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IN SITU X-RAY SCATTERING STUDIES OF THE DEPOSITION OF Ge ATOMS ON Ge (111)

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Résumé - On étudie la nucléation de Ge déposé sur Ge(111) en ultravide par réflectivité de rayons X. La mesure de l'intensité réfléchie du faisceau spéculaire en fonction de l'angle d'incidence donne accès à la distribution de hauteurs de terrasse sur la surface durant la croissance du cristal de Ge(111). Une analyse quantitative utilisant la théorie de diffusion cinématique montre que pour une température de substrat de 200 °C, la croissance se fait essentiellement couche par couche avec toutefois, une certaine rugosité. La rugosité est décrite par un modèle mettant en jeu trois niveaux incomplets de terrasse. Grâce à ce modèle, nous pouvons déterminer l'état d'occupation des trois niveaux supérieurs en fonction de la quantité totale déposée.

Abstract - The nucleation of Ge on Ge(111) has been studied by X-ray reflectivity during deposition in ultrahigh vacuum. Measurements of the specularly reflected intensity as a function of the angle of incidence reveal the distribution of terrace heights on the surface during growth of the Ge(111) crystal. A quantitative analysis using kinematical scattering theory shows that, for a substrate temperature of 200 °C, the growth proceeds predominantly layer by layer but that there is some roughening. The roughening is described by a model which includes three incomplete terrace levels. Using this model we are able to determine the occupation of the three topmost levels as a function of the total deposited amount.

The study of epitaxial growth on an atomic scale is of interest not only from a fundamental point of view. Molecular beam epitaxy (MBE) technology offers the possibility to produce multilayer semiconductor devices. A special class is formed by the so called "monolayer superlattices" [1]. A two-dimensional layer-by-layer growth mode, i.e., a growth where each layer is completely filled before the next one starts on top, allows the fabrication of such devices.

It is well known that, during MBE layer-by-layer growth, pronounced oscillations occur in the specularly reflected intensity of high energy electrons. Details of the growth mode, however, are difficult to reveal since multiple scattering effects play an important role [2] in the reflection of

electrons. Grazing incidence X-ray scattering, on the other hand, is easily described using kinematical theory. Provided the scattering geometry is well chosen, the latter technique is extremely sensitive to the surface morphology on an atomic scale [3]. In this study, the advantages of X-ray scattering are used to observe the Ge growth on a Ge(111) crystal during deposition. For this system a layer-by-layer growth mode has been reported for a substrate temperature of 200 °C [4]. By measuring the reflected intensity during deposition for several values of the perpendicular momentum transfer we are able to describe the growth in terms of occupation numbers of the topmost levels using a model that includes three incomplete terrace levels.

The experimental setup consists of an ultrahigh-vacuum chamber with up to five Knudsen effusion cells, which is coupled to a five-circle diffractometer [5]. The deposition rate of the Ge effusion cell was $(4.4 \pm 0.2) \times 10^{13}$ at./cm².min as calibrated with Rutherford backscattering (RBS). Repeated sputter and anneal cycles as described in [4] were used to clean the Ge(111) sample. After this treatment sharp $c(2 \times 8)$ diffraction patterns were observed with reflection high energy electron diffraction (RHEED) and a reflectivity curve versus perpendicular momentum transfer shows an almost perfectly flat surface. The Ge(111) crystal consists of stacked (111) bilayers (containing each 1.44×10^{15} at/cm²) separated by a distance of 3.27 Å. Maximum sensitivity for the growth of islands of bilayer height is obtained if X-rays scattered from an island and those scattered from the terrace below interfere destructively. For the wavelength of the X-ray's used (1.13 Å) this situation is obtained for an angle of incidence of 5°.

The specular reflected beam can be labeled as a point (hkl) in reciprocal space. Thereto we define a hexagonal unit cell which is related in reciprocal space to the conventional cubic cell by $(100)_{\text{hex}} = \frac{1}{3}(22\bar{4})_{\text{cub}}$, $(010)_{\text{hex}} = \frac{1}{3}(\bar{2}4\bar{2})_{\text{cub}}$ and $(001)_{\text{hex}} = (111)_{\text{cub}}$. Momentum transfer in the perpendicular direction is then represented by the index l and the specular beam at the condition of destructive interference is the $(00\frac{1}{2})$ reflection.

The reflected intensity was recorded as a function of deposition time for several values of the perpendicular momentum transfer l . For each value of the perpendicular momentum transfer a separate time-dependent reflectivity measurement was made during deposition (fig. 1). Prior to each deposition, the surface was recovered by a single sputter/anneal cycle. The recorded intensity was normalized to that of a perfectly flat bulk terminated crystal, including geometrical corrections and background subtraction. The curves in fig. 1. show damped oscillations with a parabolic shape, which are most prominent for $l = \frac{1}{2}$. Note that the period between two successive maxima (minima) is not exactly equal to the time required for the deposition of an amount equal to a bilayer.

In order to interpret the reflectivity measurements quantitatively we consider a surface with a finite number of terrace levels. Using simple diffraction theory, the time-dependent occupancies of the different terrace levels are now derived from the reflectivity curves. For an arbitrary distribution of steps in two dimensions it has been shown [6] that the reflected intensity profile in reciprocal space consists of two parts: a sharp Bragg peak and a step-broadened contribution. In our experiments the acceptance of the detector system was such that only the sharp Bragg component was measured. The total reflected intensity is given by the Fourier transform of the two-point distribution function [6]. The integrated intensity of the normalized Bragg component is given by:

$$I_B(l) = \sum \sum p_n p_m \cos[2\pi l (n-m)] , \quad (1)$$

where p_i is the one-point probability of finding a topmost atom at level i . The summation extends over all possible levels. In the case of a perfect layer-by-layer growth there are only two levels involved. In this context one layer is understood to be a bilayer. For this case one can readily derive the parabolic behaviour of the intensity versus coverage. Assume that a fraction Θ of the surface is covered by islands of bilayer height, then $p_0 = 1 - \Theta$ and $p_1 = \Theta$. For the intensity of the Bragg component we find

$$I_B(l) = \Theta^2 + (1 - \Theta)^2 + 2\Theta(1 - \Theta)\cos(2\pi l) . \quad (2)$$

For $l = \frac{1}{2}$ there is maximal sensitivity for steps: $I_B = (1 - 2\Theta)^2$. No sensitivity is obtained for $l = 0$ and $l = 1$, in agreement with the observations. The damping of the oscillations, however, cannot be explained by this two-level model. In order to explain that, we extend the total number of levels to four, of which the top three have fractional coverages Θ_1 , Θ_2 and Θ_3 . For this model, the one-point probabilities p_i are given by

$$p_0 = 1 - \Theta_1, \quad (3a)$$

$$p_1 = \Theta_1 - \Theta_2, \quad (3b)$$

$$p_2 = \Theta_2 - \Theta_3, \quad (3c)$$

$$p_3 = \Theta_3. \quad (3d)$$

Eq. (1) combined with eq. (3) gives a general expression for $I_B(l)$ which is a function of l and the fractional coverages Θ_1 , Θ_2 and Θ_3 [7]. By evaluating this expression at two different values of l and requiring that the sum $\sum \Theta_i$ equals the total deposited amount Θ , we can find solutions for Θ_1 , Θ_2 and Θ_3 . The solutions for Θ_i have a simple appearance if one evaluates the intensities at $l = \frac{1}{2}$ and $l = \frac{1}{4}$:

$$\Theta_1 = \frac{1}{4}[(1 + \Theta + \beta) \pm \sqrt{2I_B(l = \frac{1}{4}, \Theta) - (\Theta - \beta - 1)^2}] \quad (4a)$$

fig. 1. Normalized reflectivity curves during deposition, for several values of the perpendicular momentum transfer. The solid curves are calculated using a four level model as described in the text.

$$\Theta_2 = \frac{1}{2}(\Theta - \beta) \quad (4b)$$

$$\Theta_3 = \frac{1}{2}(\Theta + \beta - 2\Theta_1), \quad (4c)$$

with

$$\beta \equiv \frac{1}{2}[1 \pm \sqrt{I_B(l=\frac{1}{2}, \Theta)}]. \quad (4d)$$

From the four solutions, the physical one is selected using two boundary conditions. Firstly, there should be no overhangs, i.e. the fractional coverage of a bilayer should always be lower or equal to the coverage of the bilayer below. Secondly, the fractional coverages Θ_i should be within the range $0 \leq \Theta_i < 1$.

From the measured intensity vs. coverage curves for $l=\frac{1}{2}$ and $l=\frac{1}{4}$ we calculated the fractional coverages Θ_i as a function of the total coverage using equations (4). The results are given in fig.2.

fig. 2. The fractional coverage Θ_1, Θ_2 and Θ_3 as a function of the total deposited amount. These were derived from the experimental results as described in the text. Note that once the lowest layer is filled the second layer becomes the lowest layer.

We note that the damping of the oscillations is caused by a very small amount (5%) of atoms nucleating on top of the almost filled first layer, showing how sensitive X-ray scattering is to surface morphology on an atomic scale. Using eqs. (3) and (1) and the values of the fractional coverages as given in fig. 2. we can calculate the behaviour of the integrated Bragg intensity for values of the perpendicular momentum transfer l other than $l=\frac{1}{2}$ and $l=\frac{1}{4}$. The results are given as solid curves in fig. 1.

As long as the system has not reached a "steady state" growth mode - one in which the distribution of atoms over the levels as a function of deposition stays constant for successive deposited bilayers - the period of the oscillation is not equal to the time required for the deposition of a bilayer and the oscillation is damped. Calibration of the deposited amount using the period of the intensity oscillation, as is routinely done using specularly reflected high energy electrons [8], would in our case yield an error of 10%.

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