Analysis of Reflection High Energy Electron Diffraction Azimuthal Plots

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(Received 1 March 1993)

Reflection high energy electron diffraction data collected in the form of an azimuthal plot are analyzed theoretically for the first time. Experimental results for Si(111) at a glancing angle satisfying a Bragg condition are taken from the literature. Calculations are carried out within a multiple scattering approach. Excellent agreement between experimental and theoretical results is achieved. A simple qualitative explanation of the shape of the azimuthal plot analyzed is also presented.

PACS numbers: 61.14.Hg, 68.35.Bs, 68.55.Jk

At present, reflection high energy electron diffraction (RHEED) is widely used for controlling the growth of ultrathin films [1]. The use of RHEED became very popular after the discovery of RHEED intensity oscillations by Harris, Joyce, and Dobson [2]. It is well established that for layer-by-layer growth the intensity of the specular beam oscillates with a period corresponding to deposition of one atomic layer. There are different simplified theoretical descriptions of this phenomenon [3–5]. It has recently been shown that when diffraction conditions are specially selected it is possible to obtain good agreement between experimental and theoretical results using these descriptions [6–8]. However, it is clear from recent research that a better understanding of the phenomenon of RHEED oscillations can be achieved only after improvement in the fundamental understanding of RHEED from flat and growing surfaces.

So far, fundamental research on RHEED has been devoted to theoretical analysis of experimental rocking curves, i.e., the intensity of reflected electron beams collected while varying the glancing angle of the incident beam (assuming that the shape of the surface remains constant during measurements). There are examples of successful analyses of RHEED rocking curves for simple surfaces [9,10]. Generally, it has been found that a good quantitative description of RHEED results can be achieved when multiple scattering of electrons is considered. However, it has also been found that for interpretation of RHEED rocking curves it is necessary to consider the interaction between a large number of diffracted beams and the origin of peaks in the experimental data is difficult to understand qualitatively. This implies that theoretical analysis requires extensive numerical calculations. All the above-mentioned findings are true for a wide range of surface structures (including relatively simple ones) and are discussed in detail in the literature, for example, in Refs. [11,12]. Such a situation causes practical difficulties in attempts to analyze precisely rocking curves from surfaces with complicated structures. First, because multiple scattering effects may not be simply predictable it is not easy to recognize them when they exist together with inelastic and diffuse scattering effects. Second, calculations for complicated surfaces require enormous amounts of computer time. Recent papers on Pt(110)-(2×1) [13] and GaAs(001)-(2×4) [14] demonstrate that it is possible to overcome these difficulties successfully. However, it is also clear from these papers that the actual possibilities for carrying out similar analyses are very limited.

In this Letter we analyze for the first time experimental data collected in the form of an azimuthal plot, i.e., the specular beam intensity measured while rotating a sample around the axis perpendicular to the surface. There are a few examples of such plots [15–17] in the literature. Because they have complicated shapes with a large number of sudden peaks and valleys [see Fig. 1(a)], they were considered difficult to explain and no theoretical interpretation has been performed so far. The results

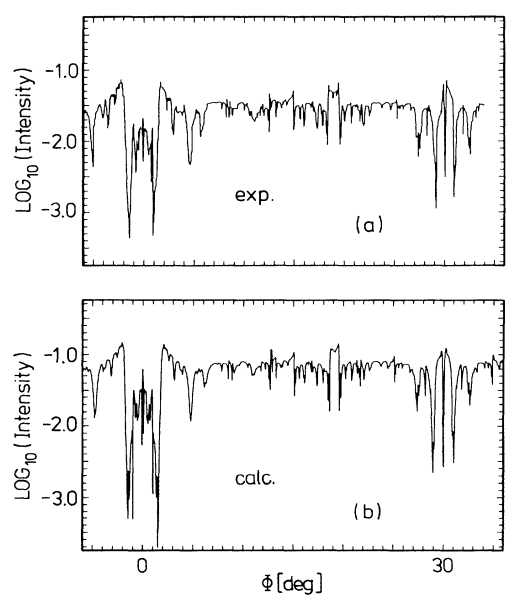

FIG. 1. (a) An experimental azimuthal plot for 40 keV electrons incident on the Si(111) surface at a glancing angle satisfying the 444 Bragg condition [15]. (b) The calculated azimuthal plot corresponding to the data shown in (a). Values of the azimuthal angle Φ of 0 and 30 deg correspond to the azimuths $\langle 10\bar{1}\rangle$ and $\langle 11\bar{2}\rangle$, respectively.

of this Letter suggest a reassessment of the usefulness of azimuthal plots: It is shown that for azimuthal plots it is possible to obtain a very good quantitative theoretical explanation and also a simple qualitative one.

Experimental data for Si(111) collected by Menadue [15] more than twenty years ago were taken from the literature. So far, these data have remained unanalyzed. Calculations were carried out using a numerical program based on the combination of a two-dimensional Bloch wave theory of RHEED [18] and a layer doubling algorithm developed originally in the theory of LEED [19]. The program has been already described in detail [9,11,20]. Results of the calculations shown in Fig. 1(b) were carried out for 35 beams. The choice of beams is discussed in a later part of the Letter. We checked that including more beams changes the intensity by an amount smaller than the experiment error [as deduced from comparison of the shape of the curve from Fig. 1(a) around the symmetrical azimuth $\langle 11\bar{2}\rangle$]. The real part of the potential used in the calculations was determined initially from the electron scattering coefficients tabulated by Doyle and Turner [21] but corrected for thermal vibrations at 300 K. Next, the real part of the potential was relativistically corrected [9] and finally a constant component was added to fit the average volume potential to the value of -12 eV determined experimentally [15]. The imaginary part of the potential was assumed to be equal to 0.1 of the real part taken without the correction of the average value. The calculations were carried out for incident electrons of 40 keV (but this value was actually taken into account after adding a relativistic correction [9]) and for the glancing angle of 1.982° which corresponds to the 444 Bragg reflection. All the abovementioned parameter values match the experimental conditions of Menadue [15]. We have assumed a bulk terminated surface for Si(111), since there is very little information on the surface reconstruction of the measured sample. However, because of the use of the glancing angle satisfying the Bragg condition we expect that diffraction effects are caused mainly by deeper layers of the crystal.

Comparing the experimental and theoretical results [Figs. 1(a) and 1(b)] we can observe very good agreement between them. All the peaks and valleys found in the measurements are well reproduced numerically. Moreover, their relative maxima and minima, respectively, are also well determined by calculations. This means that multiple scattering theory can describe properly the shapes of azimuthal plots. It seems important to consider whether we could explain the azimuthal plot from Fig. 1(a) using the simpler, single scattering theory of diffraction (usually called the kinematic theory). The kinematic theory fails because the final formulas for the intensity of the specularly reflected beam do not depend on the azimuthal angle of the incident beam (for example [22]). In other words, the intensity is predicted to remain constant during azimuthal variations. This means that analysis of azimuthal plots can be carried out only with the use of approaches including multiple scattering (dynamical) effects.

Both the experimental and calculated curves from Fig. 1 have very complicated shapes. However, we have found that they can easily be described qualitatively. Before demonstrating this we discuss some details of the two-dimensional Bloch wave approach. In this approach it is assumed that the scattering potential is periodic in the two dimensions parallel to the surface. Because of this it is possible to expand the whole electron wave function $\Psi(\mathbf{r})$ in the following form [18]:

$$\Psi(\mathbf{r}) = \sum_{\boldsymbol{\kappa}} \{Q_{\boldsymbol{\kappa}}^{-}(z)\exp(-ik_{\boldsymbol{\kappa}}z) + Q_{\boldsymbol{\kappa}}^{+}(z)\exp(+ik_{\boldsymbol{\kappa}}z)\} \times \exp[i(k_{\parallel}+\boldsymbol{\kappa})\cdot\boldsymbol{\rho}]\,, \tag{1}$$

where $\boldsymbol{\kappa}$ is a vector in the 2D surface reciprocal mesh, $\mathbf{k}_{\parallel}$ is the parallel component of the incident electron wave vector $\mathbf{k}$, and $\boldsymbol{\rho}$ is the parallel component of $\mathbf{r}$. The perpendicular wave vector magnitudes $\mathbf{k}_{\boldsymbol{\kappa}}$ obey the following relation:

$$\mathbf{k}_{\boldsymbol{\kappa}}^2 = \mathbf{k}^2 - (\mathbf{k}_{\parallel}+\boldsymbol{\kappa})^2\,. \tag{2}$$

In this approach the word "beam" usually means the term corresponding with one value of the vector $\boldsymbol{\kappa}$.

Above the surface ($z>0$) the wave function $\Psi(\mathbf{r})$ has a very simple form for which a physical interpretation exists. In this case we can write

$$\Psi(\mathbf{r}) = \exp(-ik_0z)\exp(i\mathbf{k}_{\parallel}\cdot\boldsymbol{\rho}) + \sum_{\boldsymbol{\kappa}} R_{\boldsymbol{\kappa}}\exp(+ik_{\boldsymbol{\kappa}}z)\exp[i(\mathbf{k}_{\parallel}+\boldsymbol{\kappa})\cdot\rho]\,. \tag{3}$$

The first term in (3) represents the incident electron wave. The further terms are reflected waves, which can be propagating ($k_{\boldsymbol{\kappa}}^2>0$) or evanescent ($k_{\boldsymbol{\kappa}}^2<0$). Each propagating wave can be observed experimentally as it causes the existence of a spot at the screen. This situation can be explained with the help of the well-known Ewald's construction [1]. Each rod in the two-dimensional reciprocal mesh corresponds to one beam, but only beams for which $k_{\boldsymbol{\kappa}}^2>0$ can give a contribution to the pattern on the screen. It is clear that it is possible to fulfill the following condition:

$$k_{\boldsymbol{\kappa}}^2 = 0\,. \tag{4}$$

This situation is called a beam emergence condition and it corresponds to the existence of a spot just at the shadow edge at the RHEED screen. When condition (4) for any beam is exactly fulfilled then expansion (1) cannot be applied. Because of this, such situations are excluded from the present theoretical treatment within the two-dimensional Bloch wave approach and also from calculations with the numerical program described. However, it has already been demonstrated that by using the layer doubling algorithm it is possible to carrying out convergent and stable calculations when values of $k_{\boldsymbol{\kappa}}^2$ are very close to satisfying condition (4) [23].

We now return to the qualitative description of the azimuthal plots. It turns out that most of the fluctuations in the intensity can be described qualitatively as a simple sum of effects accompanying interactions between the main beam (i.e., assigned to the zeroth vector of the two-dimensional reciprocal mesh) and those which are close to satisfying beam emergence conditions (4). Figure 2 shows the set of beams used to carry out the 35-beam calculations presented in Fig. 1(b). This set includes all the important beams for which it is possible to satisfy condition (4) while varying the azimuthal angle within the chosen range (but it also includes other beams which turned out to be relevant for precise quantitative analysis). To study effects connected with satisfying the conditions determined by Eq. (4) we calculated a number of azimuthal plots in a simplified way. The curves presented in Fig. 3 were obtained in three-beam calculations, i.e., for each curve only the main and two side beams were taken into account. The reason it is not useful to carry out calculations for less than three beams is that Eq. (4) can often be satisfied in a very narrow azimuthal range for two opposite side beams. It can be seen that most of the peaks and valleys from Fig. 1 are present in Fig. 3. It can be observed also that in a wide region between the $\langle 10\bar{1}\rangle$ and $\langle 11\bar{2}\rangle$ azimuths the shapes of the curves of Fig. 1 can be reproduced roughly by summing up intensity changes from three-beam calculations. The only exception to this rule is near the $\langle 10\bar{1}\rangle$ and $\langle 11\bar{2}\rangle$ azimuths, but this is to be expected as, for these azimuthal ranges, many beam emergence conditions exist and so strong interaction occur between many beams.

Some comment should be added on the possible origin of effects accompanying the satisfaction of the beam emergence condition (4) for one of the beams, if we assume an interaction only between that beam and the main one. We expect that for the case $k_\kappa^2 > 0$ (the propagating wave case) peaks and valleys in the specular beam intensity appear due to secondary Bragg reflections. For $k_\kappa^2 < 0$ (the evanescent wave case) we expect that changes in the specular beam intensity are caused by surface resonances. (A detailed discussion of the nature of these diffraction phenomena can be found in Refs. [19,24].)

On the basis of the results presented it is possible to draw conclusions about further investigations concerning azimuthal plots measured between principal azimuth directions. In such regions it should be possible to detect experimentally and recognize uniquely multiple scattering effects connected with the lateral arrangements of atoms. This may help to estimate the significance of such effects for the cases of reconstructed and/or strongly disordered surfaces. In addition, because the approximate three-beam treatment works for flat surfaces, it should be relatively simple to extend the theoretical work to rough surfaces (for example, by using perturbation methods). It should be possible also to develop an analytical treatment of intensity changes accompanying the beam emergence conditions.

In conclusion, we have shown that for flat surfaces it is possible to obtain an excellent quantitative and qualitative theoretical description of azimuthal plots and the key to this is the use of an accurate multiple scattering theory. The azimuthal plots may be useful for studying the fundamentals of RHEED and it should be possible to

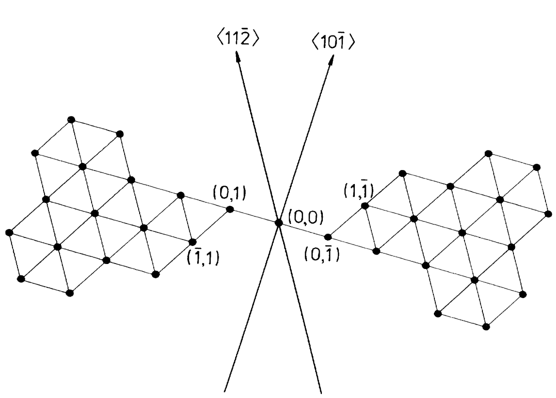

FIG. 2. The set of beams used in 35-beam calculations.

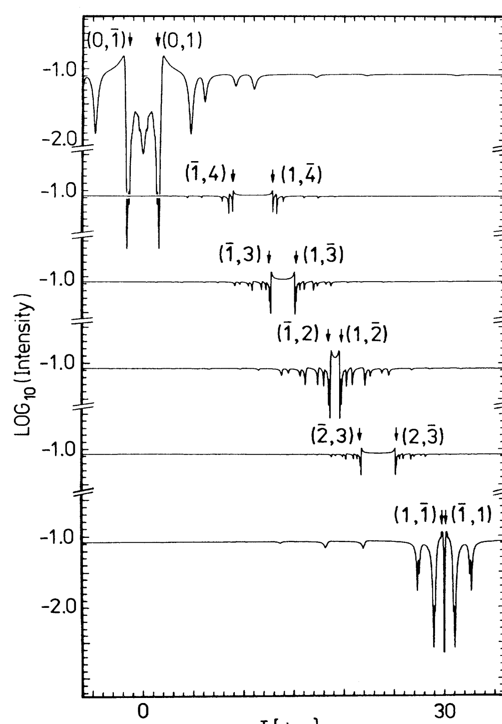

FIG. 3. Azimuthal plots obtained from three-beam calculations. The two side beams included in the calculation of each curve are given in the figure together with arrows indicating the beam emergence conditions.

exploit them in surface structural analysis.

It is a pleasure to acknowledge many helpful discussions with Professor J. L. Beeby and Dr. J. M. McCoy. One of us (Z.M.) is grateful for Royal Society financial support in the form of a one-year Postdoctoral Fellowship. This work was supported by the Computational Science Initiative of the United Kingdom Science and Engineering Research Council.

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