

Studies on the origin of reflection high energy electron diffraction oscillations observed during molecular beam epitaxy

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Reflection high energy electron oscillations were observed for the first time in the early nineteen eighties. Currently many researchers who deal with the preparation of two-dimensional systems use them to control precisely the thickness of deposited structures. However, one may easily find in the literature essentially different approaches explaining the origin of the intensity oscillations. In fact, it means that up to now the question of why the intensity oscillations appear is still open. In this paper a combined approach is used to explain the effects observed for the case when small atomic islands are formed at the surface during crystal growth. In this approach changes of the refraction conditions at the surface and changes in diffuse scattering are considered simultaneously.

1. Introduction

It was discovered in the early nineteen eighties that epitaxial growth of ultrathin films may be accompanied by regular oscillations of the intensity of scattered electron beams if reflection high energy electron diffraction (RHEED) is employed to monitor structural changes at the surfaces during the deposition of a material [1]–[3]. One period of such oscillations corresponds to the deposition of one atomic layer. Soon the observation of the oscillations became a standard tool applied to control precisely the thickness of deposited films. However, more than twenty years after the described discovery the question of why the phenomenon of the oscillations appears still remains without the widely accepted answer. In the year of 1983 two different approaches explaining this kind of oscillations were proposed. Namely, a step density model [4] (see Fig. 1a) and a terrace model [5] (see Fig. 1b) were introduced. In the first approach it was suggested that oscillations of the beam intensity are a consequence of periodic changes in conditions of random scattering of electrons by atomic island edges. In the second approach it was stated that the oscillations appear because of changes in interference conditions of electron waves reflected from different surface terraces. Thus, the two approaches were supported by solid arguments and, in fact, both of them gave valuable insight into the nature of intensity changes. However, they failed to describe the phase of oscillations [6]. Therefore it is useful to recall some details here.

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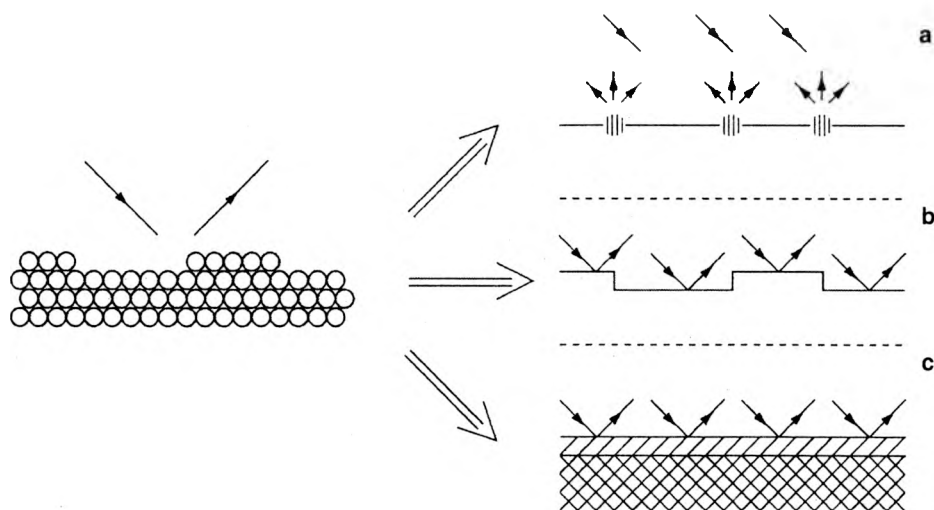


Fig. 1. Schematic illustration of the main features of different approaches suggested to explain the origin of RHEED oscillations: step density model (a), terrace model (b), and proportional model (c).

Different definitions of the phase of the oscillations are possible. Let us consider the following one which is useful for experimental work when only oscillations with simple regular shapes are observed (*i.e.*, resembling cosine-like functions). The phase is defined as follows: $\varphi = 2\pi(t_{s\min}/T - 1.5)$, where $t_{s\min}$ is the time of the occurrence of the minimum in the second oscillation period and T is the steady-state oscillation period. Both aforementioned theoretical models predict a constant value for the oscillation phase; namely, it should always be equal to 0. However, in experiment the oscillation phase strongly depends on the diffraction conditions. This is why in the nineteen nineties a third model explaining the origin of the oscillations was studied in detail in different groups. It was suggested that the intensity oscillations might appear because of periodic changes in refraction conditions at the surface [7] (see Fig. 1c). The model introduced in this approach is called proportional because it is assumed that the scattering potential of a growing layer is proportional to its coverage and to the potential of the fully completed layer. Using this approach it was possible to explain many experimental effects: the complicated shapes of the oscillations for very low glancing angles [8], [9], changes of the intensity during heteroepitaxial growth [10], and recently the oscillation phase [11], [12]. However, one may easily recognize a deficiency of the proportional model: it ignores the topography of the surface or, in other words, random scattering by step edges is totally neglected.

2. Numerical modeling of RHEED oscillations

From the description presented one may conclude that currently there exist grounds to try to formulate an overall description of RHEED oscillations in which arguments of all the three approaches aforementioned would be taken into account. It seems that it

can be achieved if two limiting cases are considered separately. For the first case, when deposited atoms condense at the surface forming large islands, one can use the terrace model [5]. For the second case, when small islands are formed, features of the step density and proportional models should be combined. In this paper we deal only with the second case. Actually, modeling of RHEED oscillations in which changes in refraction conditions and changes in diffuse scattering were considered simultaneously was introduced by DUDAREV *et al.* [13]. They used an adatom/vacancy model to compute the imaginary part of a scattering potential for partially ordered surfaces. However, as a result of the collaboration of the author of this paper with Prof. Whelan's group from the University of Oxford a new model was developed. This new model allows to consider in dynamical calculations larger effects due to diffuse scattering than the model applied in [13]. Advantages of the use of this new combined model rather than of the proportional model were discussed in detail in papers [14], [15], focusing especially on theoretical analysis of the homoepitaxial growth of metals. This is because it seems that fully convincing conclusions on the origin of RHEED oscillations can be achieved only if the simple substrates are used and only if simple growth modes are realized (in other words, if one carries out interpretations of experimental data collected during the growth of thin films on complicated surfaces, then it is impossible to avoid elements of theoretical speculations). Unfortunately, experimental data for metals which are shown in the literature are not proper for the verification of the combined model (the data do not contain all information required). So, to encourage other researchers to undertake measurements of this type, we briefly describe in this paper the combined model and then we demonstrate that for the case of homoepitaxial growth on a complicated surface the combined model can be indeed considered to be more faithful in reproducing actual experimental data than the proportional one.

The description of RHEED oscillations using the proportional and combined models is achieved with the help of the dynamical diffraction theory. Namely, it is assumed that RHEED intensities are calculated using the two-dimensional Bloch wave approach to solve the Schrödinger equation (for review see [16]). The main difficulty in carrying out calculations for growing surfaces is answering the question of how to determine the potential of a partially filled layer. For the combined model we use the following equation

$$V(\Theta, \mathbf{r}) = \Theta V_{\text{Re}}(1, \mathbf{r}) + i\Theta\alpha V_{\text{Re}}(1, \mathbf{r}) + i\Theta(1-\Theta)\beta V_{\text{add}}(\mathbf{r}) \quad (1)$$

where Θ is the growing layer coverage. Let us consider the case where there is only one atom Z per growing layer unit cell. Then to compute $V_{\text{Re}}(1, \mathbf{r})$ Doyle and Turner electron scattering factors [17] are used [16]. We assume that $V_{\text{add}}(\mathbf{r})$ can be computed in the similar manner with the help of the following scattering factor $f_{\text{add}}(s) = f_Z(0)\exp(-100s^2)$, where $s = [|\mathbf{K}|/(2\pi)]\sin v$, and $f_Z(0)$ is the value of the electron scattering factor for atom Z for $s = 0$ ($|\mathbf{K}|$ is the magnitude of the incident beam wave vector and v is the glancing angle). If there are two or more atoms in the unit cell, the total potential is obtained by summing independent contributions of all these

atoms. It should be added that if the term with the multiplier $\Theta(1-\Theta)$ is neglected in Eq. (1) then we get the proportional model. So, the basic difference between the combined model described by Eq. (1) and the proportional model is the presence of the extra imaginary term for the first model. With the help of this term random scattering by step edges is actually included in our considerations. Finally, it should be mentioned that values of α and β should be determined in detail by fitting experimental data. However, from the results of analyses of experimental RHEED rocking curves and azimuthal plots for flat surfaces which are demonstrated in the literature one may expect that values of α are from the range 0.1–0.3. Furthermore, on the basis of experimental studies of HALL *et al.* [18], conducted by transmission electron microscopy one may expect that values of the ratio β/α are from the range 0–5.

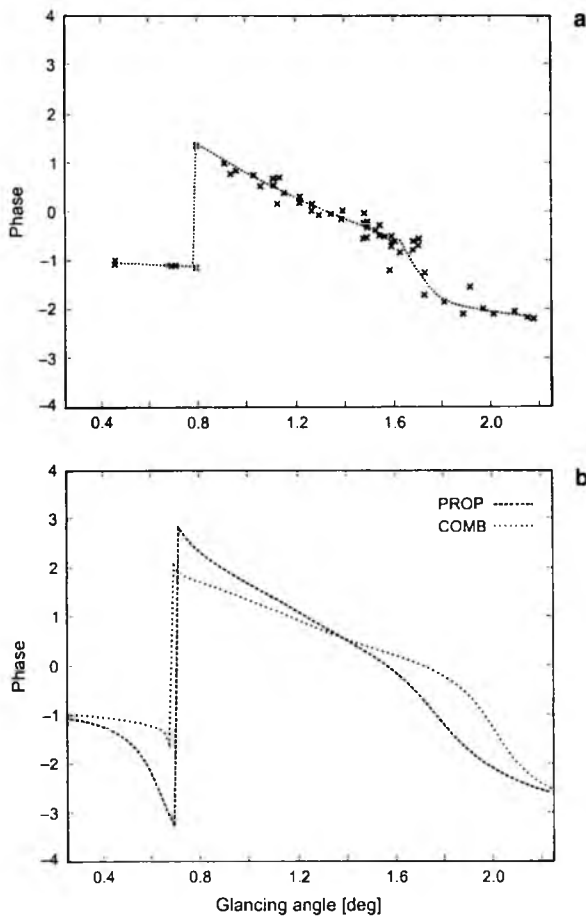


Fig. 2. Experimentally determined phase of RHEED oscillations for the growth of GaAs on a substrate GaAs(001)-2 \times 4 (the data shown are taken from the paper of CROOK *et al.* [19]) (a), and the corresponding theoretical plots of the phase, computed using the proportional model (PROP) and the combined model (COMB) (b).

It seems that the most complete experimental data collected using RHEED systems are shown in the literature for GaAs. This reflects the fact of the great importance of this material for electronic industry. However, to learn basic RHEED features, studying the effects occurring for simple metals would be much more useful. Unfortunately, such data which could be used to verify the combined model do not exist. So, we carry out verification of the combined model for the case of the growth of GaAs on the GaAs(001)-2×4 surface. Due to very complicated nature of GaAs, the theoretical interpretation, which we demonstrate, cannot be considered fully conclusive but it should be rather treated as a provisional test of the new model.

The results of the analysis of the oscillation phase measured during the growth of GaAs(001) on the GaAs(001)-2×4 substrate are presented in Fig. 2. The experimental data are taken from [19]. Measurements were carried out for an off-symmetry azimuth. We concentrate in this paper on the analysis of the phase in the angular range 0.25°–2.25° because in this range experimental error is relatively small. One can see from the Fig. 2 that using the proportional model one may predict the basic trend of the experimental plot. However, it seems that using the above described combined model one may reproduce this trend more precisely. It is worth adding that for the combined model the value of the ratio β/α was determined to be 1.5. Thus the analysis presented confirms the supposition that the inclusion of random diffuse scattering into our considerations indeed leads to a more complete description of RHEED oscillations rather than the use of the proportional model in which only changes in the refraction conditions are taken into account.

3. Conclusions

It seems that the origin of RHEED oscillations can be explained if one considers separately the cases of the formation of large and small atomic islands at the surface during epitaxial growth of the material. For the case of the formation of large islands the terrace theory of diffraction should be applied [5]. If only small islands appear during growth, then one may expect that RHEED oscillations are observed because of periodic changes in refraction conditions and because of periodic changes in random scattering of electrons by step edges. Analysis of the oscillation phase measured during the growth of GaAs on the GaAs(001)-2×4 substrate seems to be the promising confirmation of such a description of RHEED oscillations. However, further experimental data should be collected and then interpreted to obtain the wide acceptance of the aforementioned concepts. The surface of GaAs(001)-2×4 is very complicated (see, for example, [20]) and many details of the analysis may be in fact questioned. However, it is believed that the results demonstrated in this paper will encourage experimentalists (especially those who deal with simple metallic surfaces) to undertake research work to verify findings presented.

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