Determination of the stacking fault density in highly defective single GaAs nanowires by means of coherent diffraction imaging

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Abstract
Coherent x-ray diffraction imaging is used to measure diffraction patterns from individual highly defective nanowires, showing a complex speckle pattern instead of well-defined Bragg peaks. The approach is tested for nanowires of 500 nm diameter and 500 nm height predominately composed by zinc-blende (ZB) and twinned zinc-blende (TZB) phase domains. Phase retrieval is used to reconstruct the measured 2-dimensional intensity patterns recorded from single nanowires with 3.48 nm and 0.98 nm spatial resolution. Whereas the speckle amplitudes and distribution are perfectly reconstructed, no unique solution could be obtained for the phase structure. The number of phase switches is found to be proportional to the number of measured speckles and follows a narrow number distribution. Using data with 0.98 nm spatial resolution the mean number of phase switches is in reasonable agreement with estimates taken from TEM. However, since the resolved phase domain still is 3–4 times larger than a single GaAs bilayer we explain the non-ambiguous phase reconstruction by the fact that depending on starting phase and sequence of subroutines used during the phase retrieval the retrieved phase domain host a different sequence of randomly stacked bilayers. Modelling possible arrangements of bilayer sequences within a phase domain demonstrate that the complex speckle patterns measured can indeed be explained by the random arrangement of the ZB and TZB phase domains.

Introduction
In recent decades, growth techniques for semiconductor nanowires (NWs) have constantly improved, and meanwhile NWs are considered as potential building blocks for novel electrical and optoelectronic devices (Joyce et al 2011, Zimmeler et al 2010, Dayeh 2010). In comparison to thin film epitaxy, the NW technology allows to form heterostructures between materials of different lattice constant with a reduced density of defects at the interface compared to planar layer growth (Caroff et al 2008). However, the performance of the nanowire based devices strongly depends on the structural properties of the individual NWs (Han et al 2013). Therefore, detailed understanding and characterization of the atomic scale internal structure of the NWs is mandatory. It has been shown that GaAs and InAs NWs usually grow along the cubic [111] direction on a [111] oriented substrate (Heiss et al 2008). The growth can be understood as the stacking of hexagonal dense packed 2D layers, which stack in a characteristic zinc-blende (ZB) sequence ABCABC in the bulk phase. However, in nanowire growth commonly also other stacking sequences like the wurtzite (WZ) structure (ABAB) are observed, together with the occurrence of zinc-blende rotational twins, varying typically randomly between different nanowires (Biermanns et al 2013). NWs of uniform phase are hard to grow. In most cases NWs are composed of two or more of these phases and additional stacking faults (SFs). In addition more complicated stacking sequences similar to SiC may be realized (Kriegner et al 2011). Zinc-blende and wurtzite phases exhibit different physical properties such as lattice parameters (Kriegner et al 2011) and electronic band gap (De and Pryor 2010). It is
expected that the electrical and opto-electrical properties of a nanowire device may depend on the number of phase switches and the distribution of SFs within the NW because of their different band gap parameters (Jahn et al 2012). As the detailed microstructure of different NWs grown on the same sample may vary strongly, a destruction free experimental technique is desired that can provide the phase composition of a desired NW in order to correlate specific physical properties, e.g. the emission spectra or charge carrier mobility, with the crystallographic structure of the particular NWs.

In recent years, nanobeam x-ray diffraction techniques have been developed to study individual NWs (Biermanns et al 2009, Stangl et al 2013). Using x-ray nano-diffraction, the composition and arrangement of ZB and WZ phases within different NWs on the same substrate could be determined in case of nanowires with a rather high structural perfection (Biermanns et al 2012a). Compared to this, the diffraction from a highly defective NW with coherent x-ray radiation provides a complicated speckle pattern instead of a well-defined Bragg peak for specific directions, extending over a wide range in reciprocal space (Favre-Nicolin et al 2010). These speckle patterns principally contain all information about the structural phase composition of individual nano-objects. However, a direct inversion of the diffraction pattern is not possible, because the phase of the diffracted amplitude is lost during the measurement. To overcome this well-known phase problem, different experimental and numerical techniques have been developed in recent years. Using the method of phase retrieval, the lost phase is retrieved from the diffraction pattern using iterative algorithms taking into account known constraints in real and reciprocal space (Gerchberg and Saxton 1972, Fienup 1978).

However, the retrieval of stacking fault defects in NWs from speckle patterns is not straightforward and suffers from the difficulty that the diffraction pattern resembles mainly one-dimensional information. A first qualitative discussion of CDI pattern taken from a GaAs NW has been published by (Gulden et al 2011). A possible SF sequence within a single InAs NW has been reconstructed from a measured speckle pattern by means of a Monte Carlo approach (Favre-Nicolin et al 2010). However, it has not been shown that this is an unique solution. Instead of a Monte Carlo approach one may use iterative phase retrieval to find other solutions.

Although CDI has been demonstrated to evaluate strains (Robinson and Harder 2009, Cha et al 2013) or strains induced by single dislocations (Clark et al 2015) in nearly perfect single crystalline nano-objects, there is no application of the method to highly defective nanostructures so far.

In this article, we demonstrate a new approach of phase retrieval for highly defective individual NWs. We use the measured 2-dimensional intensity distribution and consider constraints for amplitude and phase variation perpendicular to the growth axis during the phase retrieval process (Minkevich et al 2007). The approach is tested for highly defective GaAs NWs which are exclusively composed by zinc-blende (ZB) and twinned zinc-blende (TZB) phase units, but contain no extended wurtzite phases. Few of these NWs grown on patterned GaAs [111] substrate have been investigated by coherent x-ray diffraction imaging (CDI) at the beamline ID1 of ESRF with two different experimental resolutions, providing 3.48 nm and 0.98 nm spatial resolutions in real space. Whereas the speckle amplitudes and distribution are perfectly reconstructed, no unique solution could be obtained for the phase structure. The number of phase switches is found to be proportional to the number of measured speckles and follows a narrow number distribution which for data with 0.98 nm spatial resolution is in reasonable agreement with estimates taken from TEM. Since even in this case the resolved phase domain still is 3–4 times larger than a single GaAs bilayer we explain the non-ambiguity in phase reconstruction by the fact that depending on starting phase and sequence of subroutines used the retrieved phase domain hosts a different sequence of randomly stacked bilayers.

**Theory**

The characterization of nano-objects is possible by means of CDI. For crystalline nanostructures, the diffraction intensity can be approximated in terms of kinematical scattering theory and the measured amplitude \( A(\overrightarrow{q}) \) of the scattered field can be expressed as the Fourier transform \( F_{\overrightarrow{r} \rightarrow \overrightarrow{q}}(\rho(\overrightarrow{r})) \) of the electron density of the object:

\[
A(\overrightarrow{q}) = \int_{V} \rho(\overrightarrow{r}) \exp \{ i2\pi \overrightarrow{q} \cdot \overrightarrow{r} \} d\overrightarrow{r} = F_{\overrightarrow{r} \rightarrow \overrightarrow{q}}(\rho(\overrightarrow{r})).
\]  

Within a conventional diffraction experiment, only \( |A(\overrightarrow{q})| = |F_{\overrightarrow{r} \rightarrow \overrightarrow{q}}(\rho(\overrightarrow{r}))| \) is measured. \( \overrightarrow{q} = \overrightarrow{k_f} - \overrightarrow{k_i} \) is the momentum transfer vector determined by diffracted and incident x-ray beams with wave vectors \( \overrightarrow{k_f} \) and \( \overrightarrow{k_i} \), respectively. A GaAs nanowire growing along a cubic [111] direction can be considered as a sequence of hexagonal close packed bilayers of group III and group IV elements. For a perfect zinc-blende structure, subsequent layers stack in a sequence ABCABC, whereas a twinned structure may be described by the inverted sequence CBACBAC. In this notation, subsequent layers are laterally shifted with respect to an initial layer, arbitrarily defined as type A (figure 1(a)). If we neglect the detailed facets of the nanowire, we may approximate that all of these close packed planes can be described by the same arrangement of atoms within the plane, however subsequent planes are shifted with respect to each other both along the growth direction and...
In this case, the electron positions in an arbitrary stacking sequence can be expressed by vectors

\[
\mathbf{r} = \mathbf{r}_{n-\text{plane}} + \mathbf{r}_{\text{plane}} + \mathbf{r}_m,
\]

where \(\mathbf{r}_{n-\text{plane}}\) denotes the relative position of each atom within the atomic bilayers \(A, B\) or \(C\) with respect to the center of the plane, \(\mathbf{r}_m\) describes the electron distribution around the \(n\)th atom in the bilayer and \(\mathbf{r}_{\text{plane}}\) describes the position of the atomic bilayer along the growth axes (figure 1(a)). The different stacking sequences can be described using a hexagonal lattice system with \(c = \sqrt{3}a_c\) and \(2a = \sqrt{2}a_c\) directed parallel and perpendicular to the growth axes, respectively, where \(a_c\) is the lattice parameter of cubic GaAs. Now we can assume that the nanowire is composed by \(M\) atomic bilayers shifted laterally and vertically with respect to each other. This displacement vector will be expressed by combination of the two vectors \(\mathbf{R}_z\) and \(\mathbf{R}_m\), describing the vertical (\(z\)) and lateral shift of the atomic planes with respect to the origin. \(\Delta z = c/3\) represents the distance between two bilayers along the \(z\)-axis. The origin of the hexagonal lattice is chosen in the center of the first atomic layer, \(A\), at \(\mathbf{R}_0 = (0, 0, 0)\) and \(\Delta z = 0\); the next following bilayer, \(B\), will fill one of the two potential minima of layer \(A\) at position \(\mathbf{R}_B = (\frac{2}{3}a_c, \frac{1}{3}a_c, \Delta z)\) and \(|\Delta z| = \Delta z = c/3\). In ZB structure the third layer \(C\) will fill the remaining potential minima not filled by layer \(B\) centered at position \(\mathbf{R}_C = (\frac{1}{3}a_c, \frac{2}{3}a_c, \Delta z)\) and \(|\Delta z| = 2\Delta z = 2c/3\) (figures 1(a), (b)). In WZ structure the third layer will be centered at \(\mathbf{R}_C = \mathbf{R}_A\) again. With this notation displacement vector \(\mathbf{r}_{\text{plane}}\) will take the following form

\[
\mathbf{r}_{\text{plane}} = m\mathbf{R}_z + \mathbf{R}_{m||},
\]

where: \(\mathbf{R}_{m||} = \begin{cases} R_A, & m = 1, 4, 7, 10 \ldots \\ R_B, & m = 2, 5, 8, 11 \ldots \\ R_C, & m = 3, 6, 9, 12 \ldots \end{cases}\) and \(\mathbf{R}_z = \Delta z\frac{2}{|\Delta z|} \mathbf{z}\).

We now model the positions of the atoms in the discrete lattice, \(P_{\text{atom}}(\mathbf{r})\), by a sequence of Dirac impulses:

\[
P_{\text{atom}}(\mathbf{r}) = \sum_{n_a} \sum_{n_b} \delta(\mathbf{r} - \mathbf{r}_{n-\text{plane}}(n_a) - \mathbf{r}_{\text{plane}}(n_b)),
\]

where \(n_a\) and \(n_b\) are the numbers of atoms in a bilayer and the number of bilayers, respectively. The individually different electron distribution around each atom in a bilayer is modeled by a continuous function \(\rho_{\text{el}}(\mathbf{r}_m, \mathbf{r})\) where the second parameter explicitly expresses the dependency of the individual electron distribution of the absolute position of the center electron in the grid. Hence we can express the total electron distribution as a convolution of the electron distribution around the individual atoms with the positioning function of the atoms in the grid.

![Figure 1](image.png)

*Figure 1.* (a) Arrangement of the atomic planes \(A\) (blue), \(B\) (red) and \(C\) (green). \(O\) represents the origin of the coordinate system \((\mathbf{r}_{n-\text{plane}} = 0 \text{ and } \mathbf{R}_A = (0, 0, 0))\). \(\Delta z\) points along the growth axis. (b) Top view: vectors \(\mathbf{R}_B\) and \(\mathbf{R}_C\) represent the shift of the origin of the layers \(B\) and \(C\) in regards to the origin of the layer \(A\) along the horizontal direction (perpendicular to the growth axes). \(\mathbf{a}_1\) and \(\mathbf{a}_2\) are the primitive lattice vectors of the hexagonal base.
\[ \rho(\mathbf{r'}) = P_{\text{atom}}(\mathbf{r'}) \ast \rho_{\text{dec}}(\mathbf{r'}, \mathbf{r}) = \sum_{n_0} \sum_{n} \delta(\mathbf{r'} - \mathbf{r}_{\text{in-plane}}(n_0) - \mathbf{r}_{\text{plane}}(n)) \ast \rho_{\text{dec}}(\mathbf{r'}, \mathbf{r}) \]
\[ = \sum_{n_0} \sum_{n} \rho_{\text{dec}}(\mathbf{r'} - \mathbf{r}_{\text{in-plane}}(n_0) - \mathbf{r}_{\text{plane}}(n_0) + \mathbf{r}_{\text{plane}}(n_0)). \]

If we had only one atom to consider the Fourier transform of the corresponding electron density would clearly depend on the position of the atom, giving rise to a space frequency expression:
\[ A_{\text{dec}}(\mathbf{q}, \mathbf{r'}) = F_{\mathbf{r'} \rightarrow \mathbf{q}} \{ \rho_{\text{dec}}(\mathbf{r'}, \mathbf{r}) \} = \rho_{\text{dec}}(\mathbf{q}, \mathbf{r}). \]

Consequently the out coming overall Fourier spectrum of the total electron density is the linear superposition of all individual spectra:
\[ A(\mathbf{q}) = \sum_{n_0} \sum_{n} \rho_{\text{dec}}(\mathbf{q}, \mathbf{r}_{\text{in-plane}}(n_0) + \mathbf{r}_{\text{plane}}(n_0)) \cdot \exp(-i2\pi \mathbf{q}^T \cdot (\mathbf{r}_{\text{in-plane}}(n_0) + \mathbf{r}_{\text{plane}}(n_0))). \]

If we now assume that the individual electron distributions do not depend on the layer, we may simplify the formula:
\[ A(\mathbf{q}) = \sum_{n_0} \sum_{n} \rho_{\text{dec}}(\mathbf{q}, \mathbf{r}_{\text{in-plane}}(n_0)) \cdot \exp(-i2\pi \mathbf{q}^T \cdot (\mathbf{r}_{\text{in-plane}}(n_0) + \mathbf{r}_{\text{plane}}(n_0))) \]
\[ = \sum_{n_0} \sum_{n} \rho_{\text{dec}}(\mathbf{q}, \mathbf{r}_{\text{in-plane}}(n_0)) \cdot \exp(-i2\pi \mathbf{q}^T \cdot \mathbf{r}_{\text{in-plane}}(n_0)) \cdot \exp(-i2\pi \mathbf{q}^T \cdot \mathbf{r}_{\text{plane}}(n_0)) \]
\[ = \sum_{n_0} \sum_{n} f_n(\mathbf{q}) \cdot \exp(-i2\pi \mathbf{q}^T \cdot \mathbf{r}_{\text{in-plane}}(n_0)) \cdot \sum_{n} \exp(-i2\pi \mathbf{q}^T \cdot \mathbf{r}_{\text{plane}}(n_0)), \]

where the first sum runs over the atoms within the bilayers determining the intensity of a Bragg peak. The second term sums up over all bilayers of the irradiated volume and determines the position of a Bragg peak. \( f_n(\mathbf{q}) \) is the atomic form factor. Introducing \( S_{\text{layer}}(\mathbf{q}) \) as the in-plane structure factor identical for each layer, we obtain
\[ A(\mathbf{q}) = S_{\text{layer}}(\mathbf{q}) \cdot \sum_{n_0} \exp(-i2\pi \mathbf{q}^T \cdot \mathbf{r}_{\text{plane}}(n_0)) \]

where:
\[ S_{\text{layer}}(\mathbf{q}) = \sum_{n_0} f_n(\mathbf{q}) \cdot \exp(-i2\pi \mathbf{q}^T \cdot \mathbf{r}_{\text{in-plane}}(n_0)) \]

and:
\[ f_n(\mathbf{q}) = \rho_{\text{dec}}(\mathbf{q}, \mathbf{r}_{\text{in-plane}}(n_0)). \]

Taking into account equation (3) and introducing momentum transfer vector as a combination of its in-plane and out of plane components \( \mathbf{q} = \mathbf{q}_a + \mathbf{q}_b \) equation (9) can be rewritten as
\[ A(\mathbf{q}) = S_{\text{layer}}(\mathbf{q}) \cdot \sum_{m=n_0} e^{im\mathbf{q}_a \cdot \mathbf{R}_b} e^{im\mathbf{q}_b \cdot \mathbf{R}_a}. \]

Using \( R_{\text{m}} \) as defined in (3), we note that the stacking sequence of bilayers leads to phase shifts of the form
\[ \Phi_{\text{b}} = \exp \left\{ i \frac{2\pi}{3} (2h + k) \right\} \quad \text{or} \quad \Phi_{\text{c}} = \exp \left\{ i \frac{2\pi}{3} (h + 2k) \right\} \]

in the reciprocal lattice of the structure defined in figure 1, where \( h \) and \( k \) define the Miller indices of the plane of \( \mathbf{q}_a \). Hence, all reflections with \( h - k \neq 3N \), \( N \in \mathbb{Z} \) are sensitive to the exact arrangement of bilayers and in hence stacking faults (Warren 1990, Barchuk et al. 2011). In particular, this holds true for reflections with \( (h, k) = (1, 0) \), leading to phase shifts \( \varphi \in [0, -2\pi/3, 2\pi/3] \). Neglecting other structural defects except SFs, the density and subsequently the scattering amplitude of the whole NW is uniform. Therefore the total scattering amplitude of the NW can be described by a uniform density but varying scattering phases depending on the particular stacking sequence of bilayers within the NW. Using coherent nano-diffraction the particular stacking sequence of a NW becomes visible by a specific speckle pattern. By use of phase retrieval analysis it is possible to retrieve the lost phase information.

**Experimental**

The investigated NWs with a diameter around 500 nm have been grown in a periodic array by selective area metal-organic vapor phase epitaxy on a GaAs substrate. The spacing between the NWs has been determined by a lithography process, resulting in a square arrangement of NWs with 3 \( \mu \)m separation. Details of the growth process can be found in (Biermanns et al. 2009). As known from ex situ transmission electron microscopy investigations, the nanowires grow predominately in the zinc-blende phase with a high density of rotational
twins (Bauer et al 2010). The x-ray diffraction experiment has been performed at beamline ID1 at ESRF in coplanar diffraction geometry. The incoming monochromatic x-ray beam with a photon-energy of 9 keV was focused down to a spot size of 500 $\times$ 300 nm$^2$ using a Fresnel Zone Plate (FZP) allowing for illumination of the entire NW. Coherent illumination has been obtained by partial illumination of the FZP with an aperture.
matching the size of the coherent volume of the beamline (Mastropietro et al 2011). A sketch of the experimental setup is presented in figure 2. The incident coherent x-ray beam is diffracted from a single nanowire at Bragg condition, and the intensity pattern is recorded at the detector. 3D diffracted speckle patterns were recorded by scanning the incidence and exit angles $\alpha_i$ and $\alpha_f$ simultaneously with different step sizes taking 2D images at each step.

Using a 2-dimensional MAXIPIX detector, three dimensional intensity pattern in reciprocal space have been recorded around the cubic (331) reflection, which is in hexagonal units the (1, 0, 7)$^h$ reflection, of the GaAs lattice sensitive to stacking faults within a $q_z$—range of 1.8 nm$^{-1}$ (low resolution case—LR, two nanowires) and 6.38 nm$^{-1}$ (high resolution case—HR, one nanowire) corresponding to spatial resolutions in real space of 3.48 nm and 0.98 nm, respectively. For the analysis of diffraction patterns using phase retrieval algorithms, the data has been integrated along the direction perpendicular to the forward scattered direction, leading to two-dimensional intensity distributions. Details of the experimental setup are described elsewhere (Biermanns et al 2009).

Figures 3(a)/(b) shows measured reciprocal space maps of NW1 and NW2 within a $q_z$ range between 47 nm$^{-1} < q_z < 48.8$ nm$^{-1}$, using a non-focused x-ray beam close to the (331) reflection of the underlying GaAs substrate. Figure 3(c) shows an enlarged $q_z$ range between 44.17 nm$^{-1} < q_z < 50.55$ nm$^{-1}$ measured from NW3. It is important to note that the $q_z$ range measured in figure 3(c) is the maximum range accessible in the experimental geometry used. Extension of this range was not possible due to collision risks of the setup. Due to the coherence properties of the incoming x-ray beam and the different individual defect distributions within the NWs, different speckle pattern are observed in all cases. The phase retrieval analysis discussed in the following section has been performed for all three NWs. However, details of the procedure are demonstrated for the pattern shown in figures 3(a) and (c) only. Since different ranges along the $q_z$, (taken for NW1 and NW3) are corresponding to different spatial resolutions the average uniform length of a phase segment is approximately 12 and 4 bilayer long, respectively. In order to apply the phase retrieval approach introduced above we made the following assumptions: (1) the NW is entirely composed by GaAs bilayers stacked along the growth axis, (2) structural defects except phase switches are excluded, (3) the electron density within a bilayer plane is homogeneous, (4) strain impact is excluded; in particular, this considers axial strain due to possible inclusion of wurztite units and strain relaxation towards the outer NW side planes. These assumptions reflect the current knowledge about the real structure of MBE and MOVPE grown GaAs NWs (Biermanns et al 2012b, Davydok et al 2012, Kriegner et al 2011, Han et al 2013, Jahn et al 2012, Joyce et al 2011). (5) The length of measured phase domain depends on experimental resolution. Neglecting any other origin for phase shifts it is supposed that the measured phase domain is composed by multiples of such bilayers. Since the phase shift between neighbor bilayers is $\phi \in [0, −2\pi/3, 2\pi/3]$ the phase shift of a uniform phase domain has to show phase $\phi^i \in [0, −2\pi/3, 2\pi/3]$. These constraints are taken into account in the phase retrieval procedure and are discussed in more detailed in the following chapters.

**Phase retrieval**

SFs are arranged along the growth direction only, suggesting the use of a 1D function. Unfortunately, success in unique 1D phase retrieval suffers from the fact that for the Fourier transform of the complex functions with the same amplitude but different phases can almost not be distinguished. To overcome this limitation when working with 1D data, we aim to retrieve a quasi 2D speaks pattern obtained from the measured 3D data. Based on the constraints introduced above we suppose that the scattering amplitude is constant along the NW (no other structural defects than SF) and (6) the scattering phase of a phase domain can only assume discrete values from the set \{-2\pi/3, 0, 2\pi/3\} considering the NW is composed by ZB and TZB phase units only. That mean we have to retrieve the phase of uniform phase domains composed by a certain sequence of bilayers.

We can apply generally accepted recipes of phase retrieval and combinations of different phase retrieval algorithms like Error-Reduction (ER) (Fienup 1982), Hybrid Input-Output (HIO) (Fienup 2013), Gerchberg-Saxton (GS) (Gerchberg and Saxton 1972), Minkevich approach (Minkevich et al 2007) and the Shrink-Wrap method (SW) (Marchesini et al 2003) to retrieve the missing phase information. The different steps of the algorithm are presented in figure 4. They are arranged in 5 blocks optimized via a trial and error approach (see supplementary material). Therefore this algorithm is specifically adjusted for data analysis of the problem in question. We arbitrarily define an area of $910 \times 690$ nm$^2$ where the object most likely will be found in its center. Within this confined area we generate an array of random phases which are multiplied to the experimental amplitudes determined from square root of measured intensities. In the first part of our reconstruction, we retrieve the shape of the object by applying the steps described in blocks 1, 2 and 3 of figure 4: The ER algorithm (about 700 iterations— block 1) followed by the alternative use of HIO and ER algorithm where the relaxation parameter in HIO is set to 0.9 (block 2). After about 3000 iterations the next step is the stepwise shrinkage of the support area to the object size performed by the SW method (block 3). It takes about $10^4$ iterations until the shape of the object is finally reconstructed (end of the block 3). In the next part, we retrieve the phase
information of the object. Here we apply the GS algorithm with a new constraint in direct space which takes into account that the density, i.e. the scattering amplitude, of the object is homogeneous over the whole length of the NW and also the phase is assumed to be constant in the direction perpendicular to the growth axis (blocks 4 and 5). These new constraints have been applied for the first time by Minkevich (Minkevich et al 2007). With these inputs from block 5 the GS algorithm is applied performing up to $10^3$ iterations each. Starting after about $\frac{3}{4}$ of these iterations the phase is homogenized along the horizontal direction and set to be either $0$, $-2\pi/3$ and $2\pi/3$ with the help of a force constraint (block 5): The phase value is set to $-2\pi/3$ when the iterated phase value is smaller than $0.4^\circ (-2\pi/3)$ and is set to $2\pi/3$ when phase value is higher then $0.4^\circ 2\pi/3$. For the remaining cases the phase value is set to zero. With the known shape and amplitude the last step of the algorithm is a combination of GS and Minkevich approach and the phase of the nanowire in real space is retrieved for the different trial phases. Above described phase retrieval algorithm was performed using $10^4$ different starting phases for each speckle pattern shown in figures 3(a), (b) and (c).

As shown in supplementary material the algorithm used is stable against change in sequences of application of the different algorithms (ER, HIO, GS aso.) and changes of numbers of the respective iteration cycles.
Results

Figure 5 is showing the distribution of the retrieved amplitude in reciprocal space for two different NWs with two trial phases and its comparison to the experiment (figure 5 (left NW1, right NW3) measured in LR and HR, respectively. Because of the constraints made for density and phase the reconstructed speckle pattern is center-symmetric with respect to the $q_x$ axis. Except this symmetry, figure 5 shows a reasonable agreement between measured and retrieved speckle pattern for both cases.

For a better comparison, the reconstructed pattern are integrated along $q_x$ and projected onto the $q_z$ axis. Figure 6 shows a comparison of the obtained line-profiles, showing that in both cases the retrieved speckle pattern agrees in most of the details with the experiment. That concerns the $q_z$ positions of maxima and minima and the total number of peaks. The achieved agreement of experimental and simulated data is similar for NW1.

Figure 6. Comparison of the linear profiles of the measured (red lines) and simulated (black and blue) amplitudes of speckle patterns. Upper and lower profiles correspond to NW1 and NW3, respectively. For better visibility experimental profile, reconstruction 1 and reconstruction 2 are shifted vertically in respect to each other.

Figure 7. Comparison of the reconstructed phase object of the NW in case of different trial phases for LR (NW1) and HR (NW3).
can simulate the speckle phases numerically and calculate the corresponding speckle pattern process, for example, bilayer A can be followed only by bilayer B or C. By random stacking of these segments we any combination of the mentioned phase segments is possible taking into account that during the growth unique solution of distribution of phases could be found independent from the spatial resolution used in segments and different arrangements of phase domains are visible in the reconstruction. This means that no 3. In contrast to amplitudes the reconstruction of phases is not unique using different trials. Different phase of phase domains

between the experimental and simulated speckle pattern is found for the case of completely random distribution of speckles as found close to supplementary material

measured from single nanowires. Therefore we conclude that the presented algorithm is well suited to retrieve the speckle patterns measured from single nanowires.

Finally the reconstructed phase sequences are presented in figure 7 for two different trials on NW1 and NW 3. In contrast to amplitudes the reconstruction of phases is not unique using different trials. Different phase segments and different arrangements of phase domains are visible in the reconstruction. This means that no unique solution of distribution of phases could be found independent from the spatial resolution used in experiment.

Although the absolute arrangement of phase segments differs among different reconstructions, the number of retrieved phase switches is rather unique for a given NW and spatial resolution. To quantify this number of planar defects, we applied $10^4$ random phase trials to retrieve the data taken from the same nanowire. It turns out that the number of individual phase domains changes slightly using different trials. The number distribution of reconstructed phase can be approximated by a Gaussian with narrow FWHM. In detail, using $10^4$ trials the average number of phase domains is about $113 \pm 5$ for NW1 and $109 \pm 5$ for NW2 (LR case) but $240 \pm 6$ for NW3 (HR case). That means the number of retrieved phase switches equals within the error bars comparing NW1 and NW2. The mean number of phase switches does change in very little using different sequence of subroutines during the phase retrieval (see supplement material, S1). On the other hand, the retrieved particular number of phase switches is same using the same starting phase and running the same sequence of subroutines during the phase retrieval procedure. The number of retrieved phase switches is a function of experimental resolution. In the HR case (NW3) the number of resolved phase switches is more than two times larger compared to the LR case (NW1 and NW2) which correlates with the more than three times higher spatial resolution and the number of resolved speckles in experiment. However, the spatial resolution of 0.98 nm in HR is still 3–4 times larger compared to the length of a GaAs bilayer which might be sufficient to distinguish between a ZB or TZB unit cell (see later), which is impossible in the LR case where the spatial resolution of 3.48 nm corresponding to the size of 12 bilayers or 3 ZB/TZB unit cells. Considering the total height of the nanowire, the data shown in figure 8 correspond to a mean defect density of $0.23 \pm 0.01$ defects/nm, $0.22 \pm 0.01$ defects/nm for NW1 and NW2 and $0.48 \pm 0.01$ defects/nm for NW3. That means that the phase segments resolved in the LR case must contain 1–2 non visible additional defects compared to the phase segments resolved in HR case. Although the defect density in HR case fits to the value taken from selected NW segments by means of HRTEM additional phase switches within the resolved phase domains cannot be excluded. In summary, we have to conclude that the detailed arrangement of phase switches cannot be retrieved by our approach.

However, since the phase retrieval approach is very complex we can try to model the phase sequence for the HR case in order to get more insight into the real structure of the NW and to explain the experimental speckle pattern. Since the spatial resolution of 0.98 nm still is longer than the ZB or TZB stacking sequence we model the size of a phase domain by 4 bilayers. Considering the given constrain for the speckle phases $\phi'$ a possible sets, but not unique, of bilayer stacking modeled as $A' = ACAB$, with $\phi' = 0$ and $B' = CBAB$ with $\phi' = -2\pi/3$ and $C' = CACB$ with $\phi' = +2\pi/3$. The choice of the $A'$, $B'$ and $C'$ was made such that in the simulated phase model any combination of the mentioned phase segments is possible taking into account that during the growth process, for example, bilayer A can be followed only by bilayer B or C. By random stacking of these segments we can simulate the speckle phases numerically and calculate the corresponding speckle pattern (see chapter S2 of supplementary material). Although we did not found complete fit of experimental data, reasonable agreement between the experimental and simulated speckle pattern is found for the case of completely random distribution of phase domains (see figure S2(b)). The result does not change qualitatively assuming another choice of model phases. Here we did not exclude subsequent stacking of equal phase segments. Considering clustering of speckles as found close to $q_z = 46 \text{ nm}^{-1}$ or $48 \text{ nm}^{-1}$ in experiment, for example, best agreement is found if equal phase sequences such as $A' A'' A''$, where $A'' = CABA$ and $A'' = BACA$, and so on, do not repeat more than 2–3
times. This verifies the suggestion that the NWs are composed by a complete random distribution of phase segments without major clustering of ZB or TZB units explaining the absence of sharp Bragg peaks.

**Discussion**

Using phase retrieval technique we tried to determine the number of phase domains from speckle data measured by CDI from individual highly defective GaAs nanowires. To overcome the limitation for phase retrieval of 1D system, we used 2D diffraction data and considered two major constraints: (1) the homogeneity of the NW density along the NW growth axis and (2) the homogeneity of density within a single sublayer. In addition we have chosen nanowires composed by ZB and TZB phases only.

Using the presented recipe we were able to reconstruct the speckle pattern taken from a single nanowire independent from the choice of the trial phase. In addition we were able to determine the mean number of phase switches in a single nanowire in an accuracy of less than 5%. This number depends on spatial resolution. In case of LR, the number of phase switches is equal for two measured objects which can be explained by the fact that all NWs have been grown onto the same substrate under same growth conditions. On the other hand the defect arrangements of both nanowires must be different because of the different speckle distribution measured in both cases. For the HR case the number of phase switches is more than twice compared to LR case. Because here the resolution is close to the length of a single ZB or TZB unit the extracted number of phase switches might be close but not equal to the real value. Indeed, HRTEM inspections taken from a small segment of another NW, from the same wafer (not shown here), give a SF density (0.37 nm\(^{-1}\)) which is close to the value obtained HR CDI experiment. In consequence, the \(q_z\) range probed in LR case revealed to be too small in order to characterize these highly defective NWs.

There are several reasons why we were not able to determine the exact arrangement of phase domains. First, we still considered a relative small range in reciprocal space which is \(\Delta q_z = 6.38\) nm\(^{-1}\) for NW3. This limits the resolution in direct space to \(d = 0.98\) nm which corresponds to 3–4 atomic bilayers. All phase switches at distances smaller than \(d\) are not accessible. Since the bilayer sequence is random and cannot be counted in numbers of ZB or TZB units we explain the non-ambiguity in phase reconstruction mainly by the fact that depending on starting phase and sequence of subroutines used during the phase retrieval the retrieved phase domain may host a different sequence of bilayers. Thus the assignment of a certain bilayer sequence to a particular phase domain differ from trail to trail and a hidden additional phase switch becomes visible in trail but not in another one.

The problem of limited resolution can be overcome by measuring a three larger range in reciprocal space, but single bilayer resolution is hardly to be realized in an experiment. A similar approach has been developed to quantify the number of dislocations introduced within a finite crystal volume by means of coherent x-ray diffraction (Jacques *et al.* 2013). In this case, similar speckle patterns have been observed whose distribution in reciprocal space can be explained by different defect distributions in real space. However, the authors pointed out that comparing the maximum intensity of the speckle pattern with a defect free reference structure can efficiently be used to determine the number of defects. For the case of nanowires, this is usually not possible as long as no phase pure structures can be grown. Nevertheless, the presented method allows the destruction free analysis of the defect density with rather high accuracy.

Second, different spatial arrangements of defects may inherently give rise to identical speckle patterns. In case of a phase object composed by 4 atomic bilayers each, sequences like ABAC, ACAB, BACA…, provide the same residual phase. The arrangement of these sequences and its inverted sequence along the growth direction will produce the same amplitude in reciprocal space. We tried to identify such similarities in the retrieved phase patterns among different trials but we could not find similarities of such sequences of length exceeding more than ten bilayers. By these reasons it is rather impossible to model the phase sequence in the LR case.

Third, the appearance of rather small domains of wurtzite phase cannot be completely excluded. It would contribute by an additional phase shift even if the distance between two subsequent twin phases is only one monolayer. The appearance of wurtzite phase segments is not included in our model so far (see constrain No (4)). The possible influence of WZ inclusions on the uniqueness of phase retrieval is a matter for future investigation.

Technically the quality of phase retrieval can be improved by ptychographic imaging, i.e. scanning the nanowire by the coherent beam from top to the bottom. The additional information gained by collecting diffraction patterns with known overlap of the real space amplitude can speed up the convergence of procedure. However, the practicability of ptychography for analysis of as grown single NWs onto substrate is challenging and has to be demonstrated.

In summary, our approach allows for a quantitative defect analysis of single nanowires by use of coherent x-ray diffraction imaging accompanied by a modified phase retrieval algorithm. For the investigated samples,
the determined mean defect density varies among the NWs grown on the same substrate and corresponds to the case of a random stacking of bilayers preventing the appearance of detectable Bragg peaks. This information might already be sufficient for many practical applications such as improvement of crystal growth and understanding of various physical properties on the nanoscale scale. It is a matter for future to show whether or not it is possible to determine the detailed defect arrangement in NWs with much lower defect density by phase retrieval. Considering recent achievements (Holler et al. 2014) the application of ptychographic methods to nanowires is most challenging.

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