma^j t_s) & \sum_{j=1}^{3} C_h^j C_h^{*j}\exp(i\gamma^j t_s) \end{pmatrix}. \quad (8)$$

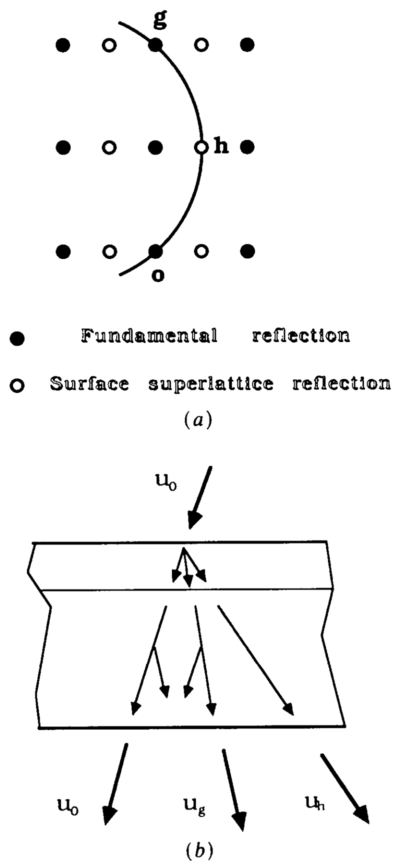

Fig. 1. Schematic diagram showing (*a*) the diffraction configuration, in which the intersection of the Ewald sphere with the zero-order Laue zone is depicted; (*b*) diffraction processes in passing through the surface layer and underlying bulk crystal.

Since the vacuum region above the surface layer contains only the incident beam (backscattered beams can be safely neglected in the Laue case), the amplitude vector $\mathbf{u}(0)$ on the upper face of the surface layer can be written as

$$\mathbf{u}(0)=\begin{pmatrix} \varphi_0(0) \\ \varphi_g(0) \\ \varphi_h(0) \end{pmatrix}=\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}. \quad (9)$$

The amplitude vector $\mathbf{u}(t_s)$ leaving the bottom face of the surface layer is given by

$$\begin{pmatrix} \varphi_0(t_s) \\ \varphi_g(t_s) \\ \varphi_h(t_s) \end{pmatrix}=\mathbf{P}_s\mathbf{u}(0)=\begin{pmatrix} \sum_{j=1}^{3} C_0^j C_0^{*j}\exp(i\gamma^j t_s) \\ \sum_{j=1}^{3} C_g^j C_0^{*j}\exp(i\gamma^j t_s) \\ \sum_{j=1}^{3} C_h^j C_0^{*j}\exp(i\gamma^j t_s) \end{pmatrix}. \quad (10)$$

Now consider the underlying bulk-crystal slab having thickness t_b. Within the underlying bulk-crystal slab, reflection $\mathbf{h}$ is a trivial one, *i.e.* $U_h=0$ in the bulk, since it is associated with a surface superlattice reflection. From (3) the corresponding three-beam eigenvalue equation is of the form

$$\begin{pmatrix} -2K\gamma & U_{-g} & 0 \\ U_g & 2K(s_g-\gamma) & 0 \\ 0 & 0 & 2K(s_h-\gamma) \end{pmatrix}\begin{pmatrix} C_0 \\ C_g \\ C_h \end{pmatrix}=0, \quad (11)$$

which gives rise to the following three eigenvalues:

$$\gamma'^{1,2}=\tfrac{1}{2}\{s_g\pm[s_g^2+(|U_g|/K)^2]^{1/2}\},$$
$$\gamma'^3=s_h,$$

where the first superscript on γ' refers to the negative sign, and the same convention will be used subsequently. The three corresponding eigenvectors are

$$\mathbf{C}^1=(C_0'^1, C_g'^1, C_h'^1)=(\cos\tfrac{1}{2}\beta, -\sin\tfrac{1}{2}\beta, 0),$$
$$\mathbf{C}^2=(C_0'^2, C_g'^2, C_h'^2)=(\sin\tfrac{1}{2}\beta, \cos\tfrac{1}{2}\beta, 0),$$
$$\mathbf{C}^3=(C_0'^3, C_g'^3, C_h'^3)=(0, 0, 1),$$

where the primes denote that all quantities are associated with the bulk crystal, to distinguish them from those of the surface layer, and β is Takagi's deviation parameter defined by (see, for example, Hirsch, Howie, Nicholson, Pashley & Whelan, 1965)

$$\cot\beta=s_g(K/|U_g|). \quad (12)$$

The scattering matrix associated with the bulk-crystal slab of thickness t_b is

$$\mathbf{P}_b=\begin{pmatrix}\cos\frac{1}{2}\beta & \sin\frac{1}{2}\beta & 0\\ -\sin\frac{1}{2}\beta & \cos\frac{1}{2}\beta & 0\\ 0 & 0 & 1\end{pmatrix}$$

$$\times\begin{pmatrix}\exp(i\gamma'^1t_b) & 0 & 0\\ 0 & \exp(i\gamma'^2t_b) & 0\\ 0 & 0 & \exp(i\gamma'^3t_b)\end{pmatrix}$$

$$\times\begin{pmatrix}\cos\frac{1}{2}\beta & -\sin\frac{1}{2}\beta & 0\\ \sin\frac{1}{2}\beta & \cos\frac{1}{2}\beta & 0\\ 0 & 0 & 1\end{pmatrix}$$

$$=\begin{pmatrix}\cos^2\frac{1}{2}\beta\exp(i\gamma'^1t_b)+\sin^2\frac{1}{2}\beta\exp(i\gamma'^2t_b) & \sin\frac{1}{2}\beta\cos\frac{1}{2}\beta[\exp(i\gamma'^2t_b)-\exp(i\gamma'^1t_b)] & 0\\ \sin\frac{1}{2}\beta\cos\frac{1}{2}\beta[\exp(i\gamma'^2t_b)-\exp(i\gamma'^1t_b)] & \sin^2\frac{1}{2}\beta\exp(i\gamma'^1t_b)+\cos^2\frac{1}{2}\beta\exp(i\gamma'^2t_b) & 0\\ 0 & 0 & \exp(i\gamma'^3t_b)\end{pmatrix}.$$

This gives the diffracted-beam amplitude associated with surface superlattice reflection **h**,

$$\varphi_h(t_s+t_b)=\varphi_h(t_s)\exp(i\gamma'^3t_b),\qquad(13)$$

and the intensity,

$$I_h=|\varphi_h(t_s+t_b)|^2=|\varphi_h(t_s)|^2,\qquad(14)$$

which is entirely unaffected by the underlying bulk scattering. A complete uncoupling is achieved in this three-beam case.

3.2. *Four-beam self-coupling case*

In the three-beam case discussed in the previous section, because the Fourier potential coefficient of the surface superlattice reflection **h** and the Fourier coefficient of the interaction potential between the superlattice reflection **h** and fundamental bulk reflection **g** are zero within the underlying bulk slab, the intensity of the superlattice reflection **h** is completely unaffected by the diffraction processes of the underlying bulk-crystal slab. A different situation may occur, however, if the Fourier potential coefficients between the surface superlattice reflections are not negligible. Shown in Fig. 2 are two four-beam self-coupling cases. By self coupling we mean that coupling occurs either among the surface superlattice reflections or among the fundamental bulk reflections.

Within the surface layer, three diffracted beams are strongly excited, a fundamental reflection **g** and two surface-superlattice reflections **h** and **l**. Similar to the derivation leading to (10), we obtain the amplitude vector $\mathbf{u}(t_s)$ leaving the upper reconstructed surface layer

$$\begin{pmatrix}\varphi_0(t_s)\\ \varphi_g(t_s)\\ \varphi_h(t_s)\\ \varphi_l(t_s)\end{pmatrix}=\begin{pmatrix}\sum_{j=1}^{4}C_0^jC_0^{*j}\exp(i\gamma^jt_s)\\ \sum_{j=1}^{4}C_g^jC_0^{*j}\exp(i\gamma^jt_s)\\ \sum_{j=1}^{4}C_h^jC_0^{*j}\exp(i\gamma^jt_s)\\ \sum_{j=1}^{4}C_l^jC_0^{*j}\exp(i\gamma^jt_s)\end{pmatrix}.\qquad(15)$$

Within the underlying bulk-crystal slab, although the interactions between the superlattice reflections **h** and **l** with either the transmitted beam or the diffracted beam resulting from fundamental reflection **g** are zero, the interaction Fourier coefficient between the surface superlattice reflections **h** and **l**, U_{h-l}, is not zero. The surface superlattice diffracted beams can therefore couple with each other strongly in passing through the bulk-crystal slab. The four-beam

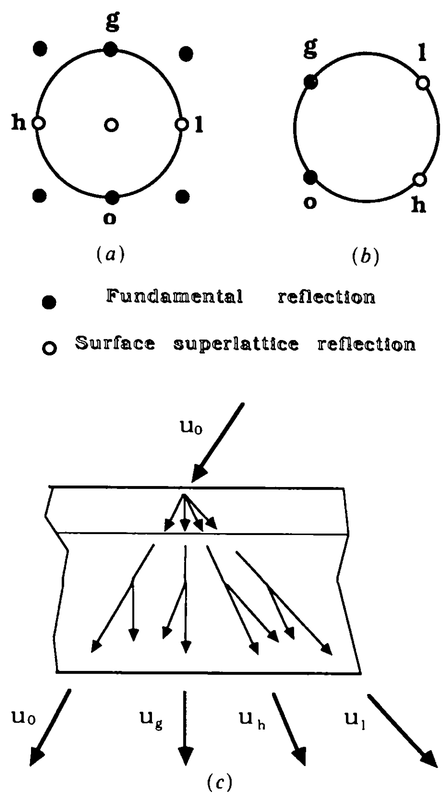

Fig. 2. Two four-beam configurations with (*a*) two symmetric surface superlattice reflections and (*b*) two asymmetric superlattice reflections. Shown in (*c*) is a schematic diagram of the relevant diffraction processes.

eigenvalue equation in this case takes the form

$$\begin{pmatrix} -2K\gamma' & U_{-g} & 0 & 0 \\ U_g & 2K(s_g-\gamma') & 0 & 0 \\ 0 & 0 & 2K(s_h-\gamma') & U_{h-l} \\ 0 & 0 & U_{l-h} & 2K(s_l-\gamma') \end{pmatrix} \times \begin{pmatrix} C'_0 \\ C'_g \\ C'_h \\ C'_l \end{pmatrix} = 0, \quad (16)$$

which gives the following four eigenvalues:

$$\gamma'^{1,2} = \tfrac{1}{2}\{s_g \pm [s_g^2 + (|U_g|/K)^2]^{1/2}\},$$
$$\gamma'^{3,4} = s_h + \tfrac{1}{2}\{(s_l - s_h) \pm [(s_l - s_h)^2 + (|U_{h-l}|/K)^2]^{1/2}\}$$

and the corresponding eigenvectors

$$\mathbf{C}^1 = (\cos\tfrac{1}{2}\beta_1, -\sin\tfrac{1}{2}\beta_1, 0, 0)$$
$$\mathbf{C}^2 = (\sin\tfrac{1}{2}\beta_1, \cos\tfrac{1}{2}\beta_1, 0, 0)$$
$$\mathbf{C}^3 = (0, 0, \cos\tfrac{1}{2}\beta_2, -\sin\tfrac{1}{2}\beta_2)$$
$$\mathbf{C}^4 = (0, 0, \sin\tfrac{1}{2}\beta_2, \cos\tfrac{1}{2}\beta_2),$$

in which β_1 and β_2 are defined by

$$\cot\beta_1 = s_g|U_g|/K,$$
$$\cot\beta_2 = (s_l - s_h)|U_{h-l}|/K.$$

The scattering matrix of the bulk slab can be written as

$$\mathbf{P}_b = \begin{pmatrix} \mathbf{P}_1 & \mathbf{0} \\ \mathbf{0} & \mathbf{P}_2 \end{pmatrix} \quad (17)$$

with

$$\mathbf{P}_1 = \begin{pmatrix} \cos^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^1 t_b) + \sin^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^2 t_b) & \cos\tfrac{1}{2}\beta_1 \sin\tfrac{1}{2}\beta_1 [\exp(i\gamma'^2 t_b) - \exp(i\gamma'^1 t_b)] \\ \sin\tfrac{1}{2}\beta_1 \cos\tfrac{1}{2}\beta_1 [\exp(i\gamma'^2 t_b) - \exp(i\gamma'^1 t_b)] & \sin^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^1 t_b) + \cos^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^2 t_b) \end{pmatrix}$$

and

$$\mathbf{P}_2 = \begin{pmatrix} \cos^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^3 t_b) + \sin^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^4 t_b) & \cos\tfrac{1}{2}\beta_1 \sin\tfrac{1}{2}\beta_1 [\exp(i\gamma'^4 t_b) - \exp(i\gamma'^3 t_b)] \\ \sin\tfrac{1}{2}\beta_1 \cos\tfrac{1}{2}\beta_1 [\exp(i\gamma'^4 t_b) - \exp(i\gamma'^3 t_b)] & \sin^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^3 t_b) + \cos^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^4 t_b) \end{pmatrix} \times \exp(-is_h t_b),$$

where here and subsequently the symbol $\mathbf{0}$ denotes a null matrix of the appropriate order.

The diffracted-beam amplitudes associated with the two surface superlattice reflections **h** and **l** are given by

$$\begin{pmatrix} \varphi_h(t_s+t_b) \\ \varphi_l(t_s+t_b) \end{pmatrix} = \mathbf{P}_2 \begin{pmatrix} \varphi_h(t_s) \\ \varphi_l(t_s) \end{pmatrix}, \quad (18)$$

which leads to

$$\begin{aligned} \varphi_h(t_s+t_b) = {} & [\cos^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^3 t_b) \\ & + \sin^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^4 t_b)] \\ & \times \exp(-is_h t_b)\varphi_h(t_s) \\ & + \cos\tfrac{1}{2}\beta_1 \sin\tfrac{1}{2}\beta_1 [\exp(i\gamma'^4 t_b) \\ & - \exp(i\gamma'^3 t_b)] \exp(-is_h t_b)\varphi_l(t_s) \quad (19) \end{aligned}$$

$$\begin{aligned} \varphi_l(t_s+t_b) = {} & \{\sin\tfrac{1}{2}\beta_1 \cos\tfrac{1}{2}\beta_1 [\exp(i\gamma'^4 t_b) \\ & - \exp(i\gamma'^3 t_b)] \exp(-is_h t_b)\varphi_h(t_s) \\ & + [\sin^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^3 t_b) \\ & + \cos^2\tfrac{1}{2}\beta_1 \exp(i\gamma'^4 t_b)] \\ & \times \exp(-is_h t_b)\varphi_l(t_s). \quad (20) \end{aligned}$$

In general, the intensities of the surface superlattice reflections are strongly coupled with each other. The intensities therefore depend on the thickness of the bulk slab and will not hold their respective kinematical values in passing through the underlying bulk-crystal slab. However, if the surface superlattice reflections **h** and **l** are symmetry related as in the case shown in Fig. 2(a), such that their kinematical intensities are equal,

$$I_h(t_s) = I_l(t_s) = \langle I(t_s)\rangle,$$

where in general $\langle\ \rangle$ denotes the mean of I_h and I_l. From (18) we have

$$\begin{aligned} \langle I(t_s+t_b)\rangle &= \tfrac{1}{2}[\varphi_h^*(t_s+t_b), \varphi_l^*(t_s+t_b)] \times \begin{pmatrix} \varphi_h(t_s+t_b) \\ \varphi_l(t_s+t_b) \end{pmatrix} \\ &= \tfrac{1}{2}[\varphi_h^*(t_s), \varphi_l^*(t_s)]\mathbf{P}_2^\dagger \mathbf{P}_2 \times \begin{pmatrix} \varphi_h(t_s) \\ \varphi_l(t_s) \end{pmatrix}, \end{aligned}$$

where $\mathbf{P}^\dagger$ denotes the transposed conjugate complex matrix. Since the scattering matrix $\mathbf{P}$ is unitary ($\mathbf{P}^{-1} = \mathbf{P}^\dagger$),

$$\langle I(t_s+t_b)\rangle = \langle I(t_s)\rangle. \quad (21)$$

The averaged intensity of the symmetry-related surface superlattice reflections is then unaffected by the underlying bulk-crystal slab. An uncoupling in the averaged intensity is realized in this four-beam self-coupling case. It should be noted that when the excited surface superlattice reflections are not symmetric, as shown in Fig. 2(b), the above averaging procedure will not be applicable.

3.3. *Three-beam cross-coupling case*

In all the previously discussed examples, it has been assumed that reconstruction occurs only on the upper surface of the bulk-crystal slab. Coupling occurs then only among diffracted beams associated either with superlattice reflections or fundamental bulk reflections when passing through the bulk-crystal slab. In other words, cross coupling between the surface superlattice diffracted beams and fundamental-lattice diffracted beams is forbidden. Such a cross-coupling case may happen, however, when both the top and bottom surfaces of the crystal slab are reconstructed.

In this section we shall consider a three-beam case as shown in Fig. 3. We assume that the structure of the reconstructed bottom surface is identical to that of the top surface, the scattering matrix of the bottom surface layer then takes exactly the same form as given in § 3.1, equation (8), for the top surface layer. The exit amplitude vector $\mathbf{u}(2t_s+t_b)$ is given by

$$\begin{pmatrix} \varphi_0(2t_s+t_b) \\ \varphi_g(2t_s+t_b) \\ \varphi_h(2t_s+t_b) \end{pmatrix} = \mathbf{P}_s \begin{pmatrix} \varphi_0(t_s+t_b) \\ \varphi_g(t_s+t_b) \\ \varphi_h(t_s+t_b) \end{pmatrix}, \qquad (22)$$

giving the diffracted-beam amplitude of the superlattice reflection $\mathbf{h}$

$$\begin{aligned} \varphi_h(2t_s+t_b) = & \left\{ \sum_{j=1}^{3} C_h^j C_0^{*j} \exp(i\gamma^j t_s) \right\} \varphi_0(t_s+t_b) \\ & + \left\{ \sum_{j=1}^{3} C_h^j C_g^{*j} \exp(i\gamma^j t_s) \right\} \varphi_g(t_s+t_b) \\ & + \left\{ \sum_{j=1}^{3} C_h^j C_h^{*j} \exp(i\gamma^j t_s) \right\} \varphi_h(t_s+t_b). \end{aligned} \qquad (23)$$

To a kinematic approximation, we may write (see Appendix for details)

$$\sum_{j=1}^{3} C_h^j C_0^{*j} \exp(i\gamma^j t_s) \propto U_h,$$

$$\sum_{j=1}^{3} C_h^j C_g^{*j} \exp(i\gamma^j t_s) \propto U_{h-g},$$

$$\sum_{j=1}^{3} C_h^j C_h^{*j} \exp(i\gamma^j t_s) \simeq 1$$

and from (13)

$$\begin{aligned} \varphi_h(t_s+t_b) &= \varphi_h(t_s) \exp(i\gamma'^3 t_b) \\ &= \left[\sum_{j=1}^{3} C_h^j C_0^{*j} \exp(i\gamma^j t_s) \right] \exp(i\gamma'^3 t_b) \\ &\propto U_h. \end{aligned}$$

Equation (23) can then be approximated as

$$\begin{aligned} \varphi_h(2t_s+t_b) \propto & \, aU_h\varphi_0(t_s+t_b) \\ & + bU_{h-g}\varphi_g(t_s+t_b) + cU_h, \end{aligned} \qquad (24)$$

where a, b and c are constants of the same order of magnitude. For cases where the projected atomic positions of the lower surface towards the surface plane do not coincide with those of the upper surface, a phase factor needs to be included in a and b to take account of the shift of the coordinate origin of the lower surface. It is obvious from the above expression that in general the intensity of the superlattice reflection $\mathbf{h}$ will not retain its kinematic value as represented by the third term. However, if the bulk slab is so thin that the transmitted beam is still much stronger than the diffracted beam after passing through the bulk crystal slab, *i.e.*

$$|\varphi_0| \simeq 1 \gg |\varphi_g|,$$

we then have from (24)

$$\varphi_h(2t_s+t_b) \propto aU_h + cU_h \propto U_h. \qquad (25)$$

It should be emphasized that this result holds only when the kinematical approximation is valid for the whole crystal system, which is composed of the top reconstructed surface layer, bulk slab and the bottom reconstructed surface layer.

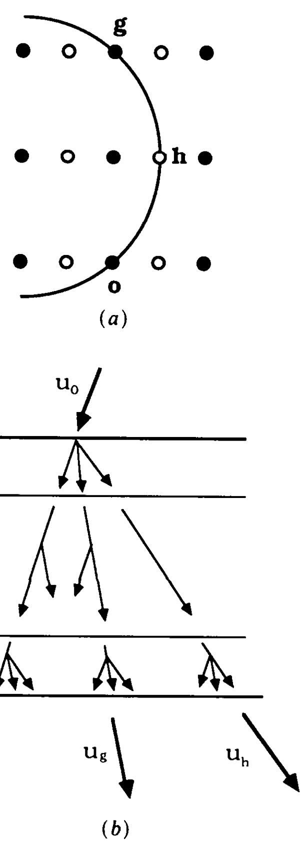

Fig. 3. Schematic diagrams of (*a*) diffraction conditions and (*b*) diffraction processes in passing through the whole crystal.

4. General cases

In general, many diffracted beams may be strongly excited, in particular when the incident electrons are propagating near a zone axis. The diffraction processes could differ appreciably from three- or four-beam cases as previously discussed. To extend the previous analysis, we now consider a general case in which many surface superlattice reflections as well as many fundamental reflections are excited.

When only the upper surface of the crystal slab is reconstructed, none of the surface superlattice reflections will be able to couple with fundamental reflections in passing through the bulk crystal slab. This is because in the bulk crystal superlattice reflections are all trivial ones in the sense that all Fourier potential coefficients associated with the scattering to fundamental reflections are zero. To see this, we write the fundamental reflections as $\mathbf{B}_i$ $(i=1,2,\ldots)$ and surface superlattice reflections $\mathbf{S}_i$ $(i=1,2,\ldots)$. The eigenvalue equation (3) can then be written in the form

$$\begin{pmatrix} -2K\gamma & U_{0B_1} & \cdots & U_{0S_1} & \cdots \\ U_{B_10} & 2K(s_{B_1}-\gamma) & \cdots & U_{B_1S_1} & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ U_{S_10} & U_{S_1B_1} & \cdots & 2K(s_{S_1}-\gamma) & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots \end{pmatrix}\begin{pmatrix} C_0 \\ C_{B_1} \\ \vdots \\ C_{S_1} \\ \vdots \end{pmatrix}=0. \tag{26}$$

Within the bulk crystal, this eigenvalue equation may be simplified, noticing that we have $U_{S_nB_m}=0$ in general, we get

$$\begin{pmatrix} \mathbf{M}_B & \mathbf{0} \\ \mathbf{0} & \mathbf{M}_S \end{pmatrix}\begin{pmatrix} \mathbf{C}_B \\ \mathbf{C}_S \end{pmatrix}=0 \tag{27}$$

where

$$\mathbf{M}_B=\begin{pmatrix} -2K\gamma & U_{0B_1} & \cdots & U_{0B_n} & \cdots \\ U_{B_10} & 2K(s_{B_1}-\gamma) & \cdots & U_{B_1B_n} & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ U_{B_n0} & U_{B_nB_1} & \cdots & 2K(s_{B_n}-\gamma) & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots \end{pmatrix},$$

$$\mathbf{M}_S=\begin{pmatrix} 2K(s_{S_1}-\gamma) & \cdots & U_{S_1S_n} & \cdots \\ \vdots & \vdots & \vdots & \vdots \\ U_{S_nS_1} & \cdots & 2K(s_{S_n}-\gamma) & \cdots \\ \vdots & \vdots & \vdots & \vdots \end{pmatrix}$$

and

$$\mathbf{C}_B=\begin{pmatrix} C_0 \\ C_{B_1} \\ \vdots \\ C_{B_n} \\ \vdots \end{pmatrix}, \qquad \mathbf{C}_S=\begin{pmatrix} C_{S_1} \\ \vdots \\ C_{S_n} \\ \vdots \end{pmatrix}.$$

For a non-trivial solution, the determinant of the matrix in (27) must be zero:

$$\begin{vmatrix} \mathbf{M}_B & \mathbf{0} \\ \mathbf{0} & \mathbf{M}_S \end{vmatrix}=|\mathbf{M}_B||\mathbf{M}_S|=0,$$

giving two groups of eigenvalues, associated with fundamental reflections and superlattice reflections, respectively,

$$\gamma_B^i\ (i=1,2,\ldots)$$
$$\gamma_S^i\ (i=1,2,\ldots),$$

which in turn result in two groups of eigenvectors

$$\begin{pmatrix} \{C_B^i\} \\ \{0\} \end{pmatrix}(i=1,2,\ldots);$$

and

$$\begin{pmatrix} \{0\} \\ \{C_S^i\} \end{pmatrix}(i=1,2,\ldots).$$

By substituting the following eigenvector matrices,

$$\mathbf{C}=\begin{pmatrix} \{C_B^i\} & \mathbf{0} \\ \mathbf{0} & \{C_S^i\} \end{pmatrix},$$

$$\mathbf{C}^{-1}=\begin{pmatrix} \{C_B^i\}^{-1} & \mathbf{0} \\ \mathbf{0} & \{C_S^i\}^{-1} \end{pmatrix},$$

and eigenvalue matrix,

$$\mathbf{Y}=\begin{pmatrix} \{\exp(i\gamma_B^it_b)\} & \mathbf{0} \\ \mathbf{0} & \{\exp(i\gamma_S^it_b)\} \end{pmatrix},$$

into (6) we obtain the scattering matrix $\mathbf{P}$ for the bulk-crystal slab

$$\mathbf{P}=\begin{pmatrix} \{C_B^i\} & \mathbf{0} \\ \mathbf{0} & \{C_S^i\} \end{pmatrix}\times\begin{pmatrix} \{\exp(i\gamma_B^it_b)\} & \mathbf{0} \\ \mathbf{0} & \{\exp(i\gamma_S^it_b)\} \end{pmatrix}\times\begin{pmatrix} \{C_B^i\}^{-1} & \mathbf{0} \\ \mathbf{0} & \{C_S^{*i}\}^{-1} \end{pmatrix}$$
$$=\begin{pmatrix} \{C_B^i\}\{\exp(i\gamma_B^it_b)\}\{C_B^i\}^{-1} & \mathbf{0} \\ \mathbf{0} & \{C_S^i\}\{\exp(i\gamma_S^it_b)\}\{C_S^i\}^{-1} \end{pmatrix}, \tag{28}$$

and the diffracted-beam amplitudes on leaving the lower surface of the crystal slab,

$$\{\varphi_{B_i}(t_s+t_b)\}=\{C_B^i\}\{\exp(i\gamma_B^it_b)\}\times\{C_B^i\}^{-1}\{\varphi_{B_i}(t_s)\} \tag{29}$$

$$\{\varphi_{S_i}(t_s+t_b)\}=\{C_S^i\}\{\exp(i\gamma_S^it_b)\}\times\{C_S^i\}^{-1}\{\varphi_{S_i}(t_s)\}. \tag{30}$$

The two groups of fundamental and superlattice reflections are therefore completely uncoupled from each other and are diffracted by the bulk crystal slab independently. Coupling is allowed, however, between reflections within the same group, *i.e.* either between fundamental reflections or between surface superlattice reflections.

The procedure of grouping may be further applied to subsets of surface superlattice reflections. A subset of superlattice reflections is defined as a group of superlattice reflections which can be connected to each other by a fundamental reflection vector, *i.e.*

$$\mathbf{S}_i^n - S_j^n = B_k,$$

where a superscript n has been used to denote the subset index. Since

$$S_i^n - \mathbf{S}_j^m \neq \mathbf{B}_k (n \neq m),$$

we then have

$$U_{S_i^n S_j^m} = 0$$

for all interacting Fourier potential coefficients between superlattice reflections belonging to different subsets, that is coupling between superlattice reflections in different subsets is forbidden in the bulk crystal. Following the same procedure leading to (30), we obtain

$$\{\varphi_{S_i^n}(t_s + t_b)\} = \{C^i_{S_i^n}\}\{\exp(i\gamma^i_{S_i^n} t_b)\} \times \{C^i_{S_i^n}\}^{-1}\{\varphi_{S_i^n}(t_s)\}. \quad (31)$$

In the extreme, when a subset of superlattice reflections consists of only one reflection, $C_{S_1^n} = 1$, *i.e.* as in Fig. 1(*a*). The diffracted-beam amplitude of this superlattice reflection after passing through the bulk slab is derived from the above general expression (31):

$$\varphi_{S_1^n}(t_s + t_b) = \exp(i\gamma_{S_1^n} t_b)\varphi_{S_1^n}(t_s),$$

and the intensity

$$I_{S_1^n}(t_s + t_b) = I_{S_1^n}(t_s),$$

which is completely unaffected by the underlying bulk-crystal slab. When the surface-layer diffraction can be treated kinematically, the intensity of this superlattice reflection will have a kinematical value which is proportional to $U^2_{S^n}$; and this kinematical value will be preserved in passing through the underlying bulk crystal regardless of its thickness t_b.

A different but interesting case occurs if within a subset of superlattice reflections only those reflections which are entirely symmetry related have been strongly excited such that

$$I_{S_1^n}(t_s) = I_{S_2^n}(t_s) = \ldots = \langle I_{S^n}(t_s)\rangle.$$

The averaged intensity of these symmetry-related superlattice reflections after passing through the underlying bulk crystal is given by

$$\begin{aligned}\langle I_{S^n}(t_s + t_b)\rangle &= N^{-1}\{\varphi^*_{S_i^n}(t_s + t_b)\}\{\varphi_{S_i^n}(t_s + t_b)\} \\ &= N^{-1}\{\varphi^*_{S_i^n}(t_s)\}\{\varphi_{S_i^n}(t_s)\} \\ &= \langle I_{S^n}(t_s)\rangle, \qquad (32)\end{aligned}$$

where N is the total number of symmetry-related reflections in the same subset which are strongly excited. The averaged intensity is thus independent of the bulk-crystal slab. We have then an uncoupling case in the averaged intensity of the subset of superlattice reflections.

5. Concluding remarks

A Bloch-wave analysis has been applied to the coupling problem in the TED analysis of reconstructed surfaces. The great success of TED in determining the structure of reconstructed surfaces is rooted in the validity of the kinematical approximation for the intensities of the surface superlattice reflections. The validity of the kinematic approximation depends primarily on the uncoupling of the surface-layer superlattice scattering from underlying bulk scattering.

Three categories of coupling, *i.e.* self coupling, cross coupling and complete uncoupling in either the intensity of a single superlattice reflection or an averaged intensity among a group of symmetry-related superlattice reflections, are distinguished.

When the electrons are incident on the top reconstructed surface layer of a bulk crystal, both fundamental and surface superlattice reflections may be excited. In the case when the surface layer is thin enough and the scattering power of the surface atoms is not very strong, a kinematical (single-scattering) approximation may be applied to the diffraction processes of the entrance surface layer.

In passing through the underlying bulk-crystal slab, only self coupling is allowed, that is coupling may occur between either fundamental reflections or superlattice reflections but not between each other. Furthermore, all superlattice reflections can be grouped into subsets in accordance with whether they are related by fundamental reciprocal-lattice vectors. Coupling between superlattice reflections in different subsets is forbidden. Diffracted beams travel through the bulk-crystal slab in groups. Each group is totally unaffected by diffraction processes resulting from reflections belonging to any other groups. On leaving the bulk-crystal slab, if a subset of superlattice reflections consists of only one reflection, the intensity of the superlattice reflection will be the same as when entering the bulk slab, and the kinematical value of the intensity is preserved. If a subset is composed of entirely symmetry-related superlattice reflections, the averaged intensity of the subset superlattice reflections will maintain its intensity constant in passing through the bulk crystal. The procedure of averaging cannot, however, be applied to reflections belonging to different subsets, and to subsets which are not composed entirely of symmetry-related reflections.

When the bottom surface of the bulk slab is also reconstructed, cross coupling is then allowed between fundamental reflections and superlattice reflections. Unless the whole-crystal slab is extremely thin so that only single-scattering processes occur in passing through the entrance reconstructed surface, the underlying bulk slab and the exit reconstructed surface, the kinematical values of the intensities of surface superlattice reflections will not be preserved in general.

A different but interesting case (*i.e.* when the surface reconstruction occurs only on the bottom face of the bulk-crystal slab) has also been recently proposed as an alternative to the above-mentioned case (Nakai, 1988; Takayanagi, 1990). It should be noted, however, that the uncoupling mechanism of superlattice reflections in the latter case is entirely different from that being discussed in the present paper. A complete uncoupling can be achieved when and only when there is no Bragg diffraction of the incident beam in passing through the bulk-crystal slab.

Lastly, it is worth mentioning that although our analysis has been applied to reconstructed surfaces, it can also be applied to any other surfaces giving reflections which do not entirely coincide with fundamental reflections.

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APPENDIX

In the transmission Laue case, the fundamental eigenvalue equation takes the form

$$2K(s_g-\gamma^{(j)})C_g^{(j)}+\sum_{h\neq g}U_{g-h}C_h^{(j)}=0.$$

By multiplying the above equation from the left by $C_l^{*(j)}$ and summing over j we obtain

$$\begin{aligned}2K\sum_j C_l^{*(j)}\gamma^{(j)}C_g^{(j)}&=2Ks_g\sum_j C_l^{*(j)}C_g^{(j)}\\&\quad+\sum_{h\neq g}U_{g-h}\sum_j C_l^{*(j)}C_h^{(j)}\\&=2Ks_g\delta_{lg}+U_{g-l}(1-\delta_{lg}).\end{aligned}$$

To a first approximation in the surface-layer thickness t_s, we may write

$$\exp(i\gamma^{(j)}t_s)\simeq 1+i\gamma^{(j)}t_s,$$

and therefore in general

$$\begin{aligned}&\sum_j C_g^{*(j)}C_h^{(j)}\exp(i\gamma^{(j)}t_s)\\&\quad\simeq\sum_j C_g^{*(j)}(1+i\gamma^{(j)}t_s)C_h^{(j)}\\&\quad=\delta_{gh}+it_s[s_g\delta_{hg}+(U_{h-g}/2K)(1-\delta_{hg})].\end{aligned}$$

For the case where $g=0$ and $h\neq 0$, we have

$$\sum_j C_h^{(j)}C_0^{*(j)}\exp(i\gamma^{(j)}t_s)\simeq(i/2K)t_sU_h\propto U_h;$$

for $g\neq h$ and $g\neq 0$, $h\neq 0$,

$$\sum_j C_h^{(j)}C_g^{*(j)}\exp(i\gamma^{(j)}t_s)\simeq(i/2K)t_sU_{h-g}\propto U_{h-g},$$

and

$$\sum_j C_h^{(j)}C_h^{*(j)}\exp(i\gamma^{(j)}t_s)\simeq\delta_{hh}=1.$$

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