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Theoretical studies on the quantitative interpretation of RHEED data

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Abstract. The importance of multiple-scattering effects for the case of reflection high-energy electron diffraction (RHEED) is demonstrated and discussed. Possible benefits from analysing two different forms of RHEED data (rocking curves and azimuthal plots) are investigated. The surface parameter sensitivity and conditions which should be satisfied when doing experimental work are examined in detail for flat surfaces. Methods to overcome possible difficulties in carrying out interpretations of data for surfaces with structural defects are discussed. It is concluded that for flat surfaces analysing RHEED rocking curves is usually most helpful, especially if one wants to extract very precise information about details of atomic arrangements at the surface. For simple characterization *in situ* of growing surfaces (usually with a number of structural defects) collecting and interpreting azimuthal plots seems to be more useful.

1. Introduction

Reflection high-energy electron diffraction (RHEED) is now widely used in experimental condensed-matter physics. It is applied by a variety of researchers whose scientific interests and aims are very different. Some are interested in the application of RHEED to get very detailed information about the arrangement of atoms at flat surfaces [1]. Their investigations can be classified as surface crystallography. Others use RHEED to monitor growth of ultrathin layers [2, 3]. In this case it is important to get information about changes in atomic arrangements at surfaces during the preparation of samples. Typically, sample surfaces in such cases cannot be considered flat so information both about their average roughness and about the arrangement of atoms within atomic islands is useful. Finally, it should be mentioned that a new, further application of RHEED has been demonstrated recently. The use of ultrafast laser pulses to drive a RHEED gun makes it is possible to collect information about the behaviour of atoms at the surface in picosecond periods of time [4].

Despite the popularity of RHEED, most of the data interpretations presented in the literature can be treated only as qualitative. This is due to the use of kinematical diffraction approaches which cannot be considered satisfactory for the description of the diffraction of electrons (this is true not only for the case of RHEED—earlier it was well recognized for low-energy electron diffraction and transmission high-energy electron diffraction). The use of kinematical approaches is, first of all, a consequence of their simplicity. However, there is also the fact that, for some time, precise theoretical analyses were presented in the literature for only relatively simple situations. So there existed a gap between realistic

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situations and those which could be considered theoretically. Precise approaches (dynamical diffraction theories) are based on the exact solution of the Schrödinger equation for scattered waves. Extensive and systematic theoretical work on RHEED started in the early eighties after the introduction by Maksym and Beeby [5] of a 2D Bloch wave approach. Following a period of development it is now possible to calculate and interpret results for surfaces directly related to experimental situations [6–11]. This opens the prospect that much more quantitative research can be undertaken using RHEED, which will, of course, require precise measurements and applications of multiple-scattering (dynamical) theory.

In this paper we investigate possible benefits from quantitative analyses of RHEED data. We consider different forms of data which are available through measurements of the intensity of the specularly reflected beam only. Although for the case of RHEED it is possible to measure and analyse the intensity of electrons scattered into different directions (i.e. to consider also the intensity of sharp spots for side beams and the intensity of diffuse scattering), specular beam intensity measurements are of particular value. Due to the equality of the incident and exit angles for the incident and specular beams, the position of the 'measurement window' at the screen can be determined easily even for surfaces with a large number of structural defects. There is no doubt about the possible usefulness of other experimental RHEED data. Important information, especially on details of surface reconstructions, can be extracted from side beam intensities. However, in practice, such data can be collected only for relatively flat surfaces. Concerning the diffuse part of scattering, recently Korte and Meyer-Ehmsen [7] have demonstrated that its interpretation may be very helpful when seeking precise information on surface phase transitions. However, in this case the analysis of experimental data can be carried out only with an extended version of the 2D Bloch wave approach software. Thus, it follows from the arguments presented above that it seems reasonable first to consider in detail possible benefits due to interpretations of specular beam intensity data. In this paper we demonstrate (section 2) that multiple-scattering effects are very important for the intensity of specularly reflected electrons although sometimes it is possible to reduce their significance. These initial considerations are rather qualitative. Next (section 3), we turn to a more precise examination of the possible usefulness of interpretations of RHEED data. We consider in detail rocking curves for main and 'random' azimuths (subsection 3.1), and azimuthal plots (subsection 3.2). The calculations are carried out for flat surfaces. However, we discuss the advantages of the forms of data presented for both flat and partially disordered surfaces (subsection 3.3).

We discuss in detail conditions which should be satisfied while recording experimental data. Currently, most RHEED set-ups are installed as parts of MBE systems. Quite often these set-ups cannot be used to collect data suitable for quantitative analyses. Unfortunately, making any improvements in MBE systems, after delivery from commercial companies and installation in laboratories, is extremely inconvenient and expensive. Thus, we hope that the considerations presented in this paper will be helpful when taking decisions during the planning of research work and the ordering of equipment.

2. Multiple-scattering effects

2.1. The geometry of RHEED measurements and the structure of $DySi_{2-x}$

Figure 1 shows a typical RHEED geometry. In this paper we deal with two methods of collecting data [12]: rocking curves and azimuthal plots. A rocking curve is a set of intensities of a diffracted beam, measured while the polar angle θ is varied (the azimuthal angle ϕ is fixed). An azimuthal plot is a set of intensities of a diffracted beam, measured

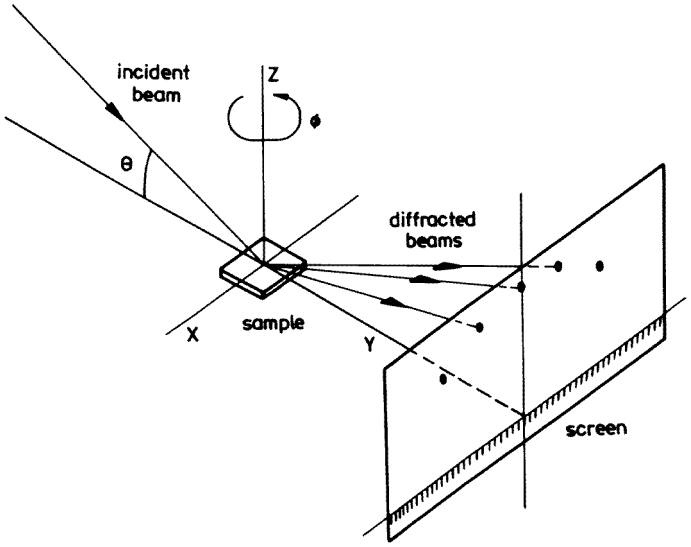

Figure 1. The geometry of RHEED measurements.

while the azimuthal angle ϕ is varied (the polar angle θ is fixed). The beam referred to could be the specular beam or any one of the side beams. As has already been discussed in section 1, in this paper we deal only with intensities of the specular beam. However, it should be stressed that the side beams also contain information about the surface and if at all possible their intensities should be measured to increase the amount of data available. Further, it seems useful to remind readers not familiar with electron diffraction literature that physicists dealing with x-ray diffraction would refer to sets of data analogous to the specular beam RHEED rocking curves as θ–2θ diffraction patterns.

To complete the discussion of the RHEED geometry we would like to add some comments. The explanation presented above on rocking curves and azimuthal plots, and the results presented later in the paper, concerns the case for which it is assumed that the incident electron beam does not diverge (in the following we will call this case standard RHEED). In other words, it is assumed that all partial incident electron waves propagate along exactly the same direction. Although in practice it is impossible to satisfy this assumption perfectly, it is expected that in actual experimental work the effort is taken to make the beam divergence as small as possible. It is worth mentioning that there exist in the literature papers on so-called convergent-beam reflection high-electron-energy diffraction (CB-RHEED) which relate to another approach. That work is concerned with RHEED intensities for the case of when the incident beam is formed from non-parallel sub-beams which are focused at the surface of the crystal. The basic idea behind this approach is an attempt to extend the amount of information contained in a single pattern observed at a screen. For example, for perfectly flat surfaces, in the case of CB-RHEED regular discs are observed at the screen in contrast to the sharp spots which are typical for standard RHEED. In fact, each incident sub-beam direction for CB-RHEED corresponds to a fixed incident beam (with properly chosen values of azimuthal and polar angles) in conventional geometry (i.e. without any divergence). Nevertheless, the interpretation of CB-RHEED experimental data can be done in a very similar manner to that used for the case of standard RHEED data. This is because in principle one CB-RHEED pattern contains the same information as a series of standard

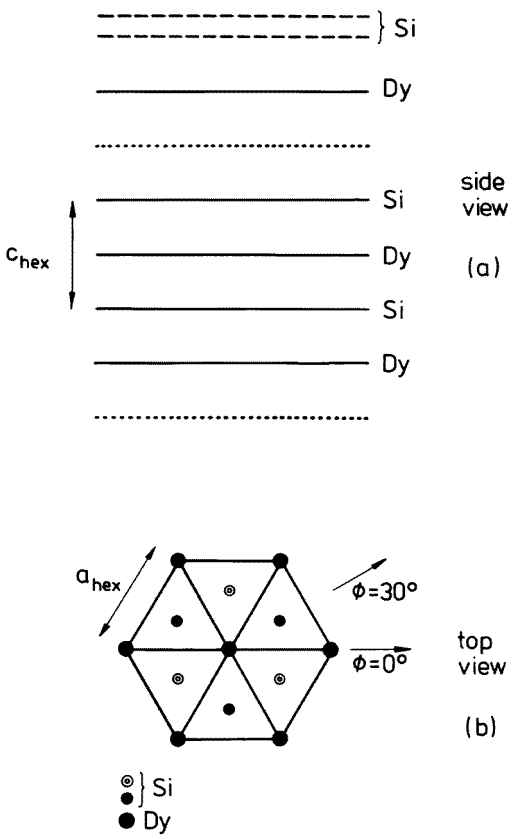

Figure 2. The structure of $DySi_{2-x}$: (a) side view, (b) top view. The values of the lattice constants are [17]: $a_{hex} = 3.831$ Å, $c_{hex} = 4.121$ Å.

RHEED rocking curves or azimuthal plots. CB-RHEED practical work was first done by Ichimiya *et al* [13] and currently we can observe a slow but continuing progress in its development [14, 15]. The shortcoming of this approach arises from its instrumental requirements. To form the incident beam properly a sophisticated experimental arrangement is necessary. Nevertheless, the basic concepts of CB-RHEED and the results achieved so far look interesting.

The structure of $DySi_{2-x}$, the material which we use as an example, is shown in figure 2. It is expected that its surface reconstruction is relatively simple so that it is well suited to the purposes of this theoretical work. Additionally, it can be noted that there is currently great interest [16] in the properties of silicides of rare-earth metals (including yttrium). $DySi_{2-x}$ thin layers present the AlB_2-type structure which can be described using a hexagonal crystallographic lattice [17]. Along the axis perpendicular to the surface there are alternate planes of Dy and Si atoms. In the Si planes there are two sublattices. It is accepted that, for all rare-earth metal silicides having a structure of the AlB_2 type, one of the Si sublattices is only partially filled [18]. We assume that it contains 67% of the possible Si atoms. We consider two models of the surface: (1) the bulk-terminated structure with Si atoms at the top, (2) one of the two Si sublattices (the one which is partially filled) is moved upward by a distance of 0.8 Å. Only the second model is though to be realistic,

having been proposed by Baptist *et al* [18] for YSi_{2-x}, a material which is structurally very similar to $DySi_{2-x}$. The structure of $DySi_{2-x}$ shown in figure 2(a) is terminated according to this model.

2.2. The method of calculations of RHEED intensities

The calculations presented in this paper were carried out using dynamical diffraction theory. This means that the results are obtained by the numerical solution of the Schrödinger equation subject to a number of approximations. It should be pointed out that the introduction of any approximations concerning the potential of a crystal must be done with great care. Such approximations should be both properly related to realistic situations and should allow one to carry out numerical calculations as quickly as possible. These are often contradictory features and finding the proper balance has been an important achievement during the development of RHEED. It seems that the most efficient choice for the case of RHEED is to assume two-dimensional periodicity of a crystal potential in planes parallel to the crystal and to allow arbitrary variation of the potential along the axis perpendicular to the surface. Formally, this condition makes it possible to expand an electron wave function in the form of a series of 2D Bloch waves. This allows one to reduce solving the 3D Schrödinger equation to solving a set of ordinary differential equations. Numerical algorithms and computer codes based on the use of 2D Bloch waves were first developed by Maksym and Beeby [5], and Ichimiya [19]. More theoretical information behind this approach can be found in a review article by Beeby [20]. There are now several numerical programs developed within the 2D Bloch wave approach and a program [21] of this type has been used to carry out the calculations presented in this paper.

We would like to discuss two practical problems which must always be solved in some way before carrying out detailed calculations of RHEED intensity.

The first problem is the proper determination of the set of 2D Bloch wave terms to be taken into account in the calculations (typically in the literature on RHEED these terms are named beams). Formally, in the mathematical equations, an infinite 2D Bloch wave series occurs. However, the series must be truncated in order to perform numerical work. The time of the computations is proportional to n up to 3, where n is the number of beams taken into account. It seems to be a commonly accepted fact that calculations for the number of beams exceeding 100 are time consuming. In fact, this number should be treated as a rough estimate only since the real time needed to carry out the calculations depends strongly on the level of sophistication and optimization of the computer code applied, the crystal structure investigated and the type of computer used. It is worth mentioning that many possible speed improvements which may be included in a RHEED program have been discussed in detail very recently by Maksym [22]. Nevertheless, the question of how many beams (or to be more precise, which of them) should be included in computations continues to be of great practical importance. Earlier, Tong and co-workers [23, 24] stated that, even for materials with relatively simple surfaces, the number of beams taken into account should be quite large. In their papers they used sets of about 40–140 beams. However, subsequently Meyer-Ehmsen [25] and Ichimiya [26] showed that while the findings presented in [23, 24] are correct in a purely numerical sense, the beam set to be considered may be substantially reduced if one takes account of the finite precision of RHEED measurements and if one omits pairs of beams partially cancelling each other (for more details see [25, 26]). Nevertheless, the choice of the set of beams must be always made with great care and carrying out some trial computations is usually unavoidable in the early stage of work with new materials.

The second practical problem which must be solved before computing RHEED

intensities is the unique choice of a procedure for determining the crystal potential. For the case of RHEED it is usual to assume that the Schrödinger equation contains a complex scattering potential. Numerically, the determination of the potential is realized in a sequence of steps. The real part is initially calculated by summing up individual contributions from crystal atoms using electron scattering factors for isolated atoms, modified using Debye–Waller factors to take into account thermal vibrations in the crystal. The data on electron scattering factors which are mostly used are those published by Doyle and Turner [27]. However, there are cases for which reference [27] does not provide sufficient information (for example, factors of some elements are not included in the tables of [27]). Other possible sources of data are tables which have been published by Jiang and Li [28], and very recently by Dudarev *et al* [29]. Returning to the procedure for the determination of the scattering potential, the volume average of the real part is further corrected by adding some constant value (usually of order of 1–2 eV) to reflect the fact that the distribution of outer electrons in crystal atoms is different from that in isolated atoms. Concerning the imaginary part of the potential, its presence reflects the occurrence of inelastic and diffuse scattering in a crystal. There are two different possible methods for the determination of the potential imaginary part. Both of them were developed originally in the field of transmission high-energy electron diffraction (THEED). The first approach (called in the following the 'proportional' model) is to assume the imaginary part of the potential to have the form of the real part of the potential reduced by some factor. In theoretical work this factor is usually assumed to have a value between 0.05 and 0.2; in work related to experiments it is often treated as an adjustable parameter. This method was introduced (originally in a slightly different form) by Hashimoto *et al* [30]. In fact, it expresses a phenomenological approach to the problem. The other method is to model the imaginary part based on actual physical processes in a crystal. Many aspects of such modelling are reviewed in a paper of Howie and Stern [31]. According to their work there are three basic contributions to the imaginary part of the potential. Respectively, they are due to: (1) electronic excitations, (2) thermal diffuse scattering, and (3) structural defects. One may expect that precise estimates of the quantities coming from these three contributions cannot be obtained simply. In fact, it turned out that even for the same experimental situations different authors could come to noticeably different final estimates (for details, see the discussion presented in a paper of Ritchie and Howie [32]). Nevertheless, so far the problem of the determination of the imaginary part has not been considered difficult to deal with by those carrying out dynamical calculations of RHEED intensity. To explain this situation it seems important to recall some general findings concerning results of such calculations. It has been observed that only variations of the average of the imaginary part influence the calculated intensity substantially, while variations of parameters describing its detailed shape cause only small changes in the output results (see, for example, [23, 24]). This is why, currently, for the case of RHEED, the use of the simplified 'proportional' model seems to be much more popular among researchers than the use of detailed imaginary potential components which are physically much more soundly based, but actually require that a number of difficulties be overcome. However, continuing progress in the development of the RHEED technique allows one to consider more and more detailed questions. So, it may be interesting to reinvestigate, in the future, the treatment of the problem of absorption. However, in our opinion research of this type should be done in connection with the interpretation of data coming from actual measurements. Detailed potential modelling requires a rather large effort and this is why it may be beneficial to start such work from trial checks of how large the differences due to possible changes of the shape of the imaginary part are, compared with the level of precision actually achieved in the measurements. To give some insight

into the problem we would like to mention some results from papers published so far. For the case of THEED, recently, a particular example has been found [33] for which the use of the simplified 'proportional' model to describe the experimental data fails, while detailed modelling is successful. Unfortunately, due to the very fine nature of the effects caused by the use of different shapes of the potential imaginary part, it is not possible to simply apply findings from THEED to the case of RHEED. Detailed modelling of the imaginary potential directly for the case of RHEED has been already employed in a few papers (for example [25, 34]). Analysing experimental data for Pt(111) in a strictly quantitative way (with the use of an advanced, automatic procedure for fitting data), Stock and Meyer-Ehmsen found that the detailed shape of the potential imaginary part is of minor importance [35]. In other words, their results support the use of the 'proportional' model for the case of RHEED. However, as stated above, because of substantial, recent progress in the development of RHEED, one may find it important to reexamine, in further research, the determination of the imaginary part of the potential. In the context of the possible interest of some researchers in detailed modelling, it seems worthwhile to mention that Dudarev *et al* [29] recently published tables containing coefficients which allow one to express the thermal inelastic scattering contribution (determined with the use of the Einstein model) in the convenient form of a sum of Gaussian functions.

All calculations of this work are carried out for an incident electron energy of 18 keV. We have used 37 electron beams (i.e. 37 terms of the 2D Bloch wave series). However, we used two different sets of 37 beams: one for calculations with the azimuthal angle ϕ in the neighbourhood of 0°, the other related to the neighbourhood of 30°. The results presented in this paper are convergent in the sense that including further beams does not alter them significantly and has no influence on conclusions. The scattering potential was computed in two stages. First, the real part of the potential was constructed using the electron scattering coefficient for Dy and Si atoms tabulated by Jiang and Li [28], assuming thermal vibrations typical for these atoms at 300 K. The imaginary part of the potential was assumed to be 0.15 of the real part. In the second stage, both the real and imaginary parts were relativistically corrected [36]. Further, a constant term of 2 eV was added to the real part in order to take into account some deformation of the distributions of outer electrons in atoms of the crystal [37]. After the determination of the potential, the value of the electron energy was modified to take into account relativistic effects [36]. The results that we present are not qualitatively dependent on these assumptions about the potential.

2.3. The dynamical nature of RHEED diffraction

Figure 3 shows a set of rocking curves calculated for different values of the azimuthal angle ϕ. The curves shown with solid lines are the results of calculations for the full 3D scattering potential. Curves shown with crosses (see parts (b), (c) and (d) of figure 3) were calculated for a simplified 1D potential. This potential was obtained after averaging the full potential in planes parallel to the surface. The operation of averaging of this type is very simple if one uses the properties of Fourier series. Calculations for the 1D scattering potential are often called one-beam calculations [38, 39]. This other name follows from the use of the formalism of the 2D Bloch wave approach. In one-beam calculations only the first term of the 2D Bloch wave series is considered. It follows from the simple analysis that taking into account only one term (beam) is equivalent to assuming the 1D potential of the crystal.

To get a general understanding of the shapes of the solid curves presented in figure 3, let us consider qualitatively diffraction in a crystal layer. If we look along the direction of the incident electron beam we can observe, depending on the chosen azimuth, different

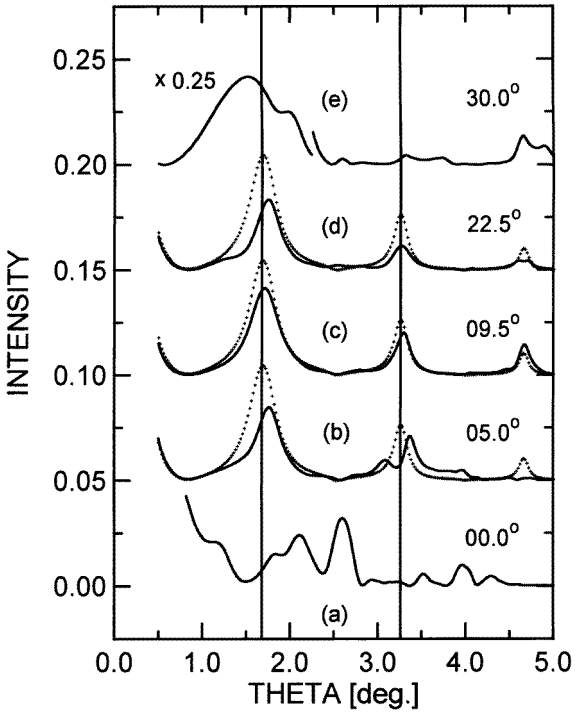

Figure 3. A set of rocking curves calculated for different azimuths for bulk-terminated $DySi_{2-x}$: (a) $\phi = 00.0^\circ$, (b) $\phi = 05.0^\circ$, (c) $\phi = 09.5^\circ$, (d) $\phi = 22.5^\circ$, (e) $\phi = 30.0^\circ$. The solid plots are for many-beam calculations; plots shown with crosses are for one-beam calculations. Two vertical lines, for 1.68° and 3.26°, are drawn in the figure to point out peak maxima found in one-beam calculations.

images of the arrangement of atoms in planes parallel to the crystal. For the main azimuths of the crystal layer we can observe very regular rows of atoms; for other azimuths the arrangement of atoms seems to be more or less 'random'. On the basis of this simple observation we can expect that if we chose the azimuth of the incident electron beam to be one of the main azimuths of the crystal layer, strong diffraction effects should occur due to movement of electrons in 'regular channels'. For other azimuths we should expect partial cancellations of effects occurring due to a lateral modulation of the potential. Figure 3 demonstrates that these qualitative considerations are in a good agreement with the results of the calculations. The shapes of curves calculated for the full 3D potential for azimuths far from the main azimuths (solid curves at figure 3(b)–3(d)) resemble the shapes of one-beam rocking curves (curves shown with crosses). Figure 3(b) represents a typical situation—despite the overall similarity there are also a number of differences in details. The solid curves shown in figure 3(c) and figure 3(d) represent cases specially chosen by us when the degree of similarity is relatively large. However, even in these cases some differences can be recognized—for example shifts of about 0.1° occur for some peaks. On the other hand rocking curves calculated for the main azimuths (figures 3(a) and 3(e)) are very complicated and their shapes are very different from those of curves for the 1D potential. Curves of this type were discussed in the literature many times and generally it is accepted that a simple qualitative description is not possible for them. Sometimes it is possible to find some diffraction effects which dominate (for example resonance effects for Pt(111) [40]).

However, in general, the shapes of rocking curves for the main azimuths are the result of the interference of many diffraction contributions and it makes them effectively unpredictable without real measurements or full dynamical calculations.

One-beam rocking curves, i.e. calculated for the 1D potential, do not depend on the azimuthal angle ϕ—this is a consequence of the determination of the simplified potential. They resemble patterns which result from calculations based on the use of kinematical diffraction theory. Actually, the positions of the peaks of the one-beam rocking curves can be determined roughly by using the Bragg law and making corrections for the refraction effects due to the difference between the scattering potential of the crystal and vacuum. However, even 'improved kinematical theory' (i.e. considering the refraction) cannot be used to describe exactly the shapes of one-beam rocking curves. The kinematical theory considers only single-scattering events, while in dynamical calculations for the 1D potential some multiple-scattering effects are included. More precisely, the kinematical theory assumes that backward scattering from the crystal can be determined by simply summing small single-scattering contributions due to individual atoms—this means that electron beams appearing because of diffraction are assumed not to be further scattered. In dynamical calculations for the 1D potential, multiple scattering of electrons between planes parallel to the surface is taken into account.

The aim of subsection 2.3 was to demonstrate the importance of dynamical effects for RHEED and to give qualitative explanations of different situations. Users of RHEED are interested in getting some information about their samples with the help of this technique. Qualitative understanding of RHEED is important for them and may enable them to obtain general insight concerning the arrangement of atoms in samples under investigation. However, the most valuable information can usually be extracted on the basis of precise quantitative analysis. In the further part of the paper we examine rocking curves and azimuthal plots from this point of view.

3. Analysis of RHEED data

3.1. Analysis of rocking curves

3.1.1. Rocking curves for main azimuths. Figure 4 shows three rocking curves calculated using the full scattering potential. We assumed in the calculations: (a) the bulk-terminated structure with Si atoms at the top and the azimuthal angle fixed to be 30° (see figure 2(b)), (b) the same structure, but with the azimuth of the incident beam changed by 0.1° and (c) the surface structure as in the Baptist *et al* model [18] with the incident electron beam along the main azimuth again. This set of curves allows us to compare changes in shapes due to possible errors of measurements with changes due to the surface reconstruction.

The changes due to the surface reconstruction are large and this is why it seems quite obvious that rocking curves can be very useful in surface structure determination. However, the 'error' of 0.1° assumed for the azimuthal angle influences intensities noticeably. We can probably consider the value of 0.1° as a limit of the error which is acceptable if one wants to ignore its consequences in an analysis. The acceptable error may actually differ for different materials and may depend on the precision of the analysis. The value given here should be treated as a general estimate which may be helpful for planning quantitative work on RHEED. The value of 0.1° seems to be larger than the divergence of the incident and reflected electron beams if high-quality RHEED equipment is used and the sample surface is flat [9]. Also, it seems that if special care is taken while changing the glancing angle θ experimentally it is possible to neglect changes in the intensity due to small but unavoidable

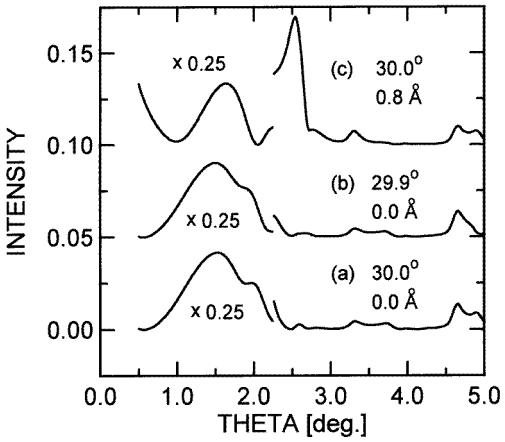

Figure 4. Many-beam rocking curves: (a) the bulk-terminated structure, $\phi = 30.0^\circ$, (b) the bulk-terminated structure, $\phi = 29.9^\circ$, (c) the Baptist *et al* model [18] of the surface, $\phi = 30.0^\circ$.

variations of the azimuth of the incident beam. However, it should be said that satisfying the condition given above is not very simple and anyone interested in real measurements should consider it in detail before doing experimental work.

3.1.2. Rocking curves for one-beam conditions. Here we discuss possible applications in analysis of one-beam rocking curves. One-beam calculation results for the bulk-terminated structure are shown in figure 5(a) and for the surface reconstructed according to the Baptist *et al* model [18] in figure 5(c). The crucial problem for one-beam rocking curves is how to measure them for real samples. There does not exist an experimental procedure for collecting data which would be fully analogous to calculations for the potential averaged in planes parallel to the surface. It is believed that this may happen for some 'random' azimuths. Ichimiya, who actually first introduced this type of rocking curve, gave some empirical advice on how to determine one-beam conditions by employing observations of Kikuchi features at the screen [39]. He also verified his predictions computationally [39]. Using his findings for Si(111) and the similarity of atomic arrangements in planes parallel to the surface for $DySi_{2-x}(0001)$ and Si(111) we can suppose that in our case we should get such a condition for an azimuthal angle ϕ of 22.5°. The curve presented in figure 5(b) was obtained using 37 beams and we can consider it as a simulation of experimental measurements. We assumed the case of the bulk-terminated structure.

The difference between curves of figure 5(a) and figure 5(b) represents the error of the one-beam calculations with respect to the full calculations. This difference is quite significant from a purely quantitative point of view. Many-beam effects influence positions of peaks (for example, for the curve shown in figure 5(a) one of the maxima appears at 1.68°, for the curve of figure 5(b) the similar maximum is observed at 1.76°). This means that results of analyses based on the use of one-beam calculations should always be treated with great care.

However, if one takes proper care while using the results of one-beam calculations it seems that in some situations useful answers may be obtained. If we compare the curves presented in figure 5(a) and figure 5(c) we can observe that the surface reconstruction may influence curve shapes noticeably. This means that one-beam rocking curves can be useful

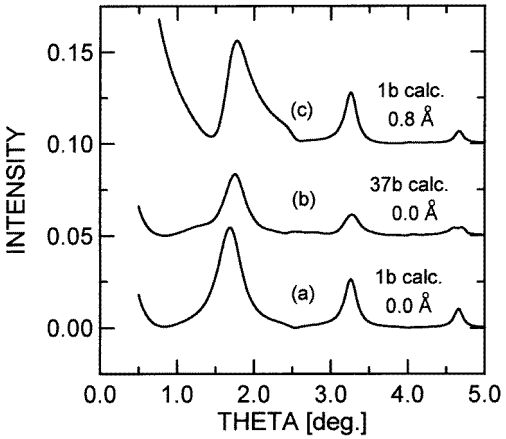

Figure 5. Rocking curves: (a) the bulk-terminated structure, one-beam calculations, (b) the bulk-terminated structure, many-beam (full) calculations for the azimuthal angle $\phi = 22.5^\circ$, (c) the Baptist *et al* model [18] of the surface, one-beam calculations.

in surface parameter determination. It is important that appropriate calculations can be carried out practically 'on-line' with present-day computers. One-beam rocking curves are sensitive to variations of surface parameters only along the axis perpendicular to the surface (this is a consequence of taking into account only the 1D potential in such calculations). Thus, analyses with the use of one-beam calculations can be done quickly and variations of surface parameters in planes parallel to the surface are eliminated from consideration, which sometimes may be very helpful. One might expect that such analyses would be much more accurate for materials composed of light atoms (for example Si) rather than for those composed of heavy atoms (for example Pt). It is known for RHEED that the importance of many-beam effects depends on the electron scattering factors of crystal atoms; these factors are relatively weak for elements with low values of the atomic number.

3.2. Analysis of azimuthal plots

Azimuthal plots of the specular beam have the strong virtue that they can be relatively easily collected in UHV chambers designed for the preparation of ultrathin films [3, 10]. These measurements are realized by recording the specular beam intensity during the rotation of a sample around the normal to the surface. However, a problem appears if one wants to compare such experimental plots with those computed. This is the problem of determining a value for the glancing angle which should be assumed in calculations. The value at which measurements of azimuthal plots are carried out can be known for experiments with a precision of about ± 0.1–0.2° (for RHEED the determination of the absolute value for the glancing angle is much more difficult than for its relative variation). On the other hand the value of the average volume potential for the crystal can be usually predicted up to ± 2 eV [37]. This means in practice that the polar angle positions of maxima and minima of the intensity of the specular beam can be theoretically predicted with a precision of about $\pm 0.1^\circ$ (more precisely, the statement that small variations of the average potential cause shifts of intensity maxima and minima should be treated as only approximately true). If we combine these uncertainties together we reach the conclusion that, in principle, experimental azimuthal plots should be compared with a set of plots calculated with glancing angles θ

in the range of about $\pm 0.3^\circ$ around the value determined experimentally.

One solution to this problem is to treat the value of the glancing angle as a free parameter to be determined during the theoretical interpretation of experimental data. Such a solution can be considered quite acceptable, although it is obvious that sometimes it may cause some practical inconvenience.

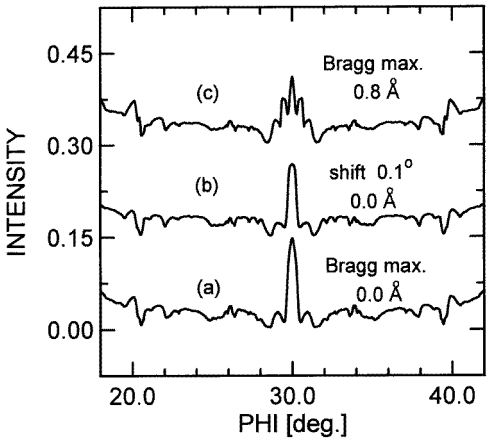

Figure 6. Azimuthal plots: (a) the bulk-terminated structure, $\theta = 1.68^\circ$ (the Bragg maximum), (b) the bulk-terminated structure, $\theta = 1.78^\circ$ (0.1° away from the Bragg maximum), (c) the Baptist *et al* model [18] of the surface, $\theta = 1.78^\circ$ (the Bragg maximum).

There is another solution which we use in this paper. We observe that if azimuthal plots were measured at Bragg reflections and were compared with azimuthal plots calculated at corresponding Bragg maxima we could practically eliminate the problem. The experimental and calculated values of the glancing angle would match automatically. Theoretically, Bragg reflections for a fixed model of the surface can be determined as maxima in the one-beam rocking curve (or in other words maxima in the rocking curve calculated for the potential averaged in planes parallel to the surface). Unfortunately, as was discussed in subsubsection 3.1.2, there is no experimental procedure allowing anyone to carry out measurements fully corresponding to one-beam calculations—it can be done only approximately. Nevertheless, from figure 3 and figure 5 it follows that it is possible to determine experimental Bragg maxima corresponding to maxima from one-beam calculations with a precision of about $\pm 0.1^\circ$. In any case, if we decide to accept even such an approximate determination of experimental Bragg reflections, we can reduce the set of azimuthal plots which we should consider theoretically. It should be mentioned that there is also some ambiguity (which is easy to handle) in the theoretical determination of Bragg reflections. The exact positions of the maxima depend on the surface models used in the calculations. This means that for each reconstruction model these positions should be found independently (more precisely, for each model a one-beam rocking curve should be calculated and from this curve Bragg maxima should be extracted).

There is no evidence that one of these two solutions to the problem of matching the values of the glancing angle is much better than the other. In this paper we prefer the solution based on the use of the concept of Bragg reflections, since it relates to basic ideas of diffraction.

Figure 6 shows the results of calculations carried out to investigate the sensitivity of

azimuthal plots to changes of surface structural parameters (figures 6(a) and 6(c)) and changes of the glancing angle (figures 6(a) and 6(b)). The azimuthal plots presented in figure 6(a) and figure 6(c) were calculated at Bragg positions for two different models of the surface: the bulk-terminated structure and the Baptist *et al* model [18]. We can observe that important changes of plot shapes appear only around the main azimuth (this corresponds to the value of 30° for the azimuthal angle in figure 6). These parts are surface parameter sensitive. The other parts of the plots seem to be related to bulk diffraction effects (the bulk for RHEED means usually the fragment of the crystal contained approximately between the fourth and tenth monolayer). The plots presented in figure 6(a) and figure 6(b) give us information on how changes of the glancing angle influence the intensity. The differences between the plots may be considered small and sometimes they might be ignored. Nevertheless, they are quite noticeable and in a precise analysis they should not be neglected. Because even the use of the concept of Bragg reflections does not allow one to determine experimentally the glancing angle better than with a precision of about $\pm 0.1^\circ$, this means that, for the calculated azimuthal plots, glancing angle variation effects should always be considered in any precise analysis of data. Additionally, from the comparison of the plots of figure 6(a) and figure 6(b) we can conclude precisely how a sample should be rotated while collecting azimuthal plots. It seems that the acceptable precision of keeping the surface parallel to an initially determined geometrical plane should be of order 0.05° if one is to neglect completely errors in the intensity. To get such precision it may be helpful to observe the left and right parts of an azimuthal plot for symmetry azimuths. Experimental work done recently has shown that in practice this can be realized quite well [10].

3.3. Discussion and conclusions

The calculations presented in subsections 3.1 and 3.2 were carried out assuming ideally flat surfaces. Comparing the possible advantages of interpretations of rocking curves and azimuthal plots, one can conclude that those interested in getting very detailed information on surface reconstructions may get more value from measurements and interpretations of rocking curves than from azimuthal plots. Examples presented in the literature of the analysis of rocking curves measured for both the specular and side beams (to increase the amount of data) look very promising [6, 9]. Concerning azimuthal plots, there may exist some limitations on extracting information on a sample as only some parts of the plots are surface parameter sensitive. Additionally, in very precise analyses of azimuthal plots it is impossible to avoid the treatment of the glancing angle as a free parameter to be determined. Nevertheless, it seems that both rocking curves and azimuthal plots may be quite useful in extracting details of structural arrangements of atoms at flat surfaces. For example, azimuthal plots may be helpful if there are apparatus limitations on measurements of rocking curves at high glancing angles or for verifications of the results of rocking curve interpretations.

The situation is different if we assume that there is some structural disorder at the surface (for example if there exist atomic islands, vacancies etc). RHEED is widely used to monitor the epitaxial growth of thin films, so we can assume that structural disorder is very typical as there are usually many defects at growing surfaces. Very often it is necessary to verify quickly whether the structure being prepared is the one desired. In such situations it is important to have some information both about the roughness at the surface and about the arrangement of atoms within large islands. To carry out structural characterization of this kind azimuthal plots may be more profitable than rocking curves. As noted earlier, rocking curves for the main azimuths of crystals have generally unpredictable shapes because they

are determined by many simultaneous diffraction processes. This makes them very sensitive to changes in surface parameters. However, this does not allow one to predict simply how the effects of surface disorder influence their shapes. The shapes of rocking curves measured for a one-beam condition are much easier to use for predictions. Unfortunately, the amount of information which can be extracted, by analysis of one-beam rocking curves only, is too little to draw complete conclusions about sample surfaces. Concerning azimuthal plots, as noted earlier, some parts of them are surface parameter sensitive while others are determined mainly by bulk diffraction effects and even a large number of structural defects at the surface do not destroy the appearance of the latter in measured plots [3, 10]. Generally, it can be expected that the increase in the number of defects causes a gradual disappearance of local maxima and minima in the parts of azimuthal plots for which the shapes are determined by bulk diffraction effects. This allows one to extract qualitative information about roughness at the surface. If it can be supposed that a surface is relatively flat, the surface-parameter-sensitive parts of azimuthal plots may be used to verify arrangements of atoms within atomic islands [41]. Thus, analysis of RHEED azimuthal plots should allow one to characterize samples during their preparations.

It can also be concluded that anyone who is interested in collecting experimental data suitable for quantitative analyses should operate a RHEED set-up which enables easy variations of polar and/or azimuthal angles of the incident electron beam. Further, only an incident beam divergence (i.e., the full width at the half-maximum) of less than 0.1° can be considered acceptable.

The considerations in this paper were based solely on numerical results. This means that further applications of RHEED to experimental work may introduce additional features. It is especially difficult to reach precise conclusions when considering surfaces with structural defects, since elements of speculation inevitably occur. However, the aim of this paper was to summarize and justify in a simple form basic information on rocking curves and azimuthal plots and to point out the most important conditions which should be satisfied experimentally when doing quantitative work on RHEED.

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