Ultramicroscopy 31 (1989) 241–244
North-Holland

SHORT NOTE

SELF-CONSISTENT SOLUTION OF THE REFLECTION DIFFRACTION PROBLEM

L.D. MARKS and Y. MA

Materials Research Center, Northwestern University, Evanston, Illinois 60208, USA

Received 2 June 1989

A general method of solving the reflection diffraction problem for high-energy electrons (and, in principle, also for X-rays) is described. The method exploits the multislice method of calculating electron diffraction in transmission through a crystal as an unconditionally convergent method of solving Schrödinger's equation for reflection electron diffraction. The method is self-consistent in that it necessarily converges with an internal consistency test. Examples are given for the use of the method to calculate RHEED patterns from the gold (110) 2×1 reconstruction and oxygen adsorbed on gold.

1. Introduction

Reflection high energy electron diffraction (RHEED) has a long history [1], but full exploitation of all the information available has not been possible due to the lack of good methods of calculating the diffraction patterns. Due to recent interest in reflection electron microscopy [2–4] and use of RHEED as an in-situ monitor of processes such as molecular beam epitaxy [5,6], theoretical efforts to understand RHEED have increased recently. For instance, Peng, Wang, Cowley and co-workers [7,8] have developed a method based upon the multislice method with an incident top-hat wave, Maksym and Beeby [9] a scattering approach with the slice parallel to the crystal surface, and Zhao et al. [10] a method using an invariant-embedding R-matrix scheme. In previous work [11,12], we have been using another approach which was based upon the Bloch wave method. In the process of this work and during efforts to combine the Bloch wave and multislice methods [13] an important theoretical result became apparent: the multislice approach to electron diffraction when used for reflection problems is, mathematically, identical to a Picard iteration [14] solution of Schrödinger's equation for the high-energy electrons. The multislice formulation due to Cowley and Moodie is given by:

$$\psi(\boldsymbol{q}, z_{n+1}) = \left[\psi(\boldsymbol{q}, z_n) \cdot P_g(\boldsymbol{q}, z_n)\right] * P_r(\boldsymbol{q}, z_n), \tag{1}$$

where P_g and P_r denote the phase grating and propagator respectively and $*$ denotes the convolution. When the crystal potential varies slowly in z axis on the range (z_0, z), (1) can be approximated as:

$$\psi(\boldsymbol{q}, z) = \left[\psi(\boldsymbol{q}, z) \cdot P_g(\boldsymbol{q}, z - z_0)\right] * P_r(\boldsymbol{q}, z - z_0). \tag{2}$$

The solution of (2) can be solved by the Picard iteration method. Since Picard iterations are unconditionally convergent, we therefore have, in principle, a general and simple method of solving for RHEED diffraction patterns; when a multislice RHEED calculation converges (as a function of thickness) the solution obtained is the true solution, ignoring any edge effects from the multislice numerical procedure.

In this note we present numerical results using this approach with the output of a Bloch wave program as the initial trial solution. These results demonstrate that this approach can be used to

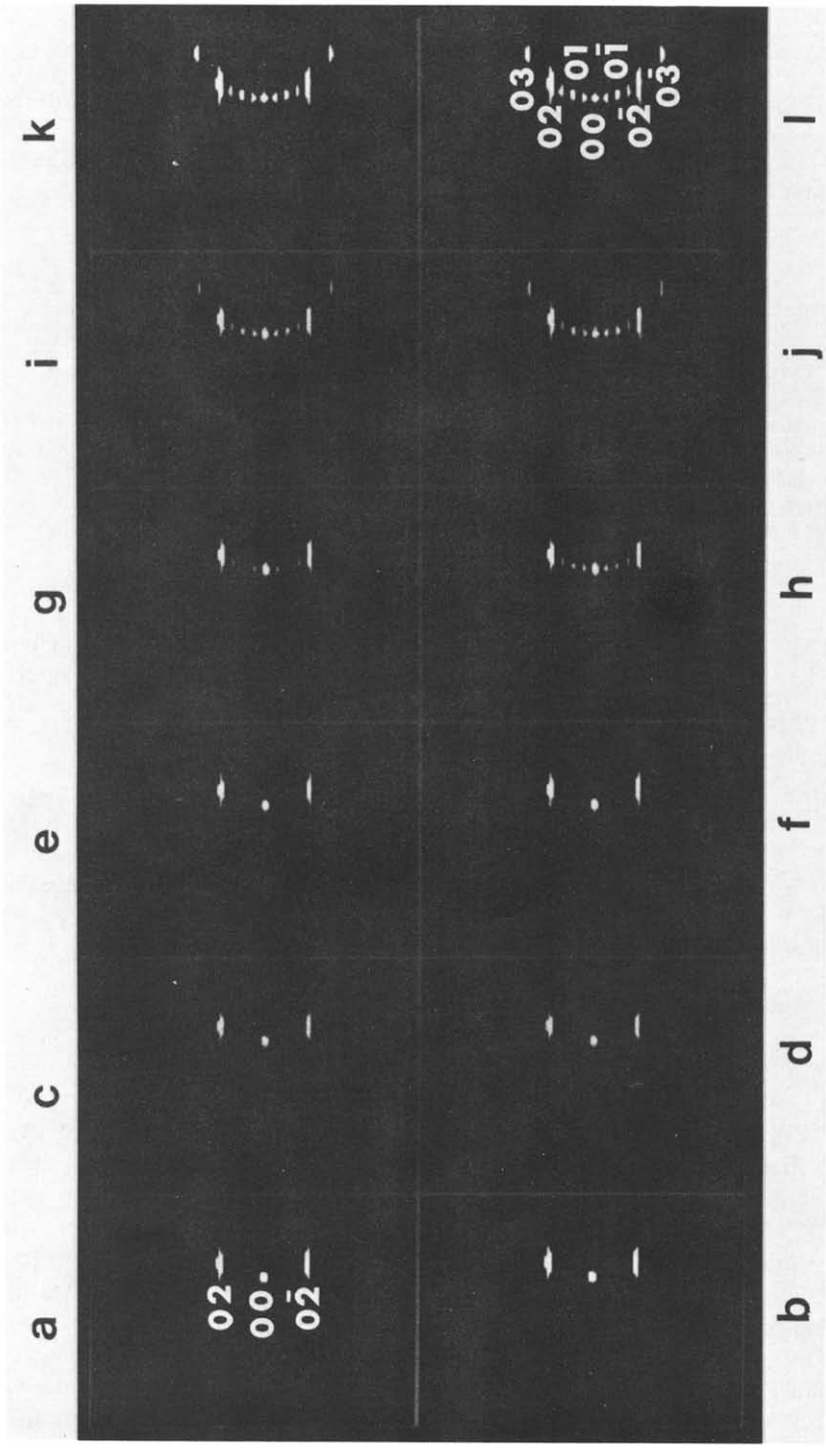

Fig. 1. RHEED patterns, excluding the incidence beam, for a 2×1 gold reconstruction. The thicknesses of the first and last slices are 0 and 536.8 Å. The thickness difference between two nearest slices is 50 Å. The beam incidence is 30 mrad.

calculate RHEED from reconstructed surfaces and surfaces with adsorbates without problems, which promises to open up interpretation of RHEED patterns. A more detailed presentation of these results is in preparation.

2. Numerical method

Our initial trial wave was calculated by using a Bloch wave approach as discussed elsewhere [11,12]; we do not have to use this trial wave but clearly the better the initial guess, the faster will be the convergence. Tests of the convergence of the solution for perfect surfaces have been presented elsewhere [13]; here we will only consider surfaces with reconstructions or adsorbates. This trial solution was then fed into a multislice program using, for the multislice, the potential of the modified surface. The output as a function of thickness was then analyzed to test the convergence of the method; in principle, this could be written into the program although we have not attempted this to date. All the results are for gold along [010] for a (001) surface and are for 100 keV electrons. The calculations were performed on an Apollo 3500 workstation (which uses a Motorola 68030 chip), and each took about 6 h of CPU.

3. Results

Fig. 1 shows calculated RHEED patterns, as a function of thickness for 5 nm increments, and fig. 2 shows the y-modulated RHEED pattern at the thickness of 607.2 Å, i.e. the last pattern in fig. 1, all for the 2×1 (001) reconstructed surface where, for simplicity, we have ignored any surface relaxations. It is apparent from both that by a thickness of about 60 nm the solutions have converged.

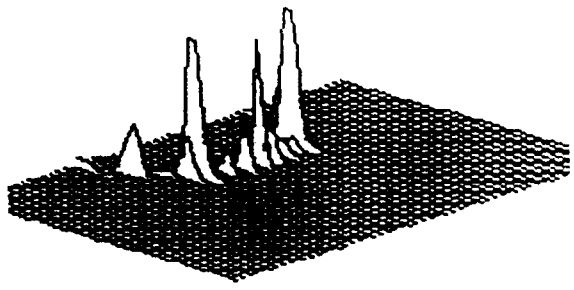

Fig. 2. Calculated y-modulated RHEED pattern for a thickness of 607.2 Å, i.e. the last pattern in fig. 1, all for the 2×1 (001) reconstructed surface where, for simplicity, the surface relaxation is ignored.

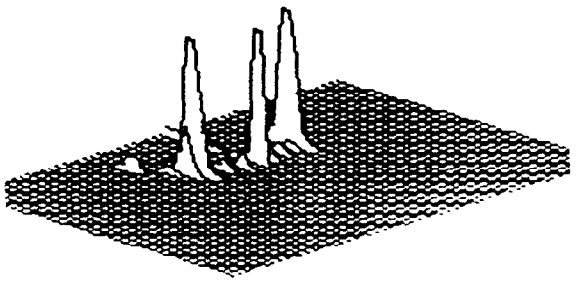

Fig. 3. Calculated y-modulated RHEED pattern where a 2×1 monolayer of oxygen is adsorbed on the surface, at the thickness of 607.2 Å.

As a second example, fig. 3 shows the calculated y-modulated RHEED pattern where a 2×1 monolayer of oxygen is adsorbed on the surface, which can be compared to the results in fig. 2 where there is a 2×1 monolayer of gold reconstruction. Whereas the effect is weak, it is apparent that the chemisorption can be detected.

4. Discussion

We have shown here that it is now realistic to calculate RHEED patterns from reconstructed or other surfaces using a method which is intrinsically self-consistent. There are some technical problems, and it would be wrong to ignore these although they are not intractable. Edge effects can lead to problems for thicknesses beyond about 60 nm in the multislice approach, and if the solution has not converged by this thickness, the approach will not work. The seriousness of the effects is also related to the incidence angle and the size of beam. However, since the method only requires a trial solution, there is no reason why one cannot patch together the wave in the vacuum and overcome these edge problems. To date we have been able to avoid these problems by manipulating the cell size for the calculation, and this may be the

easiest method to employ. The most important result is that we now can calculate RHEED using programs which are close to "turn-key".

Acknowledgement

This work was supported by the National Science Foundation through Northwestern University Materials Research Center, Grant No. DMR 85-20280.

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