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Article in *Journal of Applied Crystallography* · June 2016

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J. Appl. Cryst. (2016). **49**, 961–967

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Asymmetric skew X-ray diffraction at fixed incidence angle: application to semiconductor nano-objects

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Received 13 August 2015

Accepted 15 April 2016

Edited by G. Renaud, CEA-Grenoble DSM/INAC/SP2M/NRS, Grenoble, France

Keywords: X-ray diffraction; asymmetric skew geometry; grazing incidence; quantum dots.

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A procedure for obtaining three-dimensionally resolved reciprocal-space maps in a skew X-ray diffraction geometry is described. The geometry allows tuning of the information depth in the range from tens of micrometres for symmetric skew diffraction down to tens of nanometres for strongly asymmetric skew geometries, where the angle of incidence is below the critical angle of total external reflection. The diffraction data are processed using a rotation matrix formalism. The whole three-dimensional reciprocal-space map can be measured by performing a single azimuthal rotation of the sample and using a two-dimensional detector, while keeping the angle of incidence and the X-ray information depth fixed (FIXD method). Having a high surface sensitivity under grazing-incidence conditions, the FIXD method can be applied to a large variety of Bragg reflections, particularly polar ones, which provide information on strain and chemical composition separately. In contrast with conventional grazing-incidence diffraction, the FIXD approach reveals, in addition to the lateral (in-plane) components, the vertical (out-of-plane) component of the strain field, and therefore allows the separation of the scattering contributions of strained epitaxial nanostructures by their vertical misfit. The potential of FIXD is demonstrated by resolving the diffraction signal from a single layer of InGaN quantum dots grown on a GaN buffer layer. The FIXD approach is suited to the study of free-standing and covered near-surface nano-objects, as well as vertically extended multilayer structures.

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1. Introduction

X-ray structure analysis (Pietsch *et al.*, 2004; Cowley, 1995) reveals the atomic structure of crystalline materials and nanostructures by evaluating measured diffraction intensity patterns. The patterns contain encrypted information about the crystal symmetry and the mean setup of the unit cell, as well as the size, faceting, chemical composition and strain distribution of the nano-objects, and the statistical structure correlation properties, including the surface and interface roughness. Consequently, the properties of the three-dimensional nanostructure can be estimated, reconstructed or at least experimentally confirmed from the evaluation of the

measured three-dimensional distribution of the scattered intensity in the vicinity of suitable Bragg reflections.

Modern epitaxial semiconductor nanostructures, being nearly zero dimensional in terms of the confinement of the electron wavefunctions, are in fact complex three-dimensional nano-objects, which, according to their morphology, produce composite reciprocal-space intensity patterns, and their extremely small size (and coherent scattering volume) creates a number of challenges for X-ray studies.

With no loss of generality, we focus in the following on the example of epitaxial III–V zincblende and wurzite semiconductor systems with [001] orientation of the substrate. X-ray diffraction (XRD) is sensitive to the lattice displacement field *via* the scalar product of the displacement and diffraction vectors and is thus insensitive to the components of the displacement vector perpendicular to the diffraction vector. Therefore, a set of non-collinear reflections is required to reconstruct the strain components. In standard coplanar Bragg diffraction geometry (the surface normal is in the plane of scattering), the variety of accessible reflections is limited. For [001] zincblende semiconductor systems, for instance, the most common coplanar reflections belong to the span of symmetric 004 and asymmetric 224 reflections (Holý *et al.*, 1999, 2008; Baumbach *et al.*, 1999; Schmidbauer *et al.*, 2006, 2008). While (within the range of commonly used X-ray energies) polar Bragg reflections of the {111} series are out of reach in coplanar geometry, these reflections – if accessible – would allow the technique to be material sensitive in the case of ternary systems such as InAs/GaAs compounds. In general, the available reflections measured in non-grazing geometries are characterized by a high information depth Z_{inf} , which, according to the expression

$$\frac{1}{Z_{\text{inf}}} = \frac{1}{Z_{\text{inc}}} + \frac{1}{Z_{\text{scatt}}}, \quad (1)$$

depends on the values of the absorption depths of the incident (Z_{inc}) and scattered (Z_{scatt}) beams. Z_{inf} reaches the value of tens of micrometres in the case of coplanar reflections, and significantly exceeds the thickness of the nanostructures (usually up to hundreds of nanometres). For conventional non-grazing scattering geometries, the ratio between the intensities originating from the surface nanostructures on one hand and the substrate or buffer layers on the other, being proportional to the respective scattering volumes, could easily be as low as 10^{-4} or even less. Therefore, the weak scattering signal from the nano-objects often becomes indistinguishable, especially in the close vicinity of the Bragg peaks from the substrate or the buffer layer (Krause *et al.*, 2014). Certainly, for selected reflections and X-ray energies, coplanar diffraction could also be realized under grazing-incidence conditions (Höche *et al.*, 1988; Rhan *et al.*, 1993; Kimura *et al.*, 1994; Holý *et al.*, 1995), resulting in enhanced diffraction signals from the surface structures. However, the coplanar scattering geometry has the intrinsic drawback that the angle of incidence changes during collection of the reciprocal-space map.

Grazing-incidence diffraction (GID) (Afanas'ev & Melkonyan, 1983; Aleksandrov *et al.*, 1985) is realized in a strongly

noncoplanar diffraction geometry with the diffraction vector parallel to the surface, and allows us to probe the crystalline properties of surfaces (Dosch *et al.*, 1986), thin films, multilayers (Baumbach *et al.*, 1995; Stepanov *et al.*, 1995) and surface nanostructures (Pietsch *et al.*, 2004). GID is particularly sensitive to free-standing surface structures and close-to-surface nano-objects, since it allows tuning of the information depth down to extremely small depths under the condition of total external reflection (TER):

$$l_0 = \frac{1}{2k \operatorname{Im}(\alpha_m)}, \quad (2)$$

where k is the wavevector length and α_m is the complex angle between the surface and the $\mathbf{k}$ vector in a crystal. Thus, the GID geometry allows the separation of the scattering contribution of the near-surface structures from that of objects located deeper than l_0 . If the incident angle exceeds the critical TER value α_c , the GID regime converts to the extremely asymmetric Laue case of diffraction. The output of diffracted intensity above the sample horizon drops drastically, while the information depth grows rapidly. Having the main components of the scattering vector parallel to the crystal surface, GID gains high sensitivity to the in-plane strain components (Lübbert *et al.*, 1999; Schmidbauer *et al.*, 1999; Baumbach *et al.*, 2000; Kegel *et al.*, 2000; Schülly *et al.*, 2003; Grigoriev *et al.*, 2003) but lacks any sensitivity to the normal (z) component of the displacement field (Hanke *et al.*, 2004).

In order to overcome the limitations of standard coplanar Bragg diffraction and GID, more general noncoplanar scattering geometries could be used (Kriegner *et al.*, 2013). As an example, Bragg diffraction in the skew geometry is widely used in its noncoplanar symmetric form for investigations of GaN films (Heinke *et al.*, 2000; Moram *et al.*, 2009; Kaganer *et al.*, 2005). It also provides access to reflections of the {111} series for [001]-grown III–V semiconductor systems, which are not accessible within the methodology of the coplanar geometry. But usually, the information depth for the symmetric skew geometry remains high. In order to remedy the shortcoming of high penetration depth, an extremely asymmetric skew geometry in grazing-incidence conditions (see Fig. 1) has been developed (Haertwig, 1977; Hoche *et al.*, 1986; Stepanov & Kohler, 1994; Bushuev & Oreshko, 2003). Here, a sample rotation around the diffraction vector $\mathbf{Q}$ [$|\mathbf{Q}| =$

Figure 1
Overview of asymmetric skew geometry with grazing incidence and steep exit. $\mathbf{k}^i$ and $\mathbf{k}^f$ indicate the incident and diffracted beams, respectively, and $\mathbf{Q}$ denotes the diffraction vector. The plane of scattering is marked in green. The yellow cone corresponds to all possible combinations of the incident and diffracted beams.

$Q = (4\pi/\lambda)\sin(\theta_B)$, where θ_B is the Bragg angle and λ is the wavelength of the incident radiation] is performed in order to create the extremely asymmetric skew geometry, where the angle of incidence is small (grazing incidence) and the exit angle is steep. The usual θ or $\theta-2\theta$ scans in this configuration change the angle of incidence during the scan with θ and consequently change the information depth too (Bruel *et al.*, 1990; Ress *et al.*, 1998). Recently, this approach was applied to an investigation of crystal truncation rod intensity distribution with a point detector (Fister *et al.*, 2013) and to a strain and composition study of SiGe islands (Stangl *et al.*, 2001).

In the present paper, we describe an approach which combines the surface sensitivity of GID with the advantages of the conventional coplanar and skew geometries. In particular, it facilitates access to polar reflections and vertical strain components. By this approach it is possible to measure the three-dimensional reciprocal-space intensity distribution in an extremely asymmetric skew geometry, realized by a single azimuthal rotation of the sample at a fixed incident angle. In this regime of diffraction, the boundary conditions of wave propagation for the incident wave, and hence the information depth, can be changed smoothly by choosing the angle of incidence. In the following sections we first describe this technique, which we will call FIXD, and its technical realization, performing scans and transforming the measured data into reciprocal space using a rotation matrix formalism. Finally, we apply the FIXD approach to strain and composition detection of small (InGa)N quantum dots (QDs) with low In content.

2. The FIXD approach

2.1. Accessing the Bragg reflection in the basic FIXD setup

The following considerations are based on a six-circle diffractometer layout, presented in Fig. 2 and described by You (1999). In our case, a sample scan can be organized in the most effective way if two circles are used for detector positioning and three for sample orientation, with the last (free) motor dedicated to the scan itself. Let us define the laboratory coordinate system (x, y, z) and the diffractometer rotation axes $(\varphi, \chi, \theta, \nu, \delta, \mu)$. The axes of the laboratory coordinate system are oriented as follows: the x axis is parallel to the incident beam, the z axis points in the vertical direction and the y axis is defined as the vector product $\mathbf{z} \times \mathbf{x}$. In the initial conditions (all angular values of the diffractometer are set to 0), we assume that all the rotation axes of the diffractometer except the χ axis are perpendicular to the incident beam and the incident beam hits the centre of the detector. The χ axis is parallel to the incident beam and the x axis.

The sample coordinate system is defined as follows. The lateral component of the reciprocal node under investigation Q_{rad} defines the axis x_s . At this point, the axes x_s, y_s and z_s coincide with the corresponding axes of the laboratory system (Fig. 3a). This arrangement corresponds to the state of a well adjusted sample at the beginning of an experiment. The Bragg

Figure 2
The six-circle diffractometer, showing its rotational axes and the laboratory coordinate system.

Figure 3
The basic geometry of FIXD. x, y and z correspond to the laboratory coordinate system, while x_s, y_s and z_s belong to the sample. α_i is the angle of incidence. The Bragg condition is satisfied when the sample is rotated by $180 - \Phi$ degrees about the z_s axis (φ motor). The centre of the Ewald sphere is marked as E . $\mathbf{k}^i$ and $\mathbf{k}^f$ are the wavevectors of the incident and scattered beams, respectively. Q^B is the reciprocal vector position after transformation (4).

condition for a given reciprocal vector $\mathbf{Q}$ is achieved in two simple but important steps.

First, we set the incident angle α_i to the chosen value by rotating the sample around the y axis in the negative direction.

Second, to satisfy the Bragg condition in the Laue formulation, the equation $|EQ| = |EO|$ needs to be valid. This happens for two azimuthal (rotation by the motor φ about the z_s sample axis) positions $\varphi^* = \pm(180 - \Phi)$ of the sample, where Φ is the angle between the x_s axis and the projection of the incident beam onto the sample surface (see Fig. 3c). Here and in the following the asterisk sign is used to denote the actual angular position of the given goniometer motor. With the wavevector length $k = 2\pi/\lambda$, we can express Φ as

$$\Phi = \arccos \left[\frac{Q_{\text{rad}}^2 + Q_z^2 - 2k(\sin \alpha_i) Q_z}{2k(\cos \alpha_i) Q_{\text{rad}}} \right]. \quad (3)$$

The angle Φ is a complex number if the desired reflection can be achieved in a coplanar geometry and the chosen angle α_i is smaller than the incident angle for coplanar diffraction (see Fig. 1).

In this subsection we consider all rotations being performed in the laboratory coordinate system (see Fig. 2). To simplify the arithmetic, we first make a rotation about the azimuthal motor axis (φ). In this case, the axis z_s still coincides with z and the whole sequence of rotations will be reflected in the transformation matrix (Vlieg, 1997, 1998; Schlepütz *et al.*, 2010)

$$R^{\text{Bragg}} = R_Y(-\alpha_i) R_Z(180 - \Phi). \quad (4)$$

In the following, R_X , R_Y and R_Z denote the rotation matrices about the corresponding laboratory coordinate axes and ω is the rotation angle.

$$\begin{aligned} R_X(\omega) &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos \omega & -\sin \omega \\ 0 & \sin \omega & \cos \omega \end{pmatrix}, \\ R_Y(\omega) &= \begin{pmatrix} \cos \omega & 0 & \sin \omega \\ 0 & 1 & 0 \\ -\sin \omega & 0 & \cos \omega \end{pmatrix}, \\ R_Z(\omega) &= \begin{pmatrix} \cos \omega & -\sin \omega & 0 \\ \sin \omega & \cos \omega & 0 \\ 0 & 0 & 1 \end{pmatrix}. \end{aligned} \quad (5)$$

To denote the real diffractometer (motor) rotations, Greek letters will be used: R_φ , R_θ and R_δ , according to Fig. 2. In contrast with the laboratory axes, some of the diffractometer axes may change their orientation. Expression (4) given in motor axes is

$$R^{\text{Bragg}} = R_\theta(-\alpha_i) R_\varphi(180 - \Phi). \quad (6)$$

In the sample Bragg position, the reciprocal vector $\mathbf{Q}$ according to transform (4) is described by its components in the following way (laboratory system):

$$\mathbf{Q} = \begin{pmatrix} Q_x^{\text{B}} \\ Q_y^{\text{B}} \\ Q_z^{\text{B}} \end{pmatrix} = R^{\text{Bragg}} \begin{pmatrix} Q_{\text{rad}} \\ 0 \\ Q_z \end{pmatrix}. \quad (7)$$

The wavevector of the diffracted wave obtains the form

$$\mathbf{k}^{\text{f}} = \begin{pmatrix} k_x^{\text{f}} \\ k_y^{\text{f}} \\ k_z^{\text{f}} \end{pmatrix} = \begin{pmatrix} Q_x^{\text{B}} + k \\ Q_y^{\text{B}} \\ Q_z^{\text{B}} \end{pmatrix}. \quad (8)$$

The components of the vector $\mathbf{k}^{\text{f}}$ determine the angles μ^* and δ^* required for positioning the detector:

$$\begin{aligned} \delta^* &= \arcsin(k_z^{\text{f}}/k), \\ \mu^* &= \arctan(k_y^{\text{f}}/k_x^{\text{f}}). \end{aligned} \quad (9)$$

Equations (3), (4), (7), (8) and (9) illustrate the sequence of operations in the rotation matrix formalism needed to find the extremely asymmetric noncoplanar reflection:

- (i) Set the desired angle of incidence α_i using motor θ .
- (ii) Find the corresponding Φ value and use motor φ for rotation $R_\varphi(180 - \Phi)$ to set the sample in the Bragg position.
- (iii) Calculate the angles μ^* and δ^* of the diffracted beam and move the detector accordingly.

Fig. 3(c) suggests that the optimal way of scanning reciprocal space, while keeping the incident angle constant, is a one-motor scan about the z_s axis, namely a φ scan. In the next subsection we will analyse the φ scan in detail.

2.2. Scans in reciprocal space and probed reciprocal volume

Fig. 3 represents the horizontal sample mounting. In fact, the type of sample mounting (horizontal or vertical) plays no role for laboratory sources. At the same time, synchrotron beams demonstrate much better quality in terms of size and divergence in the vertical direction compared with the horizontal one. Additionally, synchrotron radiation is essentially polarized in the horizontal plane. This means that the sample, by rotation around the x axis (χ rotation), has to be set in the position which brings the scattering plane to the vertical position. In fact, the diffraction process remains invariant concerning the rotation of the crystal about the x axis. Hence, by a combination of the proper rotation matrices it is possible to create any desired configuration of the scattering, either with a vertical (horizontal) orientation of the sample, or with a vertical (horizontal) plane of the scattering. In the latter case, a simple four-circle diffractometer (3 + 1) is sufficient to perform FIXD experiments. Thus, FIXD can also be realized utilizing a suitable laboratory X-ray source. For some special applications, however, for example *in situ* growth experiments using an appropriate molecular beam epitaxy instrument (Slobodskyy *et al.*, 2012), the vertical sample position is the only option.

Let us illustrate the idea of rotation around the x axis using vertical sample placement as an example. For this case, we do not need to develop the geometry of the experiment from the very beginning. Instead, we need to modify equation (4) by introducing a rotation about the x axis (circle χ of the diffractometer in Fig. 2). As a result, we obtain a new

experimental setup (the sample is in the Bragg position if $\varphi^* = 180 - \Phi$ in the laboratory coordinate system):

$$R^{\text{Exp}}(\alpha_i, \varphi^*) = R_Z(-\alpha_i) R_Y(-\varphi^*) R_X(90). \quad (10)$$

The same operation can be written using the names of the diffractometer circles:

$$R^{\text{Exp}}(\alpha_i, \varphi^*) = R_\eta(-\alpha_i) R_\varphi(\varphi^*) R_\chi(90). \quad (11)$$

The operations (10) and (11) create the diffraction geometry shown in Fig. 4. This scheme is very similar to Fig. 3(c), but now the angle of incidence is defined by rotation about the η axis. At the upper right, D and the green surface denote the segment of the Ewald sphere probed by the two-dimensional detector. The easiest way to evaluate the detector position in k coordinates $[Q_{x(i,j)}^D, Q_{y(i,j)}^D, Q_{z(i,j)}^D]$ for each pixel (i, j) of the detector (the visible part of the Ewald sphere) consists of two steps.

In the first step the detector pixels have to be calibrated utilizing the primary beam around the initial position ($\mu = 0, \delta = 0$) and keeping in mind the coordinates of the point E ($-k, 0, 0$). In the second step we have to apply the rotation matrix formalism and evaluate the detector pixel positions ($Q_{x(i,j)}^D, Q_{y(i,j)}^D, Q_{z(i,j)}^D$) in reciprocal space (laboratory system) under the conditions of the experiment:

$$\begin{bmatrix} Q_{x(i,j)}^D(\mu^*, \delta^*) \\ Q_{y(i,j)}^D(\mu^*, \delta^*) \\ Q_{z(i,j)}^D(\mu^*, \delta^*) \end{bmatrix} = R_Z(\mu^*) R_Y(-\delta^*) \begin{bmatrix} Q_{x(i,j)}^D(0, 0) \\ Q_{y(i,j)}^D(0, 0) \\ Q_{z(i,j)}^D(0, 0) \end{bmatrix}. \quad (12)$$

We implement a single rotation scan to probe the volume around the reciprocal node $\mathbf{Q}$. As we see from Fig. 4, during the scan by a sample rotation round the z_s axis (φ scan), the reciprocal node (shown as the rose sphere) moves along the arc from the beginning point (labelled b) to the end point (labelled e). It crosses the Ewald sphere (marked in green) at

Figure 4
A FIXED scan with a vertical sample position. A scheme of the scan in the laboratory coordinate system is shown. The green surface marked with D corresponds to the segment of the Ewald sphere covered by the detector window. The scan is executed by a rotation about the z_s axis ($\Delta\varphi$). The reciprocal node (rose sphere) passes along the trace between the points b (beginning of the scan) and e (end of the scan). All other symbols correspond to Fig. 3(c). The inset demonstrates the reciprocal volume in the vicinity of the main reflection (red sphere) covered by the detector window during the scan from b to e in the sample reciprocal coordinates.

the exact Bragg position. It is very important to mention that a φ scan does not alter the angle of incidence α_i . At each step of the scan the absolute position of the reciprocal node in the laboratory coordinate system can be evaluated by substituting the actual value of φ^* into $R_Y(R_\varphi)$ and equations (10) and (11), and using equation (7):

$$\begin{pmatrix} Q_x^{\text{Exp}} \\ Q_y^{\text{Exp}} \\ Q_z^{\text{Exp}} \end{pmatrix} = R^{\text{Exp}}(\alpha_i, \varphi^*) \begin{pmatrix} Q_{\text{rad}} \\ 0 \\ Q_z \end{pmatrix}. \quad (13)$$

The matrix $R^{\text{smp}} = R^{\text{Exp}}(\alpha_i, \varphi^*)^{-1}$ projects the node coordinates in the laboratory system back to the initial position (Fig. 3a). This matrix, being applied to the coordinates of the detector in the laboratory system $[Q_{x(i,j)}^D(\mu, \delta), Q_{y(i,j)}^D(\mu, \delta), Q_{z(i,j)}^D(\mu, \delta)]$, transforms them to the sample coordinate system, keeping their relative positions with respect to the reciprocal node constant.

The sequence of ‘back-transformed’ detector frames, taken at each step of the scan, fills the three-dimensional volume around the reciprocal node (blue box in the inset of Fig. 4).

The approach described above, based on rotation matrices, facilitates the non-trivial geometric calculations for three-dimensional reciprocal-space mapping in a skew geometry. However, it is not restricted to this single problem and can be universally applied for any other geometry (coplanar, non-coplanar, GID) and any trajectory of the reciprocal node and detector. The only requirement is access to the motor positions, which are usually available through the log files of the scan or (preferably) recorded in the file header of each frame. The practical advantages of the rotation matrix algorithm have been demonstrated and analysed comprehensively by Vlieg (1997, 1998) and Schlepütz *et al.* (2010), for instance.

2.3. Difficulties and limitations of the FIXED method

It is also necessary to discuss some difficulties and limitations of this method. In the present paper we have assumed the kinematic approximation of X-ray diffraction, which allows us to use a relatively simple approach to describe reciprocal space and calculate the diffraction vector coordinates. The presence of any dynamic effects may lead to additional features in reciprocal space and could make the interpretation of the diffracted intensity more complicated. Moreover, FIXED as a noncoplanar geometry has a complicated resolution function in three-dimensional reciprocal space. Owing to the convolution of a diffraction pattern with the resolution element, the resulting diffraction might have a non-trivial shape and contain mixed information about the sample and the X-ray source. In some cases, this could be a limiting factor for laboratory sources and an additional reason for using third-generation synchrotrons.

3. Experimental application of the FIXED approach: InGa_N quantum dots with low In content

Here, we demonstrate the method’s sensitivity to the chemical composition and strain of surface nano-objects. We exploit the

versatility of FIXD by measuring a Bragg reflection with the essential Q_z component of the scattering vector at grazing-incidence conditions.

The InGaN quantum dot structures were grown by metal-organic vapour-phase epitaxy. At 1323 K, a 2 μm thick GaN buffer layer was deposited on a 330 μm thick sapphire (0001) substrate. It should be mentioned that GaN buffer layers typically have $5 \times 10^8 \text{ cm}^{-2}$ screw dislocations and $5 \times 10^9 \text{ cm}^{-2}$ edge dislocations (Krause *et al.*, 2014). Because of this very high defect density, the InGaN quantum dots can be considered to be grown on separate GaN blocks, which are tilted and twisted with respect to each other.

The InGaN QDs were grown at 873–1523 K according to the spinoidal decomposition mechanism (Tessarek *et al.*, 2011), resulting in the formation of a so-called meander structure which consists of a system of small nearly connected nano-islands (quantum dots) with a low indium content. Additionally, relatively large (InGa)N islands with a high indium content (above 90%) and of truncated pyramidal shape with a lateral size of 40–50 nm appear, due to excess In accumulated during the spinoidal decomposition.

According to Krause *et al.* (2014), the large pyramidal islands could be successfully resolved in an asymmetric geometry of X-ray diffraction, for instance, using the $10\bar{1}5$ reflection in non-grazing conditions. However, dislocations in the GaN buffer layer cause a broad area of intense diffuse scattering in reciprocal space (Moram & Vickers, 2009). This strong scattering from the GaN layer completely conceals the weak signal from the low-In (InGa)N QDs. Applying FIXD, it is possible to measure the $10\bar{1}3$ reflection in grazing-incidence conditions and thus resolve the signal of the low indium concentration quantum dots.

The following experiments were performed on the ID03-beamline at the ESRF (Grenoble, France) at an energy of 13 keV using a six-circle diffractometer and a two-dimensional Maxipix detector (Ponchut *et al.*, 2011).

Figure 5
 $Q_{\text{rad}}-Q_z$ intensity maps for the $10\bar{1}3$ reflection with incident angles of (a) 10° and (b) 0.3° . The positions of GaN and low-In QDs in reciprocal space are marked by circles and pentagons, respectively. Arrows point to the InN reciprocal node (relaxation direction), which deviates slightly from the direction to the coordinate origin.

The $Q_{\text{rad}}-Q_z$ intensity distributions of the $10\bar{1}3$ reflection obtained by FIXD are shown in Fig. 5 for incidence angles of 10° (Fig. 5a) and 0.3° (Fig. 5b). This figure represents only the vicinity of the main GaN $10\bar{1}3$ reciprocal node. The signal of the In-rich InGaN QDs is not within the scanned region. However, this signal was well investigated by Krause *et al.* (2014) (see Fig. 5d in that work). Fig. 5(a) shows no indication of the presence of QDs, since the weak signal of the low-In QDs vanishes beneath the strong and broad signal originating from the GaN buffer layer. In Fig. 5(b) a well pronounced cloud of intensity shifted in the direction of the InN $10\bar{1}3$ reciprocal node (relaxation direction) can be seen, in addition to the signal from the GaN buffer layer (compare Fig. 5a). We attribute this additional feature to the intensity scattered by the low-In (InGa)N QDs in the meander structure. The position of the QD signal in the $Q_{\text{rad}}-Q_z$ plane implies almost relaxed material. Hence, Vegard's law can be applied, which yields a mean In content of $\sim 3.5\%$ in the small low-indium-concentration (InGa)N QDs. The FWHMs of the QD signal are $0.00476 \text{ \AA}^{-1}$ in the direction of the diffraction vector and $0.00685 \text{ \AA}^{-1}$ in the perpendicular direction. The FWHMs of the signal are related to the mosaicity of the highly defective GaN buffer layer. It is possible to consider QDs grown on slightly tilted and twisted GaN blocks. Owing to the twist and tilt of an individual QD, the resulting diffraction pattern is merged into a diffuse cloud. Broadening of this diffuse intensity in the case of an asymmetric reflection (such as $10\bar{1}3$) is specified by the simultaneous influence of the tilt, twist and possible gradient of the In content, which makes its interpretation complicated.

Using FIXD, the signals of the (InGa)N QDs were successfully resolved owing to the simultaneous Q_z sensitivity of the scattering vector and grazing-incidence conditions. Without Q_z sensitivity (like in the case of GID; Krause *et al.*, 2014) both clouds merge vertically, and evaluation of relaxation and composition is not feasible for the small low indium concentration (InGa)N QDs. The possibility of obtaining the Q_z resolution in combination with the high surface sensitivity due to the grazing incidence is the obvious advantage of FIXD compared with conventional GID. The method could be successfully applied not only to free-standing (InGa)N QDs, but also to any free-standing and buried semiconductor nanostructures, as well as to a single-layer or multilayer structure.

4. Conclusions

In this manuscript we have described mathematical methods and experimental procedures to perform X-ray diffraction experiments in an asymmetric skew geometry with a fixed angle of incidence (FIXD). The described approach is suited to application on both laboratory setups and synchrotron beamlines. It includes finding an asymmetric non-coplanar reflection, the realization of a scan of this reflection at a fixed incidence angle and, finally, the evaluation of the measured intensity distribution in three-dimensional reciprocal space. We have demonstrated the potential of this approach by

investigating (InGa)N quantum dots with low In content grown on a GaN buffer layer. Traditional X-ray diffraction methods fail to reveal the signal from the low-In quantum dots because of intense diffuse scattering from the GaN buffer layer. By applying FIXD in grazing-incidence conditions, we have successfully measured the signal of these low-In (InGa)N quantum dots and were able to estimate their mean In content to be about 3.5%.

Acknowledgements

The authors acknowledge beamtime at the ESRF and express their gratitude to the staff of the ID01 beamline for their support during the experiment.

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