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Explanation of the initial phase change vs. incident angle of the RHEED intensity oscillation

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Abstract

Reflection high-energy electron diffraction, which is a very widely used monitoring technique of molecular-beam-epitaxial growth processes, has still some unexplained features. An interesting example of these, the so-called $t_{3/2}/T$ phenomenon, is investigated in this work. The first period of the intensity oscillations of reflection high-energy electron diffraction shows a singular behaviour. An interpretation for the initial change of the phase and of the period duration dependence on the incident angle of the electron beam using the notion of surface coherence length is given here. This particular phenomenon is satisfactorily explained in the case of a GaAs (001) surface. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Reflection high-energy electron diffraction; Molecular beam epitaxy; $t_{3/2}/T$ phenomenon; GaAs

1. Introduction

Reflection high-energy electron diffraction (RHEED) is a widely used monitoring technique during molecular-beam-epitaxial (MBE) growth. The reconstruction and roughness of the surface can be observed by the RHEED pattern. The intensity of the RHEED pattern oscillates under appropriate conditions during the growth process. One period of these oscillations corresponds to the growth of one single monolayer (ML) in a layer by layer mode.

RHEED patterns and its oscillations of intensity are very complex phenomena. These effects can be used as a versatile tool for in-situ monitoring of the epitaxial layer growth, in spite of the fact that we do not know many details of its nature. Several mechanisms of the behaviours of RHEED oscillations are not yet understood. For example, some of these problems involve different phases of the specular and non-specular RHEED beams [1], or differences in behaviour of the oscillations in the case of III–V and II–VI materials [2]. The description of these phenomena was attempted by several authors. A thoroughly review of these models is given in Joyce et al. [3]. The anomalies of the initial phase of the RHEED oscillations, the so-called $t_{3/2}/T$ phenomena, are investigated in this work. Making use

of computer simulations we provide an explanation for the initial phase shift.

2. The $t_{3/2}/T$ phenomenon in experiments

The initial period and the amplitude of RHEED oscillations differ from what follows those seen after a prolonged observation. Except for the first period, the measured decay of the oscillations fits well to an exponential function [4]. The incident electron beam impinges on the surface with an angle between 0.7° and 3° . If the incident or the azimuthal angle are changed, then the initial phase of the oscillations also changes. The measured ‘rocking’ data for the specular spot according to Resh et al. [1] and Joyce et al. [3] are shown in Fig. 1. Data points were obtained by measuring the time to the second minimum, so-called $t_{3/2}$, and normalising with respect to the period at a steady state, T . These data are obtained vs. incident angle at two different azimuthal directions for the GaAs (001) surface. The temperature of substrate during growth was 600°C [1] and 580°C [3], respectively. The electron beam energy (12.5 keV) was mentioned only in the second reference [3].

3. Results and discussion

3.1. The notion of the surface coherence length

The simple kinematic theory [1] does not predict the phase shift of the oscillations, which would additionally

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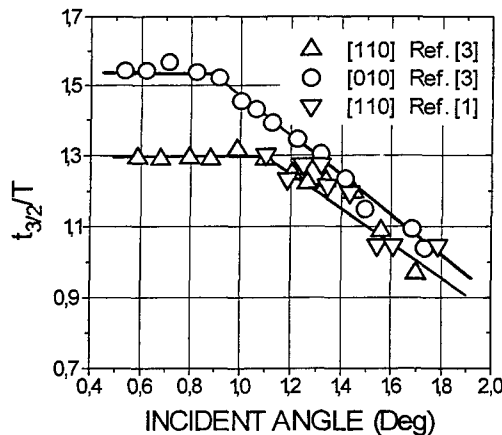


Fig. 1. The experimental 'rocking' data of RHEED oscillation, that is the ratio of $t_{3/2}/T$ as a function of the incident angle of the electron beam in [110] and [010] azimuthal directions of a GaAs (001) surface. The data values originate from Resh et al. [1] and Joyce et al. [3]. The lines serve only as guides to the eye.

depend on the condition of the incidence of the electron beam. The contribution of inelastic processes, such as Kikuchi scattering, to the phase shift phenomenon is not completely taken into account in this approach [1]. The RHEED phenomenon is partly reflection-like and partly diffraction-like. The effect of the phase shift is described by noting the exact positions of the minima of the oscillations. The detailed behaviour of the minima and maxima of the oscillations can be explained also using a geometrical picture, which will be employed also in the present paper. Because the specular spot is not a reflected beam, the interaction of the electron beam and the target surface must be described quantum mechanically. The glancing-incidence angle electron beam interacts with the surface over a relatively large area. However, the reflected-diffracted information obtained does not come from the whole of this area. The interaction between the surface and the electron beam occurs only under special conditions, therefore, we need to consider the notion of surface coherence length (w). Beeby [5] introduced the quantum mechanics definition of this quantity. Here we briefly point the major steps introducing this quantity.

However, now we have supposed that the surface coherence length w is of the same order as the coherence length (Λ) of the beam [6]. In the above-mentioned experiments the energy of the electron beam is in the order of $E = 10$ keV, corresponding to de Broglie wavelength $\lambda = 12.2 \times 10^{-12}$ m. The coherence length of the electron beam Λ can be determined as [7]:

$$\Lambda = \frac{\lambda}{2\beta\sqrt{1 + (\Delta E/E)^2}}$$

where β and ΔE are the angular spread and the thermal width (i.e. the energy spread) of the electron energy of

the incident beam, respectively. The beam divergence ranges usually between 10^{-4} and 10^{-5} rad and the energy spread is approximately 0.5 eV [8]. The spatial width for the wave packet for highly collimated beam can be estimated, to extend by: $\Lambda_0 = 6.1$ nm with these values. Another estimate of the wave packet from interference investigations gives 300λ [9]. In this way we can obtain similar value of $\Lambda_0 = 3.7$ nm for the wave packet.

The spot size of the illuminating electron beam on the surface (L) in the incident direction depends strongly on the incident angle (α). If the cross-section of the illuminating electron beam is b then the touching area dependence on incident angle can be expressed by the following simple trigonometrical function $L = b/\sin\alpha$. This dependence is very strong in the vicinity of an incident angle of 1° . Therefore, we can suppose that the surface coherence length depends on the incident angle and is described by a similar function ($\Lambda = w/\sin\alpha$) [6].

3.2. The size of characteristic growth terraces

The relation between the domain size (s) and the surface coherence length (w) in the case of a polycrystalline surface was investigated in Beeby [5]. This concept can be applied in our case if we consider, instead of domains, identically oriented growth units (or growth terraces). An estimate of the characteristic dimension of a growth terraces can be made from experimental data. The terrace average width (s) and the migration length of Ga (l) depend on the substrate temperature. The RHEED oscillations are present if $l \leq s$ and absent if $l \geq s$. In our case the migration length is 7 nm because the substrate temperature is 580°C [3].

The binding energy on the (001) surface in the directions [110] and $[1\bar{1}0]$ is different, which can be explained with the different dangling bonds in different directions [4]. This anisotropy is manifested in the different growth rates. The growth rate in the [110] direction is larger than that in the perpendicular direction [10]. This anisotropy is apparent not only in the growth of the crystal but also in the etching (i.e. during decomposition of the crystal). This factor can be estimated with the help of etch-pit shapes (see Fig. 4 in Nemcsics et al. [11]), where the ratio of the sides of the rectangle shaped pits is a factor of approximately 2.4. Here is supposed that the growth rates in the [110] and $[1\bar{1}0]$ directions differ by a similar amount (see Fig. 2).

3.3. Initial phase dependence on the incident angle

It is supposed that the surface coherence length and the average terrace width have similar dimensions at glancing-incidence angles ($w \approx s$), because the touching length of the electron beam (assumed to be the same

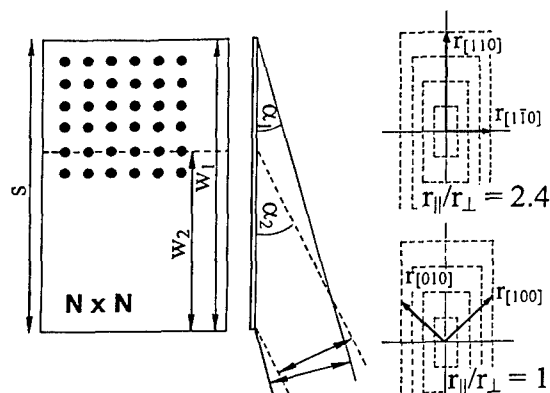


Fig. 2. The left side of the figure shows view of one island with lattice nodes in the growth model consisting of islands of $P \times P$ ($P=4$) terraces of $N \times N$ ($N=36$) lattice sites, where the relation between the terrace dimension and surface coherence length is illustrated. The right side illustrates the ratio of $r_{\parallel}/r_{\perp}$, if the observation direction changes towards $[110]$ and $[010]$.

as the surface coherence length) changes very abruptly at angles less than 1° and in this region the function $t_{3/2}/T$ is constant, since $w > s$ (see Fig. 1). The relation of the surface coherence length and average terrace width is changed with the changes of the incident angle. If the incident angle increases, the surface coherence length becomes smaller than the average terrace width ($w < s$), so and thus the reflected–diffracted information comes from only a part of the average terrace.

The polynuclear growth model in the two-dimensional case was used for our calculations [12]. The simplified picture takes into consideration diffraction contributions only from the topmost ML and the RHEED intensity is taken as proportional to the smooth part of the surface top layer [4]. The computing model assumes $N \times N$ lattice sites in a $P \times P$ growth terrace (Fig. 2) [1,6]. The relation between the terrace size and the area of the surface coherence is shown in left side of Fig. 2. It is clear that the information supplied about the probed surface area decreases with an increasing incident angle of the beam. The different crystallographic directions mean different growth rates. Here the ratio of $r_{[110]}/r_{[1\bar{1}0]}$ are estimated to be 2.4. The oscillations were computed for two different ratios of $r_{\parallel}/r_{\perp}$, where $r_{\parallel}$ and $r_{\perp}$ are the growth rates for the observation direction parallel to the electron beam and perpendicular to it, respectively. Thus, the assumed value of the ratio in the case of the beam along $[110]$ is 2.4. The ratio in the $[010]$ direction taken as 1 (see right side of Fig. 2). The calculated oscillations in the case of both ratios can be seen in Fig. 3. Perfect layer-by-layer growth was assumed in the calculation, so only the actual top monolayer was investigated. The calculated function of $t_{3/2}/T$ vs. the azimuthal angle in the two different directions is plotted in Fig. 4. The growth time for one complete ML in the two different directions is the same

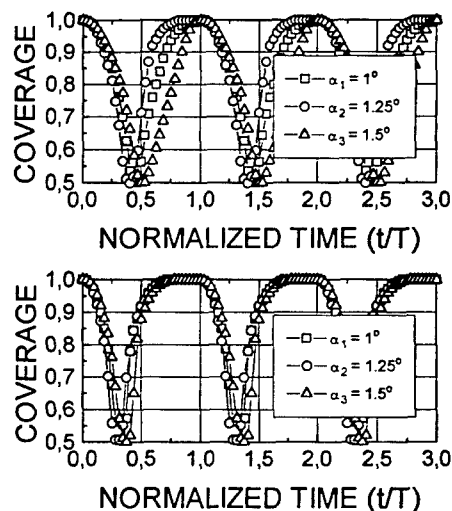


Fig. 3. The computed oscillations at different incident angles. The symbols $\square$, $\circ$ and $\triangle$ mean incident angles of 1° , 1.25° and 1.5° , respectively. The change of incident angle means that the reflected–diffracted information comes from 100%, 80% and 60% of the terrace area, respectively. The upper part of the figure shows the case of $r_{\parallel}/r_{\perp} = 2.4$ and the lower part shows $r_{\parallel}/r_{\perp} = 1$.

(T), but the phase is different ($t_{3/2}$) because of the anisotropic growth rate. These curves correspond very well to the measured ‘rocking’ data in Fig. 1. If the surface coherence length is larger than the average terrace width then the $t_{3/2}/T$ ratio remains constant (with the value is determined by the $r_{\parallel}/r_{\perp}$ ratio). If the surface coherence length is smaller than the average terrace width, then the $t_{3/2}/T$ ratio decreases also.

The behaviour of $t_{3/2}/T$ vs. incident angle was investigated for glancing-incidence angles, under 1.8° . Our model describes the incident angle dependence of $t_{3/2}/T$ in this range only. In real situations, the diffracted–reflected electron beam gets information not only from the topmost ML, but a larger incident angle causes also a larger penetration depth. The description of this phenomenon at larger incident angles probably can be

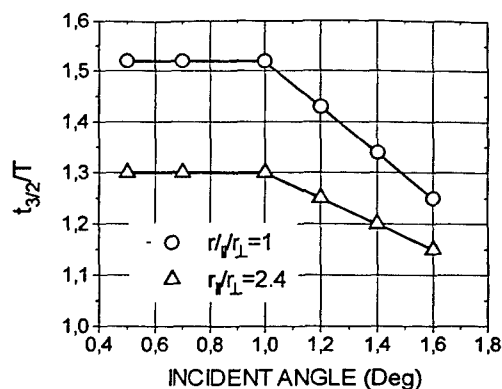


Fig. 4. The computed $t_{3/2}/T$ ratio vs. incident angle in different crystallographic directions (in the case of $r_{\parallel}/r_{\perp} = 2.4$ and $r_{\parallel}/r_{\perp} = 1$).

improved by considering more MLs below the surface during the growth process.

4. Conclusion

The phenomenon of the initial phase variation of the RHEED intensity oscillation is explained here with the help of the concept of the surface coherence length. The surface coherence length corresponds to the surface area probed by a coherent beam, which changes with the incident angle of the beam. A geometrical description was used for the interpretation. Our explanation is demonstrated by a computer simulation. The demonstration was carried out in two different crystallographic directions, where a realistic estimation was made concerning the ratio of the crystal growth rates. The calculations were carried out with a discrete lattice node arrangement. The topmost ML was taken into account. The calculated curves for the decay of the oscillations correspond very well with the measured 'rocking' data in the investigated range.

Acknowledgments

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