

- MATHIESON, A. MCL. (1985*b*). *Acta Cryst.* **A41**, 603-605.
 MATHIESON, A. MCL. (1985*c*). *J. Appl. Cryst.* **18**, 506-508.
 MATHIESON, A. MCL. (1988). *Acta Cryst.* **A44**, 239-244.
 MATHIESON, A. MCL. & STEVENSON, A. W. (1985). *Acta Cryst.* **A41**, 290-296.
 MATHIESON, A. MCL. & STEVENSON, A. W. (1986). *Acta Cryst.* **A42**, 435-441.
 PANTAZATOS, P. & WERNER, S. A. (1973). *Acta Cryst.* **A29**, 557-558.
 SCHOENBORN, B. P. (1983). *Acta Cryst.* **A39**, 315-321.
 SEQUEIRA, A. (1974). *Acta Cryst.* **A30**, 839-843.
 SJÖLIN, L. & WLODAWER, A. (1981). *Acta Cryst.* **A37**, 594-604.
 SPENCER, S. A. & KOSSIAKOFF, A. A. (1980). *J. Appl. Cryst.* **13**, 563-571.
 STEVENSON, A. W. (1988). *Acta Cryst.* A. In the press.
 WERNER, S. A. (1971). *Acta Cryst.* **A27**, 665-669.
 WILKINS, S. W. (1983). *Phys. Rev. Lett.* **50**, 1862-1865.
 WILLIS, B. T. M. (1960). *Acta Cryst.* **13**, 763-766.

Acta Cryst. (1988). **A44**, 1042-1044

Bethe's Correction Method for Dynamical Calculation of Reflection High-Energy Electron Diffraction Intensities from General Surfaces

BY AYAHIKO ICHIMIYA

Department of Applied Physics, School of Engineering, Nagoya University, Chikusa-ku, Nagoya 464-01, Japan

(Received 22 February 1988; accepted 8 June 1988)

Abstract

The correction method for weak diffraction of Bethe [*Ann. Phys. (Leipzig)* (1928), **87**, 55-129] is applied to dynamical calculation of reflection high-energy electron diffraction (RHEED) intensities from general surfaces based on a multi-slice method. For high-step-density surfaces, it is shown that the corrected surface potential (Bethe potential) depends on the incident direction of electrons for step directions. Furthermore it is shown that the Bethe potential is approximately proportional to the coverage of adsorbed atoms or of terraces of high-step-density surfaces. For the RHEED intensity from stepped surfaces, the intensity oscillation during molecular beam epitaxial growth is discussed. An appropriate calculational formula for reconstructed surfaces is also obtained.

1. Introduction

Practical methods of reflection high-energy electron diffraction (RHEED) intensity calculation were proposed by several theoretical studies (Masud & Pendry, 1976; Maksym & Beeby, 1981; Ichimiya, 1983; Peng & Cowley, 1986). These methods are appropriate to perfect crystal surfaces, but most of the methods are not available for imperfect surfaces. The first theoretical formalism for imperfect surfaces was presented with a perturbation method by Beeby (1979). Peng & Cowley (1986) proposed a new method for calculating RHEED intensities which would allow the calculation of intensities from imperfect surfaces. Electron diffraction intensity distributions from imperfect surfaces were interpreted using kinematic diffraction theory by several authors (Matysik, 1974; Henzler, 1977; Holloway & Beeby, 1978; Holloway, 1979; Van Hove, Lent, Pukite &

Cohen, 1983; Lent & Cohen, 1984; Pimply & Lu, 1985; Pukite, Lent & Cohen, 1985). Recently some theoretical approaches were proposed to interpret RHEED intensity oscillations (Van Hove, Lent, Pukite & Cohen, 1983; Kawamura, Maksym & Iijima, 1984; Kawamura & Maksym, 1985; Ichimiya, 1987). In a previous paper (Ichimiya, 1987) RHEED intensities from a surface with low step densities were obtained analytically from Kirchhoff's diffraction theory. From the calculations it was shown that integrated intensities of RHEED scarcely depend on the step distributions and terrace coverage, but the intensities on reciprocal rods depend sensitively on these factors. Kawamura & Maksym (1985) have shown that the oscillation property of RHEED intensities during molecular beam epitaxial (MBE) growth depends on crystal orientation and the step directions for the incident beam because of dynamic diffraction effects. In their calculation dynamic diffraction as high density and periodic distribution of steps was taken into consideration.

In the present work Bethe's correction method (Bethe, 1928; Ichikawa & Hayakawa, 1977) for weak beams in the dynamical theory of electron diffraction was applied to calculation of RHEED intensities from general surfaces such as stepped, reconstructed, distorted or adsorbed surfaces based on the multi-slice method (Ichimiya, 1983).

2. Bethe's correction for general surfaces

According to the multi-slice method of RHEED dynamical theory (Ichimiya, 1983), the Schrödinger equation at the j th slice parallel to the surface is expressed by a two-dimensional (2D) position vector \mathbf{r} and a 2D wave vector \mathbf{k}_0 , which are parallel to the

surface, as

$$(\nabla^2 + \mathbf{k}_0^2)\psi_j(\mathbf{r}) + V_j(\mathbf{r})\psi_j(\mathbf{r}) = 0, \quad (1)$$

where $\psi_j(\mathbf{r})$ is a 2D wave function and $V_j(\mathbf{r})$ a 2D potential multiplied by $2me/\hbar^2$ at the j th slice. For a 2D periodic potential field, $V(\mathbf{r})$ and $\psi(\mathbf{r})$ (hereafter the suffix j is removed for simplicity) can be expanded in Fourier series

$$V(\mathbf{r}) = \sum_n U_n \exp(i\mathbf{B}_n \mathbf{r}) \quad (2)$$

$$\psi(\mathbf{r}) = \sum_n \varphi_n \exp(i\mathbf{k}_n \mathbf{r}) \quad (3)$$

where \mathbf{B}_n is a 2D reciprocal-lattice vector (reciprocal-rod vector) and

$$\mathbf{k}_n = \mathbf{k}_0 + \mathbf{B}_n. \quad (4)$$

Substituting (2) and (3) into (1), one obtains a set of homogeneous equations for the 2D periodic field as

$$(k_0^2 - k_n^2)\varphi_n + \sum_m U_{n-m}\varphi_m = 0. \quad (5)$$

For a nonperiodic potential, (2) and (3) are rewritten as

$$V(\mathbf{r}) = \int U(\mathbf{s}) \exp(i\mathbf{s}\mathbf{r}) \, d\mathbf{s} \quad (6)$$

$$\begin{aligned} \psi(\mathbf{r}) &= \int \varphi(\mathbf{s}) \exp[i(\mathbf{k}_0 + \mathbf{s})\mathbf{r}] \, d\mathbf{s} \\ &= \int \varphi(\mathbf{s}) \exp(i\mathbf{k}_s \mathbf{r}) \, d\mathbf{s}. \end{aligned} \quad (7)$$

Upon substitution of (6) and (7) into (1), the set of equations for the nonperiodic potential field becomes

$$(k_0^2 - k_s^2)\varphi(\mathbf{s}) + \int U(\mathbf{s} - \mathbf{s}')\varphi(\mathbf{s}') \, d\mathbf{s}' = 0. \quad (8)$$

When the 2D potential field is slightly distorted from a periodic one, $U(\mathbf{s})$ can be written as

$$U(\mathbf{s}) = \sum_m U_m \delta(\mathbf{s} - \mathbf{B}_m) + \Delta U(\mathbf{s}), \quad (9)$$

where $\delta(\mathbf{s})$ is Dirac's δ function and $\Delta U(\mathbf{s})$ is a term of the distortion from the periodic potential field which gives rise to weak diffuse scattering. Substituting (9) into (8), we obtain

$$\begin{aligned} (k_0^2 - k_s^2)\varphi(\mathbf{s}) + \sum_m U_m \varphi(\mathbf{s} - \mathbf{B}_m) + \int \Delta U(\mathbf{s} - \mathbf{s}')\varphi(\mathbf{s}') \, d\mathbf{s}' \\ = 0. \end{aligned}$$

When we put $\varphi(\mathbf{B}_n) = \varphi_n$ for $\mathbf{s} = \mathbf{B}_n$, the above equation becomes

$$\begin{aligned} (k_0^2 - k_n^2)\varphi_n + \sum_m U_{n-m}\varphi_m + \int \Delta U(\mathbf{B}_n - \mathbf{s}')\varphi(\mathbf{s}') \, d\mathbf{s}' \\ = 0. \end{aligned} \quad (10)$$

From (8),

$$\varphi(\mathbf{s}) = -\frac{\int U(\mathbf{s} - \mathbf{s}')\varphi(\mathbf{s}') \, d\mathbf{s}'}{k_0^2 - k_s^2}. \quad (11)$$

Since dynamical diffraction occurs predominantly between the waves diffracted into reciprocal rods,

$\varphi(\mathbf{s}')$ in (11) is put approximately equal to

$$\varphi(\mathbf{s}') = \sum_m \varphi_m \delta(\mathbf{s}' - \mathbf{B}_m).$$

Then (11) becomes

$$\varphi(\mathbf{s}) = -\frac{\sum_m U(\mathbf{s} - \mathbf{B}_m)\varphi_m}{k_0^2 - k_s^2}. \quad (12)$$

Substituting (12) into (10) and using (8), we obtain

$$\begin{aligned} (k_0^2 - k_n^2)\varphi_n \\ + \sum_m \left[U_{n-m} - \int \frac{\Delta U(\mathbf{B}_n - \mathbf{s}) \Delta U(\mathbf{s} - \mathbf{B}_m)}{k_0^2 - k_s^2} \, d\mathbf{s} \right] \varphi_m = 0, \end{aligned} \quad (13)$$

because $\Delta U(\mathbf{B}_n - \mathbf{B}_m) = 0$. When we put

$$U'_{n-m} = U_{n-m} - \int \frac{\Delta U(\mathbf{B}_n - \mathbf{s}) \Delta U(\mathbf{s} - \mathbf{B}_m)}{k_0^2 - k_s^2} \, d\mathbf{s}, \quad (14)$$

(13) is reduced to the same formalism as (7) for a periodic potential as

$$(k_0^2 - k_n^2)\varphi_n + \sum_m U'_{n-m}\varphi_m = 0. \quad (15)$$

U'_n is named the 2D Fourier component of the Bethe potential, because the second term of (14) is an effect of diffuse scattering. Thus RHEED intensities of reciprocal rods for general surfaces can be calculated from (15) by the usual methods of RHEED dynamical theory (Maksym & Beeby, 1981; Ichimiya, 1983).

3. Bethe potential for stepped and adsorbed surfaces

A Fourier component of the 2D potential for stepped or adsorbed surfaces is

$$U(\mathbf{s}) = \sum_j u(\mathbf{s})S_j(\mathbf{s}) \exp(i\mathbf{s}\mathbf{r}_j), \quad (16)$$

where $u(\mathbf{s})$ is the 2D atomic scattering factor, \mathbf{r}_j the 2D original position vector of the j th terrace or island, and

$$S_j(\mathbf{s}) = \sum_k \exp[i\mathbf{s}(\mathbf{r}_k - \mathbf{r}_j)],$$

which is the shape function of the j th terrace or island, and \mathbf{r}_k is the 2D position vector of the k th atom. For a completely flat surface,

$$U(\mathbf{B}_n) = U_n = N_0 u(\mathbf{B}_n), \quad (17)$$

where N_0 is the number of atoms in the surface. For stepped or adsorbed surfaces, where atoms are at correct lattice sites,

$$\begin{aligned} U(\mathbf{B}_n) &= \sum_j u(\mathbf{B}_n)S_j(\mathbf{B}_n) \\ &= \sum_j u(\mathbf{B}_n)N_j, \end{aligned} \quad (18)$$

where N_j is the number of atoms in the j th terrace or island. If the terrace or island coverage θ_j is defined

as $\theta_j = N_j/N_0$,

$$U(\mathbf{B}_n) = \sum_j \theta_j U_n = \theta U_n, \quad (19)$$

where $\theta = \sum_j \theta_j$ is total coverage of terraces or islands. From (19), the Bethe potential U'_{n-m} becomes

$$U'_{n-m} = \theta U_{n-m} - \int \frac{\Delta U(\mathbf{B}_n - \mathbf{s}) \Delta U(\mathbf{s} - \mathbf{B}_m)}{k_0^2 - k_s^2} ds, \quad (20)$$

where

$$\Delta U(\mathbf{s}) = \sum_j u(\mathbf{s}) S_j(\mathbf{s}) \exp(i s r_j) - \theta \sum_n U_n \delta(\mathbf{s} - \mathbf{B}_n). \quad (21)$$

For the one-dimensional step distributions treated by Kawamura & Maksym (1985), (20) becomes

$$U'_{n-m} = \theta U_{n-m} + \int [\Delta U(\mathbf{B}_n - \mathbf{s}) \Delta U(\mathbf{s} - \mathbf{B}_m)] / s^2 ds, \quad (22)$$

when the incident beam is parallel to the step edge, $\mathbf{k}_0 \perp \mathbf{s}$; and

$$U'_{n-m} = \theta U_{n-m} + \int \frac{\Delta U(\mathbf{B}_n - \mathbf{s}) \Delta U(\mathbf{s} - \mathbf{B}_m)}{2k_0 s + s^2} ds, \quad (23)$$

when the incident beam is perpendicular to the step edge, $\mathbf{k}_0 \parallel \mathbf{s}$. Since the Bethe potential depends on the incident directions as (22) and (23), RHEED intensity oscillation behaviour during MBE growth is expected to differ with direction as was shown by Kawamura, Sakamoto & Ohta (1986). The second term of (22) becomes very small for a high-step-density surface, because $2k_0 s$ becomes very large. Therefore the Bethe potential for such a high-density stepped surface is approximately given as $V'(\mathbf{r}) \approx \theta V(\mathbf{r})$, as pointed out by Kawamura & Maksym (1985). On the other hand, although the second term of (20) becomes significant for low-step-density surfaces, in such cases the treatment by Kirchhoff's diffraction theory is effective as described by Ichimiya (1983).

4. Bethe potential for reconstructed surface

For reconstructed surfaces, many fractional-order spots are observed in the RHEED pattern. Therefore many beams for dynamical calculation are required to be taken into account, and the calculations involve long CPU times of a computer. The Bethe correction method is also appropriate to such reconstructed surfaces.

For reconstructed surfaces, a Fourier component of the 2D potential can be written as

$$U(\mathbf{s}) = \sum_m U_m \delta(\mathbf{B}_m - \mathbf{s}) + \sum_n U_n \delta(\mathbf{B}_n - \mathbf{s}), \quad (24)$$

where suffix m is for integral-order reflections and n is for fractional-order ones. Substituting (24) into (14) we can obtain the Bethe potential

$$\begin{aligned} U'_{n-m} &= U_{n-m} - \sum_l \frac{U_{n-l} U_{l-m}}{k_0^2 - k_l^2}, \\ &= U_{n-m} + \sum_l \frac{U_{n-l} U_{l-m}}{2\mathbf{kB}_l + B_l^2}, \end{aligned} \quad (25)$$

where the summation is performed for fractional orders. The second term of (25) is appreciable for the zeroth Laue zone, because $\mathbf{kB}_l = 0$. For the other Laue zones the second term becomes very small because of the large dominator, including large wave number. Therefore in the many-beam dynamical calculation, we have to take only the zeroth-Laue-zone reflections into account.

Thus, the use of the present Bethe correction method allows dynamical diffraction effects of fractional-order reflections on rocking curves of integral-order reflections to be estimated without numerous computations.

References

- BEEBY, J. L. (1979). *Surf. Sci.* **80**, 56-61.
 BETHE, H. (1928). *Ann. Phys. (Leipzig)*, **87**, 55-129.
 HENZLER, M. (1977). In *Electron Spectroscopy for Surface Analysis*, edited by H. IBACH, pp. 117-149. Berlin: Springer.
 HOLLOWAY, S. (1979). *Phys. Lett.* **71A**, 476-480.
 HOLLOWAY, S. & BEEBY, J. M. (1978). *J. Phys. C*, **11**, L247-L251.
 ICHIKAWA, M. & HAYAKAWA, K. (1977). *J. Phys. Soc. Jpn*, **42**, 1957-1964.
 ICHIMIYA, A. (1983). *Jpn. J. Appl. Phys.* **22**, 176-180.
 ICHIMIYA, A. (1987). *Surf. Sci.* **187**, 194-200.
 KAWAMURA, T. & MAKSYM, P. A. (1985). *Surf. Sci.* **161**, 12-24.
 KAWAMURA, T., MAKSYM, P. A. & IJIMA, T. (1984). *Surf. Sci.* **148**, L671-L676.
 KAWAMURA, T., SAKAMOTO, T. & OHTA, K. (1986). *Surf. Sci.* **171**, L409-L414.
 LENT, C. S. & COHEN, P. I. (1984). *Surf. Sci.* **139**, 121-154.
 MAKSYM, P. A. & BEEBY, J. L. (1981). *Surf. Sci.* **110**, 423-438.
 MASUD, N. & PENDRY, J. B. (1976). *J. Phys. C*, **9**, 1833-1844.
 MATYSIK, K. J. (1974). *Surf. Sci.* **46**, 457-472.
 PENG, L. M. & COWLEY, J. M. (1986). *Acta Cryst.* **A42**, 545-552.
 PIMBLY, J. M. & LU, T. M. (1985). *J. Appl. Phys.* **58**, 2184-2189.
 PUKITE, P. R., LENT, C. S. & COHEN, P. I. (1985). *Surf. Sci.* **161**, 39-68.
 VAN HOVE, J. M., LENT, C. S., PUKITE, P. R. & COHEN, P. I. (1983). *J. Vac. Sci. Technol.* **B1**, 741-746.