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Theoretical analysis of reflection high-energy electron diffraction (RHEED) and reflection high-energy positron diffraction (RHEPD) intensity oscillations expected for the perfect layer-by-layer growth

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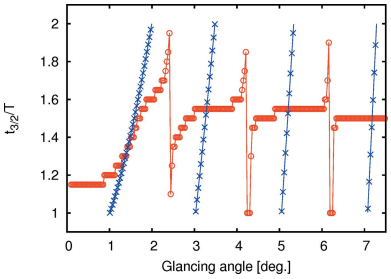
Predictions from two theoretical models, allowing one to determine the phase of intensity oscillations, are compared for reflected beams of electrons and positrons. Namely, results of the precise dynamical calculations are compared with results obtained using a simplified approach. Within the simplified model, changes in the specularly reflected beam intensity, expected to occur during the deposition of new atoms, are described with the help of interfering waves and the effect of refraction, and respective approximate analytical formulas are employed to determine the phase of the oscillations. It is found that the simplified model is very useful for understanding the physics ruling the appearance of intensity oscillations. However, it seems that the model with the realistic potential is more suitable for carrying out interpretations of experimental data.

1. Introduction

Reflection high-energy electron diffraction (RHEED) is a relatively old technique used to investigate surfaces of crystalline solids (Ichimiya & Cohen, 2004; Peng *et al.*, 2004). However, it is interesting to note that the theory of RHEED is still incomplete. In fact, experimental data can be precisely interpreted mostly for flat surfaces. For flat samples, a two-dimensional Bloch wave approach (Maksym & Beeby, 1981; Ichimiya, 1983; Zhao *et al.*, 1988) can be used, or alternatively, it is possible to apply a properly modified (Peng & Cowley, 1986; Ma & Marks, 1992) multislice method originally developed for the case of transmission geometry. However, for surfaces with steps, islands *etc.*, quantitative theoretical treatment is available only for special cases. The case worth mentioning is the growth of thin layers with the accompanying appearance of three-dimensional small islands at the surface. Then transmission diffraction features are observed on the screen and advanced analysis of experimental data is possible (Wang & Lu, 2014).

Reflection high-energy positron diffraction (RHEPD), in comparison to RHEED, is a relatively new technique, developed mainly in Japan (Fukaya *et al.*, 2013, 2014). It is interesting that respective dynamical calculations can be executed for both techniques using the same computer program (Ichimiya, 1992). Thus, it seems valuable to discuss theoretical results for RHEED and RHEPD together.

The goal of this paper is to analyse theoretically the effect of RHEED and RHEPD intensity oscillation. RHEED oscilla-



tions can be actually recorded experimentally in some situations. This occurs if thin films of the material are precisely grown (*e.g.* using molecular beam epitaxy) and conditions of the layer-by-layer growth mode are satisfied. The period of such intensity oscillations corresponds closely with the time needed for the deposition of one monolayer of the material. However, the phenomenon of RHEED oscillation is explained only partially (Ichimiya & Cohen, 2004; Peng *et al.*, 2004). Concerning RHEPD oscillations, so far, they have not been observed in practice, although RHEPD has not yet been used to monitor the growth of thin films. However, in the context of the recent development of RHEPD, undertaking such research work may be expected in the near future. This is why it seems interesting to discuss features of possible RHEPD oscillations.

Our goal is to compare predictions from two theoretical models. Within the first model precise dynamical calculations for the realistic, one-dimensional scattering potential are executed. It is also assumed that the potential of the growing layer is proportional to the potential of the fully completed layer and the coverage of the layer. In the second model, the interference of partial, reflected waves is considered and the final results are obtained with the help of approximate formulas. However, to describe in detail the motivation for carrying out the current work, it seems important to recall some facts. Namely, the first model mentioned was introduced in the literature by Peng & Whelan (1990). These researchers also analysed theoretically the behaviour of the phase of oscillation, showed runs of oscillations for the ideal and non-ideal modes of the layer-by-layer growth, and also discussed theoretical oscillations for heteroepitaxy (Peng & Whelan, 1991*a*,*b*,*c*). Further, this approach was verified successfully in the analysis of the experimental oscillation phase for an off-symmetry azimuth (Mitura *et al.*, 1998). Additionally, Daniluk *et al.* (1996) showed the usefulness of the approach in the interpretation of oscillation runs for heteroepitaxial growth. Actually it seems that in some situations it may be important also to include in considerations scattering by step edges. It can be done by the introduction of an extra imaginary part in the crystal potential as discussed in Dudarev *et al.* (1994) and in Mitura *et al.* (1998). Now, concerning the second model used in the current paper, its basic concepts were introduced by Horio & Ichimiya (1993) to explain qualitatively the effect of double minima in RHEED oscillations. This model is called a simple potential model and it is also based on the assumption that the scattering potential of the growing layer is proportional to its coverage. However, there are two possible versions of this model (Horio & Ichimiya, 1993; Mitura, 2014). In the first version, after the introduction of two values of the potential to describe the scattering properties of the crystal (one value is for the bulk crystal and the other for the growing layer), the Schrödinger equation is precisely solved (Horio & Ichimiya, 1993; Braun *et al.*, 1998). Such a procedure has some virtues – it takes account of amplitudes of scattered waves. However, obtaining simple analytical equations is not possible for such a case. We do not follow this line in the current work. We use the other possible version of the interference model. Namely, by introducing some further simplification, mentioned in Horio & Ichimiya (1993), one can obtain simple approximate formulas. In the book of Ichimiya & Cohen (2004), the values of the phase obtained within such formulas were compared with RHEED experimental results for GaAs of Zhang *et al.* (1987) and of Braun *et al.* (1998), and reasonable agreement was found. However, when researchers at Harvard University used a similar approach in their work for Ge, they found that the agreement which they achieved in the course of interpretation of experimental data was not as good as they had expected (Shin *et al.*, 2007; Shin, 2007). They concluded that the dynamical theory of RHEED, based on the assumption that the scattering potential of the growing layer is proportional to the layer coverage and the potential of the fully completed layer, may fail to explain their results. However, in Mitura (2013), it was discussed that the use of the precise theory employing the realistic, one-dimensional model of the scattering potential allows one to reproduce the experimental results shown in Shin *et al.* (2007). Nevertheless, the general question of how good are the predictions of the simplified interference model remained unclear. This is actually the reason why the current research work was conducted. We decided to compare, from a possibly broad perspective, the results obtained using the simplified model introduced by Horio & Ichimiya (1993) with the results of precise dynamical calculations. Subsequently, the analysis was performed for the growth of two materials and for two types of diffraction.

Our paper is organized as follows. In §2, details of the models and computations are described. Next, in §3, the results obtained for the homoepitaxy of Ge and GaAs are shown. Finally, in §4, our conclusions are presented.

2. Details of calculations

2.1. The case of RHEED

First, we describe how to perform precise dynamical calculations for the one-dimensional model of the potential. Next, we show how to apply simplified formulas to compute the phase of intensity oscillations. For details, see also Mitura (2013).

It is assumed that above the crystal surface the incident and reflected beams can be expressed with the help of the wavevectors $\mathbf{K}_\mathrm{i}$ and $\mathbf{K}_\mathrm{f}$, respectively, and the following relation is satisfied: $|\mathbf{K}_\mathrm{i}|^2 = |\mathbf{K}_\mathrm{f}|^2 = K^2$. Furthermore, the z components of these wavevectors are, respectively, equal to $-k$ and k, where $k = |K| \sin\vartheta$ and ϑ is the glancing angle. The entire crystal is assumed to be contained between z_B and z_T, where $z_T > z_B$, and within the crystal, the following Schrödinger equation is satisfied: $\nabla^2\Psi(\mathbf{r}) - v(z)\Psi(\mathbf{r}) + K^2\Psi(\mathbf{r}) = 0$. The potential $v(z)$ appearing in this equation can be found by summing the contributions from all two-dimensional meshes of atoms.

Now, we need to explain how to find the intensity I of the reflected wave. One of the possibilities is to employ a numerical program used for solving similar problems for the three-dimensional potential (Mitura *et al.*, 1998; Mitura, 2013).

However, in fact the case of the one-dimensional potential can be treated separately and then numerical calculations can be executed in a simpler manner. Herein we describe a new algorithm developed specifically for such a case.

The crystal is divided into W very thin slices having the same thickness h. It is assumed that the boundaries of the w-th slice are determined by z_{w-1} and z_w, where $z_w > z_{w-1}$. Additionally, $z_0 = z_B$ and $z_W = z_T$. We determine a series of coefficients r_w, where w is an integer with values ranging from 0 to W. We begin with $r_0 = 0$. The other coefficients r_w are found computationally using the following formula:

$$r_w = \frac{r_{w-1}[1 + h(ik + \frac{v_w}{2ik}) + \frac{h^2}{2}(-k^2 + v_w)] + h\frac{v_w}{2ik}}{1 - h(ik + \frac{v_w}{2ik}) + \frac{h^2}{2}(-k^2 + v_w) - r_{w-1}h\frac{v_w}{2ik}}, \qquad (1)$$

where v_w is the value of the scattering potential in the middle point of the slice. Finally, the amplitude r of the specularly reflected beam is given by $r = r_W$ and the intensity I of the reflected beam is determined as $I = |r|^2$. Results obtained with the computer code based on this algorithm are practically identical to those obtained using the general code.

An algorithm specific for the one-dimensional potential has already been introduced in Peng & Whelan (1990). Another algorithm of this type was shown in Ichimiya (1991). Actually Ichimiya used a different form of differential equations than the form used by Peng & Whelan. However, in both cases, the crystal was divided into thin slices and exact solutions of respective differential equations within each slice were considered. Next, the transfer matrices for the entire crystal, allowing one to compute easily the amplitude of the reflected wave, were found. Thus, it seems interesting to discuss the advantages and disadvantages of our approach in relation to the approaches mentioned above. In this context, the algorithm defined by equation (1) can be considered to be slightly more convenient for practical use than the previous ones. This is because we do not find the exact solution of differential equations within a slice, but rather employ Heun's method (Chapra & Canale, 2010) to obtain the numerical solution. Because of this, we do not need to use advanced mathematical functions of a complex variable (subsequently, computer codes can be prepared very easily). Moreover, we do not find the transfer matrix for the entire crystal, but rather determine, at the top of each slice, the ratio of amplitudes of backward and forward waves moving in the crystal. This allows one to avoid some trouble with fixing the thickness of the crystal before running computations. In fact, it has already been discussed in the literature that the finding of transfer matrices for the thick layers may cause computational divergences. Therefore, Ichimiya (1983) and Zhao *et al.* (1988) suggested that in practical calculations special matrices, relating some parts of the solutions of differential equations, should be determined at tops of slices. In this context we can say that our approach is similar to the method suggested in Ichimiya (1983). In fact the proposals of Ichimiya (1983) and Zhao *et al.* (1988) are different; however, it was discussed in the literature (Watanabe *et al.*, 1998) that the application of the concepts of Ichimiya (1983) gives nearly the same results as the application of concepts of Light & Walker (1976) which were employed by Zhao *et al.* (1988). Coming back to the algorithm defined by equation (1), it should be said that there are also some disadvantages in its use. Namely, the form of equation (1) is not very informative, *i.e.* one cannot learn from it any basic properties of the solution, while extracting such information is easily possible from the descriptions of the two other algorithms (Peng & Whelan, 1990; Ichimiya, 1991). Also, it seems that, in some situations, slices with very small thicknesses may be required for use in equation (1) to achieve similar numerical accuracy as in the case of the application of exact solutions within individual slices. Thus we can conclude that the method presented here and the methods of Peng & Whelan (1990) and of Ichimiya (1991) should be treated more as complementary methods rather than as alternative ones.

We executed calculations employing the algorithm given by equation (1) for the homoepitaxial growth of Ge(001) and GaAs(001). The perfect layer-by-layer growth was assumed for both materials. Thermal vibrations were not included in the calculations. Also possible reconstructions at the surface were ignored for both materials. The scattering potentials were determined using electron scattering factors for isolated atoms (Peng *et al.*, 2004) assuming respective multipliers of 0.85 and 0.20 for the real and imaginary parts of the potential (Mitura, 2013). The phase of the intensity oscillations was determined for each glancing angle according to the prescription given in Zhang *et al.* (1987). Namely, the phase denoted $t_{3/2}/T$ is defined as the time of the occurrence of the minimum in the second period of oscillations divided by the time period of the oscillations.

Now, we describe how the phase can be found in an approximate way; for more details see Mitura (2013). Namely, it is assumed that the amplitude of the reflected wave can be expressed as the sum of the contributions from two partial waves: the one reflected at the boundary of the vacuum – the growing layer, and the one reflected at the boundary of the growing layer – the bulk crystal. To obtain an estimation of the value of $t_{3/2}/T$, the condition of the occurrence of the destructive interference needs to be analysed in detail. This condition can be expressed as follows: $2\kappa d = (2n + 1)\pi$, where $\kappa = (k^2 - \Theta\widetilde{v}_{\rm Re})^{1/2}$ and d is the thickness of the growing layer (Horio & Ichimiya, 1993). Further, Θ is the coverage of the growing layer and $\widetilde{v}_{\rm Re}$ is the volume-averaged value of the real part of the potential in the bulk crystal (the imaginary part of the potential is ignored in these considerations). On this basis, one can obtain the following condition for $\Theta_{\rm min}$ when the minimum of the intensity is expected (Ichimiya & Cohen, 2004):

$$4(K^2 \sin^2\vartheta - \Theta_{\rm min}\widetilde{v}_{\rm Re})d^2 = (2n + 1)^2\pi^2. \qquad (2)$$

Using the formula (2) and additionally taking into account the relation

$$t_{3/2}/T = \Theta_{\rm min} + 1, \qquad (3)$$

one can find values of $t_{3/2}/T$. It is worth mentioning that the theoretical curve shown in Shin *et al.* (2007) and in Shin (2007) can be reproduced with the help of equations (2)–(3). This is

why we suppose that the authors of the papers cited used the same (or very similar) formulas as displayed by us. Further, it is worth noting that from the use of equations (2)–(3) we should expect to obtain a plot that consists of different branches, where each branch represents the dependence of $t_{3/2}/T$ as a function of the glancing angle for a single value of n. However, we also need to assume that $t_{3/2}/T$ can take values only between 1 and 2. Actually, equations (2)–(3) can be combined and then the following useful formula is obtained (Mitura, 2013): $t_{3/2}/T = [4d^2K^2\sin^2\vartheta - (2n+1)^2\pi^2]/(4d^2\widetilde{\nu}_{\rm Re}) + 1$. Finally, the values of $\widetilde{\nu}_{\rm Re}$ needed to use equations (2)–(3) can be determined on the theoretical basis as discussed in Mitura (2013) and we used such an approach in this paper.

2.2. The case of RHEPD

Some time ago, Ichimiya suggested in a theoretical paper that calculations for RHEPD can be executed using computer codes originally developed for RHEED (Ichimiya, 1992). In this paper, we followed a similar route. Execution of the precise RHEPD calculations required modification of the scattering potential used for RHEED. Namely, its real part was taken with the opposite sign (the imaginary part was left unmodified because this part of the potential does not depend on the particle charge but rather on a detailed mathematical form selected to describe propagating waves).

Simplified calculations of $t_{3/2}/T$ with the application of equations (2)–(3) were also performed for RHEPD. The calculations required only the earlier replacement of $\widetilde{\nu}_{\rm Re}$ by $-\widetilde{\nu}_{\rm Re}$.

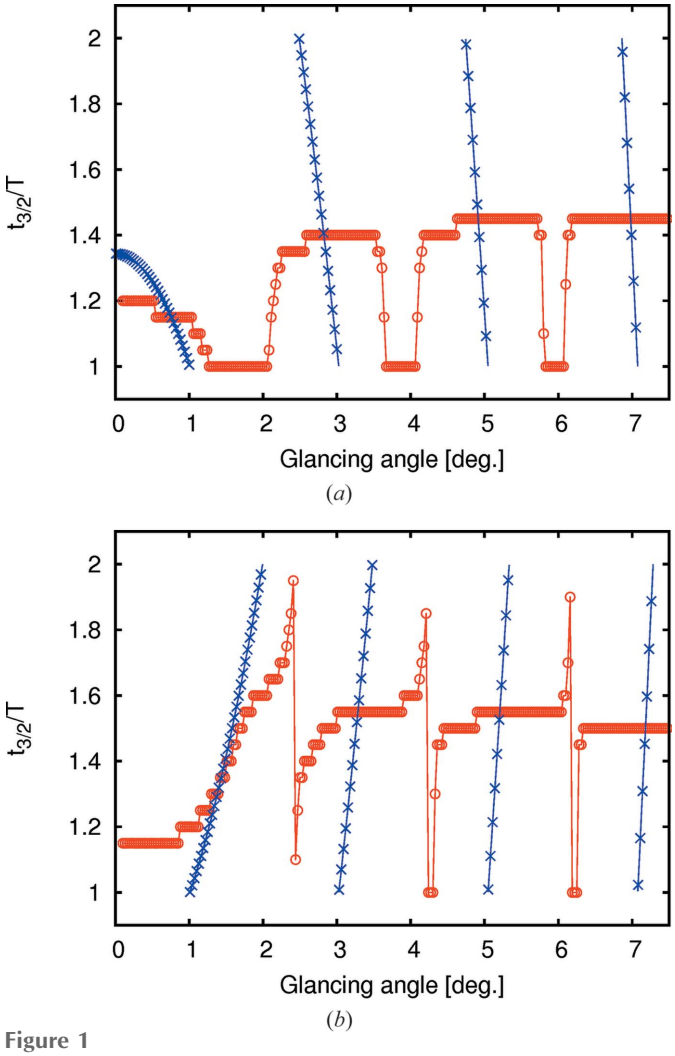


Figure 1
The plots of the phase $t_{3/2}/T$ of the intensity oscillations for the homoepitaxial growth of Ge(001) layers for RHEED (*a*) and RHEPD (*b*). In both parts of the figure, the results of the precise dynamical calculations are shown with circles and the results obtained using the approximate approach are displayed with crosses. The energy for electrons and positrons is taken to be 15 keV.

3. Results

Our main results are shown in Fig. 1 and Fig. 2. The basic difference between calculations for the growth of Ge(001) and the growth of GaAs(001) is that for Ge the growing layer is about two times thinner than that of GaAs. For the first case,

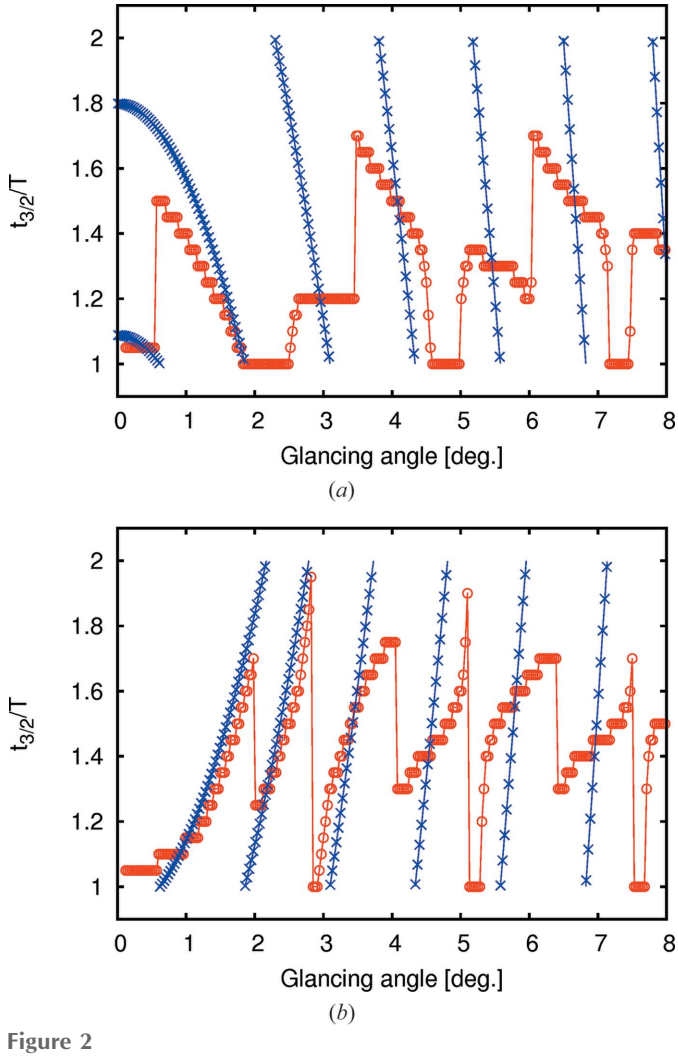


Figure 2
The same as Fig. 1 except the plots are for the homoepitaxial growth of GaAs(001) layers and the energy for electrons and positrons is taken to be 10 keV.

the growing layer contains one mesh of atoms, whereas for the second case, two meshes are involved in the growth.

We computed values of $t_{3/2}/T$ with the angular step of 0.01°. However , to make our figures clearer we displayed plot points with the step of 0.03°.

If we look at the results for Ge (see Fig. 1), we observe that for small glancing angles the predictions of the phase $t_{3/2}/T$ obtained with the help of the simplified approach agree reasonably well with the results of precise calculations. However, for higher glancing angles, only general trends are reproduced with the use of the analytical formulas (2)–(3). However, the agreement between precise and approximate results is much better for the case of GaAs (see Fig. 2). Namely, for some angular ranges, we indeed observe very similar features. Generally, it also seems that the values of $t_{3/2}/T$ obtained applying equations (2)–(3) are more accurate for positrons than for electrons.

Additionally, it is worth briefly discussing the results concerning the possibility of the occurrence of two minima in one period of oscillations. According to theoretical investigations, such an effect may occasionally appear for some glancing angles, but only if the growth mode of the material is relatively close to the perfect layer-by-layer mode. A detailed discussion of different aspects of the appearance of double minima is given in Ichimiya & Cohen (2004). In our case, we can say that, for the assumed growth of Ge, we did not find this effect (for either RHEED or RHEPD). However, for GaAs, the situation is different. If we take a look at the plots obtained with the help of equations (2)–(3) (see Fig. 2) then we can recognize that double minima can be expected in the angular range 0–0.61° for RHEED. For RHEPD, a similar situation occurs for angles of 1.86–2.16°. This is because in the above angular regions for each glancing angle there are two values of $t_{3/2}/T$. However, it is interesting to compare the approximate results with the results obtained using the precise dynamical theory. For calculations for the realistic one-dimensional potential, double minima can be recognized only by directly examining oscillation runs (*i.e.*, the analysis of Fig. 2 is not proper for these calculations because, for this case, during the determination of $t_{3/2}/T$ only the smallest value in a period was identified, as explained in §2). However, we found in the course of the additional examination of individual oscillation runs that double minima (for the perfect layer-by-layer growth) can be expected in the range of 0–0.80° for the case of RHEED and in the range of 1.92–2.10° for RHEPD. Thus, we can say that there exists very good correlation between the results obtained using the simplified approach [equations (2)–(3)] and those obtained employing the dynamical calculations for the one-dimensional scattering potential.

4. Conclusions

We conclude that the concept to explain features of RHEED and RHEPD intensity oscillations expected to be observed for the perfect growth of materials employing two interfering waves and taking into account the refraction effects is very valuable. This concept, introduced in Horio & Ichimiya (1993), helps us to understand the physics ruling the appearance of intensity oscillations. However, using only the analytical approximate formulas (which are available within the simplified approach), we should not expect, in general, to go beyond the level of the qualitative predictions. We admit that in many situations the application of more advanced theoretical models may be required. In particular, for off-symmetry azimuths, conducting calculations employing the realistic one-dimensional potential seems to be more appropriate if precise theoretical predictions of the features of intensity oscillations are needed. However, we should also admit that in some situations, to realize such a goal, the inclusion of the effects of diffuse scattering may prove to be crucial.

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