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Measuring three dimensional strain and structural defects in a single InGaAs nanowire using coherent x-ray multi-angle Bragg projection ptychography

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3 strain imaging; stacking faults

4 **Abstract**

5 III-As nanowires are candidates for near infrared light emitters and detectors that
6 can be directly integrated onto silicon. However, nanoscale to microscale variations
7 in structure, composition, and strain within a given nanowire, as well as variations
8 between nanowires, pose challenges to correlating microstructure with device perfor-
9 mance. In this work, we utilize coherent nano-focused x-rays to characterize stacking
10 defects and strain in a single InGaAs nanowire supported on Si. By reconstructing
11 diffraction patterns from the $2\bar{1}\bar{1}0$ Bragg peak, we show that the lattice orientation
12 varies along the length of the wire, while the strain field along the cross-section is
13 largely unaffected, leaving the band structure unperturbed. Diffraction patterns from
14 the $01\bar{1}0$ Bragg peak are reproducibly reconstructed to create three-dimensional im-
15 ages of stacking defects and associated lattice strains, revealing sharp planar boundaries
16 between different crystal phases of wurtzite (WZ) structure that contribute to charge
17 carrier scattering. Phase retrieval is made possible by developing multi-angle Bragg
18 projection ptychography (maBPP) to accommodate coherent nanodiffraction patterns
19 measured at arbitrary overlapping positions at multiple angles about a Bragg peak,
20 eliminating the need for scan registration at different angles. The penetrating nature
21 of x-ray radiation, together with the relaxed constraints of maBPP, will enable *in*
22 *operando* imaging of nanowire devices.

23 Nonplanar semiconductor heterostructures provide opportunities for novel and efficient
24 functionality over a broad range of applications. For example, ternary III-As nanowire
25 heterostructures are promising near-IR emitter/detectors for applications including on-chip
26 photonic information transfer due to their bandgap tunability and high electron mobility.¹⁻⁴
27 Additionally the nanowire geometry enables direct integration of III-V's onto silicon, as the
28 small interface area mitigates the formation of dislocations and anti-phase domain bound-
29 aries.⁵⁻⁷ However, III-As nanowires commonly exhibit nanoscale structural inhomogeneties
30 such as stacking faults, polytype insertions, and nanofaceting.^{8,9} In addition, composition
31 fluctuations in ternary alloys and the resulting lattice strain can modify the electronic
32 bandgap.^{8,10,11} When nanoscale defects occur together with composition and strain vari-
33 ations on multiple lengthscales, it can be challenging to establish the physical origins of
34 properties and device behaviors that are probed over microscale volumes. Therefore, the
35 necessary optimization of nanowire materials for specific electronic and optoelectronic de-
36 vices will require improved approaches to map local inhomogeneities in crystal structure and
37 composition throughout a nanowire, preferably using approaches that enable *in operando*
38 analysis.

39 Although the present work includes analysis of previously unreported perturbations in
40 nanowire structure encompassing nanometer to micron lengthscales, we are especially mo-
41 tivated to probe structural features that strongly influence optical emission and electronic
42 transport properties. In particular, there is a strong correlation between the density of
43 stacking defects and mobility in III-As nanowires.^{12,13} Comparing high resolution transmis-
44 sion electron microscopy (TEM) images of free standing nanowires with position dependent
45 field effect mobility measurements on InAs nanowire devices, Schroer et al¹² showed that
46 low densities of stacking faults localize electrons, leading to transport characteristics con-
47 sistent with quantum dot formation even in devices with low resistance Ohmic contacts.
48 Irber et al¹³ later showed that diffusive quantum transport in quasi 1-D sub-bands can be
49 observed in modulation doped GaAs nanowires even in the presence of stacking faults, but

50 as the stacking fault density increases, quantum features are washed out due to increased
51 scattering. It is also well established that crystal phase switching between wurtzite (WZ)
52 and zinc blende (ZB) polytypes, which exhibit a type II band alignment,¹⁴ leads to the
53 formation of quantum dots that act as single photon emitters.¹⁵ Further, Jahn et al¹⁴ ob-
54 served that GaAs nanowires with the same average WZ/ZB content may luminesce above
55 or below the ZB bandgap, dependent on the thickness of the ZB insertions. The interpreta-
56 tions advanced in the works cited above require *a priori* knowledge of the spatial variation
57 in the density of stacking defects. Methods such as transmission electron microscopy have
58 contributed greatly to our understanding of structure/property relationships in nanowires,
59 and the development of complimentary approaches compatible with more complex sample
60 environments (*e.g.* nanowire devices fabricated on standard Si wafers) is needed to deepen
61 our understanding.

62 A promising avenue lies in coherent x-ray diffraction imaging (CDI), which offers the ad-
63 vantage of probing strain and other structural features in nanowires over a larger field of view
64 on thicker substrates and embedded in operating devices. To date, 2D CDI methods have
65 been used to view longitudinal projections¹⁶ or cross sectional cuts¹⁷⁻¹⁹ of lattice strain in
66 III-V nanowires. However, scaling the approach to three dimensions and towards multi-scale
67 imaging is not straightforward because high resolution in 3D is needed of a high-aspect-ratio
68 sample. Existing 3D CDI techniques are not well suited for measuring extended structures
69 such as nanowires that are larger than the x-ray beam footprint. Further, abruptly vary-
70 ing features, such as crystal phase switching at few-nanometers length scales in nanowires,
71 cannot be reliably imaged using traditional CDI methods.^{20,21}

72 Here we overcome the limitations of conventional 3D CDI by adapting Bragg ptychog-
73 raphy, a variant of CDI based on scanning focused coherent x-ray beam measurements, to
74 robustly reconstruct 3D images of strain and stacking defects in single InGaAs nanowires.
75 The analysis focuses on InGaAs nanowires grown by catalyst-free molecular beam epitaxy
76 (MBE) which have been shown to have wide compositional tunability and can be used as

77 a foundation for epitaxial core-shell heterostructures^{22,23} for near-IR optoelectronics. We
78 demonstrate reconstruction of single stacking defects and lattice strain in InGaAs nanowires
79 on Si substrates with a spatial resolution better than 3 nm. To do so, we introduce an
80 implementation of Bragg ptychography (named multi-angle Bragg Projection Ptychography,
81 maBPP) in combination with coarse-scanning Bragg nanodiffraction analysis to provide a
82 holistic view of the hierarchical structure of a single InGaAs nanowire spanning from nanome-
83 ters to several microns. The methodological framework we present, and the proof of princi-
84 ple we demonstrate, can enable new insights into the impact of stacking faults and crystal
85 phase switching on the characteristics of individual substrate-supported nanowire devices *in*
86 *operando*.

87 Prior to describing the data and analysis, we briefly introduce 3D Bragg ptychography
88 and the motivation for the specific advance in methodology that was required to resolve
89 single stacking faults in a nanowire with a high density of these defects. We utilize 3D
90 Bragg ptychography as it satisfies many attributes necessary for characterizing complex
91 III-V nanowires: nanoscale resolution, sensitivity to different structural features, and the
92 potential for mapping extended crystals. With Bragg ptychography, nanoscale variations in
93 crystal structure can be imaged by numerically inverting coherent diffraction intensity pat-
94 terns measured in the vicinity of a Bragg peak.^{24,25} The approach utilizes a localized scanning
95 x-ray probe (typically focused with an x-ray optic) and entails measuring oversampled Bragg
96 coherent diffraction patterns at different overlapping scan positions at one²⁴ or more²⁵ angles
97 near the Bragg diffraction condition of an extended crystal. Gradient-based iterative inver-
98 sion algorithms have been developed²⁶⁻²⁸ to retrieve the phases (which cannot be measured
99 experimentally) of the intensity patterns in such a data set and to provide a real-space image
100 of the complex-valued structure factor of the measured Bragg peak. Variation in the phase
101 of these structure factor images can then be interpreted in terms of various phenomena, in-
102 cluding lattice displacement from elastic strain fields,²⁹ defects in atomic stacking order,^{30,31}
103 individual dislocations,³² and ferroelectric polarization.³³ Typically, these phenomena can be

104 observed with a spatial resolution of 5-50 nm, depending on the signal-to-noise ratio (SNR)
105 of the measurement and other factors such as scattering geometry, degree of probe overlap,
106 and choice of reconstruction algorithm.²⁸

107 Traditional 3D Bragg ptychography utilizes data sets with fine angular steps about the
108 Bragg peak (known as rocking curves),^{25,34} requiring that high SNR diffraction data be
109 collected for all angles at each probe position. 3D reconstruction algorithms require that
110 scan positions be commensurate at every diffraction angle to within a few percent of the
111 beam diameter, a requirement that is highly challenging with state-of-the-art hard x-ray
112 focusing optics that produce focii of <100 nm and operate in fly-scan mode.^{35,36} Alternatively,
113 methods such as Bragg projection ptychography (BPP and 3DBPP) have been developed
114 that only require a single angle measurement. Scanning Bragg nanodiffraction data at a
115 single-angle can then be inverted into 2D³⁷ and 3D²⁴ images of lattice structure within a
116 material. However, single-angle 3D BPP requires high diffraction angles ($>\sim 60^\circ$) that can
117 be difficult to reach experimentally and at which Bragg peaks scatter more weakly.

118 We address these challenges by utilizing a generalized 3D multi-angle Bragg projection
119 ptychography approach, which is described in more detail in the Algorithm Description
120 section. maBPP relaxes experimental constraints such that a set of coherent diffraction in-
121 tensity patterns measured at arbitrary angles and positions can be incorporated into a single
122 3D reconstruction, without requiring any position registration. Specifically, we implement
123 maBPP by adapting the Ptychographic Iterative Engine (PIE),²⁶ a phase retrieval algorithm
124 shown to be well suited for ptychographic imaging.

125 $\text{In}_{0.86}\text{Ga}_{0.14}\text{As}$ nanowires with diameters of 100-200 nm were grown by catalyst-free molec-
126 ular beam epitaxy under conditions similar to those in Reference 38. Nanowires of this
127 diameter range, and even larger diameters, are of interest for IR optoelectronics because
128 optical modes are insufficiently confined at smaller diameters.³⁹ HRTEM investigation of
129 similar samples revealed a primarily WZ crystal phase with a high density of stacking faults,
130 typically spaced by <10 nm. No extended regions (>1 nm) of ZB were observed at these

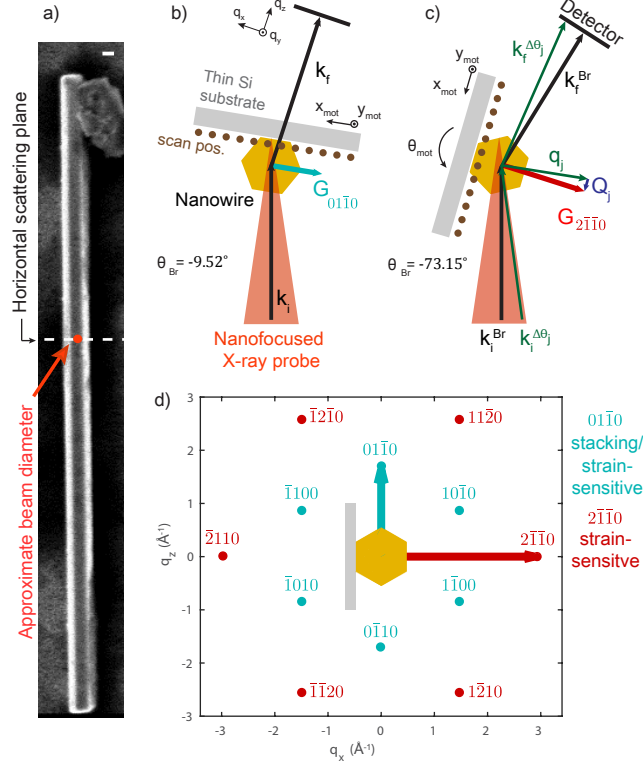


Figure 1: Experimental geometry at HXN beamline (a) SEM image of the investigated InGaAs nanowire. Scale bar is 100 nm. The focused x-ray probe (red circle) is approximately 50 nm in diameter. Scattering geometries used for the $01\bar{1}0$ (b) and $2\bar{1}\bar{1}0$ (c) conditions. θ_{Br} is the angle of the integrated intensity maximum of the rocking curve. k_i^{Br} and k_f^{Br} are the initial and final scattering vectors at θ_{Br} , defined by the momentum transfer vector G . $k_i^{\Delta\theta_j}$ and $k_f^{\Delta\theta_j}$ are the initial and final scattering vectors for the j th angle away from θ_{Br} , defined by the momentum transfer q_j , Q_{θ_j} away from G . (d) The reciprocal space lattice in the radial plane of the nanowire (cyan and red points) and a schematic of the facets of the InGaAs nanowire studied (yellow hexagon). The family of $2\bar{1}\bar{1}0$ peaks of the WZ lattice correspond to the $20\bar{2}$ family of peaks in the ZB structure, and they are sensitive to lattice strain fields within the nanowire. The $01\bar{1}0$ peaks have no analog in the cubic ZB structure. These peaks are sensitive to stacking faults in the WZ phase as well as a component of lattice strain. Bragg ptychography nanodiffraction area raster scans were performed on the same nanowire at the $2\bar{1}\bar{1}0$ and $01\bar{1}0$ Bragg peaks and reconstructed into complementary 3D images.

131 growth conditions.³⁸ To prepare a sample for structural imaging with maBPP, the nanowires
132 were drop-casted onto a 10- μm -thick silicon substrate that transmits hard x-rays prepared
133 for this application via selective etching and lithography by Norcada Inc., and the location of
134 nanowires relative to chromium fiducial markers on the substrate was determined with scan-
135 ning electron microscopy (SEM), prior to x-ray investigations. The SEM characterization
136 revealed that each nanowire was fixed to the substrate with an a -plane ($2\bar{1}\bar{1}0$) facet parallel
137 to the Si surface. (We adopt hexagonal four-index notation in this work consistent with the
138 hexagonal WZ crystal structure.) Figure 1(a) shows a SEM image of the 200-nm-diameter
139 nanowire investigated.

140 Bragg ptychography coherent nanodiffraction measurements were performed at the Hard
141 X-ray Nanoprobe (HXN) beamline of the National Synchrotron Light Source II (NSLS-
142 II).^{40–42} A coherently-illuminated x-ray zoneplate with an outermost zone width of 40 nm
143 was used to focus 10.4 keV monochromatic x-rays at the sample, forming a minimum spot
144 size of 49 nm with an 80 mm focal length. The wavefront of the probe was characterized
145 with standard direct-beam ptychography of a known reference sample²⁷ prior to the nanowire
146 measurements. Scanning probe fluorescence measurements were used to locate an individual
147 nanowire with the long axis aligned vertically. The vertical nanowire orientation enabled
148 two different Bragg peaks ($01\bar{1}0$ and $2\bar{1}\bar{1}0$) to be accessed in the horizontal scattering plane,
149 each sensitive to a different structural component of the nanowire. Figure 1(b-d) shows
150 depictions of the scattering geometries used to reach the Bragg peaks measured here, as well
151 as their reciprocal space orientation. At both Bragg conditions, 2D nanodiffraction maps
152 were measured at a series of angles about the Bragg peak while simultaneously measuring
153 Ga K-edge fluorescence.

154 Positional scans were done in a fly-scan mode, moving the sample with motors oriented
155 parallel to the Si membrane surface (x_{mot} , y_{mot} in Figure 1(c)), and the angle was adjusted
156 in 0.02° steps about the Bragg condition with a rotational stage (θ_{mot}) that rotated the
157 nanowire along its long axes. Fly scans, now being increasingly utilized for ptychography

158 measurements,³⁵ were implemented with an average dwell time per scan point of 0.2 seconds
 159 in order to minimize scan time overhead and eliminate motor settling time. A Merlin pixel
 160 array detector was used with 512×512 square pixels with $55\mu\text{m}$ edges and a sample-to-
 161 detector distance of 500 mm and 330 mm for the $2\bar{1}\bar{1}0$ and $01\bar{1}0$ Bragg peak measurements
 162 respectively. These peaks were found at θ motor positions of $\theta_{Br}^{2\bar{1}\bar{1}0} = -9.52^\circ$ and $\theta_{Br}^{01\bar{1}0} =$
 163 -73.15° , with the detector positioned 33.7° and 19.04° off the direct beam respectively, as
 164 shown in Figure 1. At each angle, about both Bragg peaks, overview nanodiffraction raster
 165 maps measured with coarse step sizes (~ 100 nm) were performed of the entire wire, which
 166 was used to correct for error from uncertainty in the center of rotation of the θ motor.
 167 Then fine-stepped raster scans (step size ~ 25 nm) were used for Bragg ptychography data
 168 in specific regions of the wire. We note that we did not attempt to register probe scan
 169 positions as a function of angle, as this would be impractically difficult for a 50 nm beam.
 170 This emphasizes the need for the new maBPP approach, which allows for incommensurate
 171 positions to be incorporated into the phase retrieval.

172 The two Bragg peak measurements in this study were chosen to image different types
 173 of lattice structure in the InGaAs nanowires via the sensitivity of the Bragg structure fac-
 174 tor. As illustrated in Figure 1(d), the family of $01\bar{1}0$ Bragg peaks originate only from the
 175 hexagonal WZ phase (this peak is forbidden in the cubic ZB structure). As has been de-
 176 rived previously,^{20,43} the structure factor of a WZ $01\bar{1}0$ -type peak changes by $\pm 2\pi/3$ across
 177 a $\langle 0001 \rangle$ c -axis stacking fault. In addition to the spatial variations in structure factor from
 178 WZ stacking faults, any overall distortions of the crystal due to elastic strain, dislocations,
 179 etc. will also be encoded in the structure factor, and correspondingly in the phase of $01\bar{1}0$
 180 Bragg ptychography reconstruction. The second Bragg peak belongs to the $(2\bar{1}\bar{1}0)$ family
 181 of WZ Bragg peaks which is not sensitive to WZ stacking faults, and is indistinguishable
 182 from the cubic ZB $(20\bar{2})$ type peaks. As a result, images derived from a $2\bar{1}\bar{1}0$ Bragg peak
 183 will reveal more subtle structural perturbations such as those due to elastic strain fields. In
 184 the remainder of the paper, we examine the qualitative differences between Bragg scattering

185 patterns measured at the $2\bar{1}\bar{1}0$ and $01\bar{1}0$ Bragg peaks, show an analysis of $2\bar{1}\bar{1}0$ diffraction
 186 that reveals micron-scale structure in the NW, and conclude by discussing 3D images of
 187 nanoscale strain fields and stacking order obtained from maBPP reconstructions of both
 188 Bragg conditions.

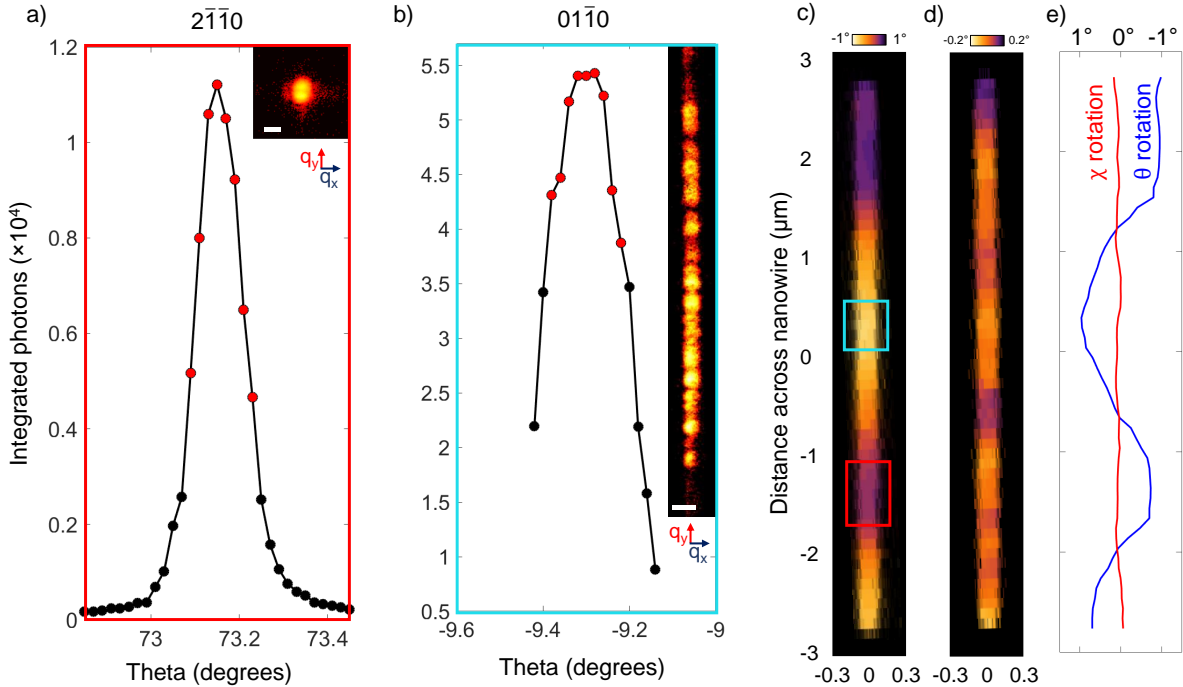


Figure 2: Sample rocking curves measured for the $2\bar{1}\bar{1}0$ (a) and $01\bar{1}0$ (b) peaks were taken taken from the center of red and cyan boxes in (c) respectively. Data was collected at each plotted point, but 3D maBPP reconstructions were performed in these regions using only the angles marked in red. Example 2D diffraction patterns (logarithmic intensity) at the Bragg maximum are shown in an inset. The diffraction pattern insets span different distances, with scale bars of 3 nm^{-1} (a) and 50 nm^{-1} (b). 2D diffraction peak mapping obtained from the $2\bar{1}\bar{1}0$ intensity patterns reveals a relative twist (c) about the long axis (about θ) and (d) a bending in the plane of the Si substrate (about χ) as a function of position across the nanowire. (e) 1D line cuts of twisting (blue) and bending (red) through the center of the nanowire. Variations in angle for (c),(d), and (e) are relative to their Bragg maximum near 73.15 degrees.

189 The characteristics of typical scattering patterns measured at both Bragg conditions high-
 190 light their sensitivity to different local structure in the nanowire. Figure 2(a,b) shows rocking
 191 curves of the $2\bar{1}\bar{1}0$ and $01\bar{1}0$ Bragg peaks measured near the middle of the outlined regions in
 192 Figure 2(c). (The rocking curves were obtained by first registering the series of 2D overview

193 nanodiffraction maps to one another using Ga fluorescence maps. The integrated-intensity
 194 rocking curves shown were then extracted from a fixed pixel position of the aligned nanowire
 195 fluorescence maps.) The coherent nanodiffraction patterns measured at the maxima of these
 196 rocking curves are inset in Figure 2. The diffraction pattern insets span different distances
 197 in q_x , q_y : (a) 15 nm^{-1} , 15 nm^{-1} and (b) 100 nm^{-1} , 1000 nm^{-1} . The $2\bar{1}\bar{1}0$ Bragg peak is
 198 predominantly composed of a central annulus-shaped speckle,⁴⁴ and can be used to map the
 199 orientation and spacing of the $(2\bar{1}\bar{1}0_{WZ})/(20\bar{2}_{ZB})$ lattice planes. By contrast, the diffraction
 200 pattern at the $01\bar{1}0$ Bragg peak is made up of many annular speckles scattering over a broad
 201 range of q_y originating from the closely spaced stacking fault boundaries illuminated by the
 202 beam that act as an interference grating.⁴³ Any variation in the position or intensity of in-
 203 dividual annular speckles within the $01\bar{1}0$ peak encodes differences in the local arrangement
 204 and nature of stacking boundaries within the illuminated volume.

205 By extracting the angle and position of the $2\bar{1}\bar{1}0$ Bragg peak maximum from the coarse
 206 nanodiffraction maps (an analysis approach similar to previous work⁴⁵⁻⁴⁷), we find that
 207 the lattice orientation varies continuously as a function of position. Figure 2(c) shows the
 208 twisting of the nanowire about the θ axis (rotation about y_{mot} as defined in Figure 1(c)), while
 209 Figure 2(d) shows the rotation about the incident beam direction (denoted as χ rotation),
 210 which is extracted by mapping the $2\bar{1}\bar{1}0$ peak center of mass along q_y . From these maps,
 211 we found that the Bragg peak angle varied by $\pm \sim 1.0^\circ$ from the mean in θ , indicating that
 212 the nanowire lattice is twisted along its growth axis. Variations of up to $\pm \sim 0.2^\circ$ from the
 213 mean in χ also indicate a bending of the nanowire. We note that all nanowires examined
 214 showed bending and twisting of a similar magnitude that could arise either during growth
 215 or during transfer to the Si membrane substrate. Regardless, the above analysis provides a
 216 micron-scale view of the lattice structure that would be useful for monitoring, for example,
 217 strain within functioning nanowire devices, and from which one can “zoom in” to specific
 218 regions of interest with ptychography.

219 Further analysis based on the maBPP approach enables reconstruction of a higher-

220 resolution 3D image of strain, and furthermore, provides a means to invert the more com-
 221 plicated speckle patterns measured at the $01\bar{1}0$ Bragg peak into 3D real space images. The
 222 reconstructions were performed for the red and cyan regions marked in Figure 2(c) for the
 223 $2\bar{1}\bar{1}0$ and $01\bar{1}0$ Bragg peaks respectively. Different regions of the nanowire were imaged
 224 in order to avoid possible beam induced damage,⁴⁸ though later measurements reveal the
 225 nanowire was structurally robust under continuous focused x-ray probe exposure.

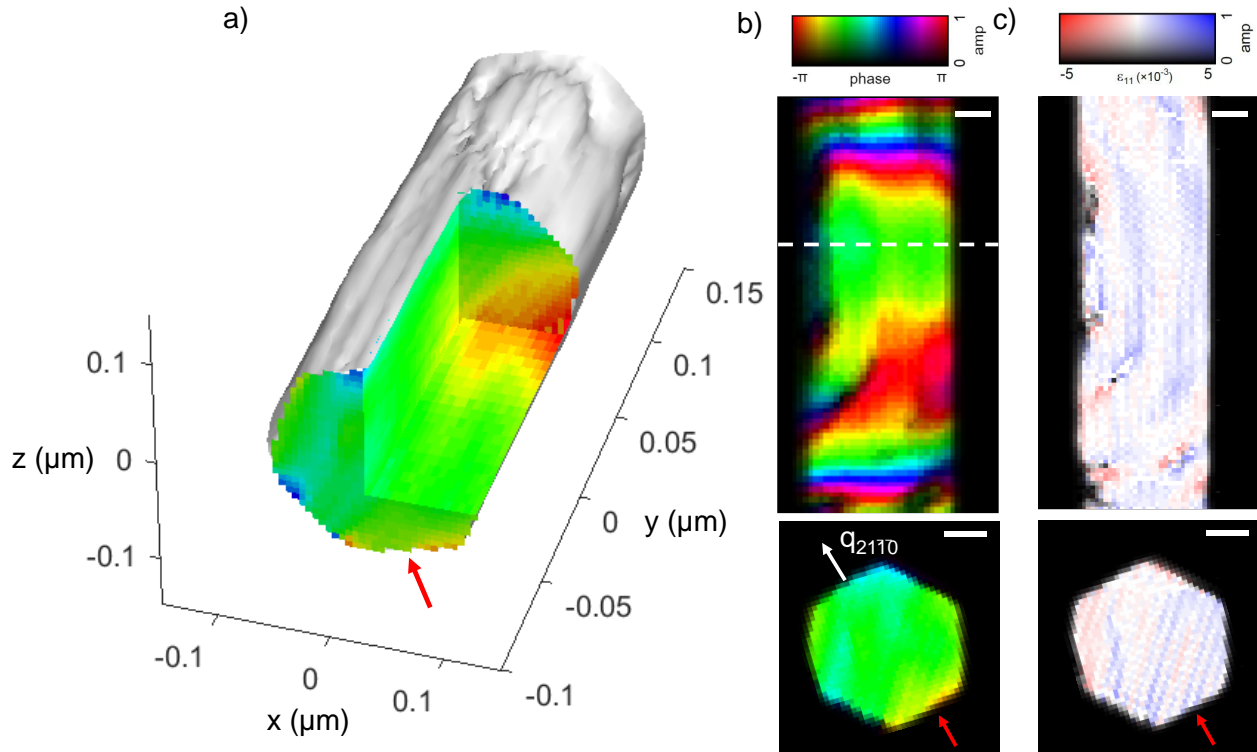


Figure 3: maBPP reconstruction of the $2\bar{1}\bar{1}0$ peak. A cut into the 3D reconstruction (a) and 2D cuts (b) taken from this volume. The cross section cut was taken from the line marked (dashed white). This reconstruction gives sensitivity to lattice displacement along $q_{2\bar{1}\bar{1}0}$ (white arrow). The same 2D cross sections converted to strain (ϵ_{11}) (c). Pixels at which the strain derivative wraps over in phase are not shown, as they are non-physical. Red arrows identify the NW facet that was adhered to the Si substrate. All scale bars are 50 nm.

226 Figure 3(a) shows a section of the nanowire (red box in Figure 2) reconstructed from
 227 the $2\bar{1}\bar{1}0$ Bragg peak nanodiffraction patterns. (Details on maBPP phase retrieval of these
 228 data are presented in the Algorithm Description section.) Because this Bragg peak is in-
 229 sensitive to stacking faults in this material, the phase of the reconstruction ($\phi_{2\bar{1}\bar{1}0}$) can be

230 related to the relative displacement of ($2\bar{1}\bar{1}0$) planes in the direction of the diffraction vec-
 231 tor ($u_{2\bar{1}\bar{1}0} = \phi_{2\bar{1}\bar{1}0}/|\mathbf{G}_{2\bar{1}\bar{1}0}|$). 2D cross sections of the displacement fields are shown in 3(b).
 232 We note that the reconstruction was performed with the Bragg condition along the white
 233 line in Figure 3(b) set as a reference. As a result, this region shows relatively flat phase
 234 due to the locally homogeneous structure that evolves axially away from the line due to
 235 the twist shown in Figure 2. These same cross sections converted to units of relative com-
 236 pressive/tensile strain along the diffraction vector are shown in 3(c), derived via the spatial
 237 derivative $\partial u_{2\bar{1}\bar{1}0}/\partial x_{2\bar{1}\bar{1}0}$,⁴⁹ where $x_{2\bar{1}\bar{1}0}$ is defined as the direction normal to the $2\bar{1}\bar{1}0$ planes.
 238 Further, analysis of the change of phase along the growth direction reveals that outside a
 239 length window of ~ 60 nm the lattice orientation varies appreciably (>10 % change) by the
 240 twist observed in Figure 2. However, we find that this twist does not strongly influence
 241 the strain component $\partial u_{2\bar{1}\bar{1}0}/\partial x_{2\bar{1}\bar{1}0}$, as evidenced by the fact that the strain field across the
 242 entire 600 nm window in Figure 3(c) varies by less than $\pm 3 \times 10^{-4}$, the $1\text{-}\sigma$ of the Gaussian
 243 distribution of strain values in the volume near the dashed line in Figure 3b. We take this
 244 value to be the strain sensitivity limit of this particular measurement, and we note that
 245 the striations in strain that fall within this range in Figure 3(c) are artifacts arising from
 246 uncertainty in the incident angle of the beam (See Supporting Information (SI) Figure S1).
 247 Further, the breadth of strain variations is comparable to strain variations expected from
 248 random alloy fluctuations assuming a binomial distribution of group III elements on group
 249 III sites (see SI Figure S2). Therefore, we do not expect significant perturbations of the
 250 band structure from any long-range strain variations present in these nanowires.

251 Finally, we note that an isotropic spatial resolution of ~ 50 nm in x , y , and z was estimated
 252 for this image, commensurate with the 53 nm full-width-at-half-maximum of the amplitude
 253 of the probe. In the x and z directions, this was done by fitting the amplitude of the facet
 254 edges to an error function. In the y direction, since no sharp features were present in the
 255 field of view, the estimate is based on the angular extent of the diffraction patterns, which
 256 does not exceed the annulus given by the beam size.

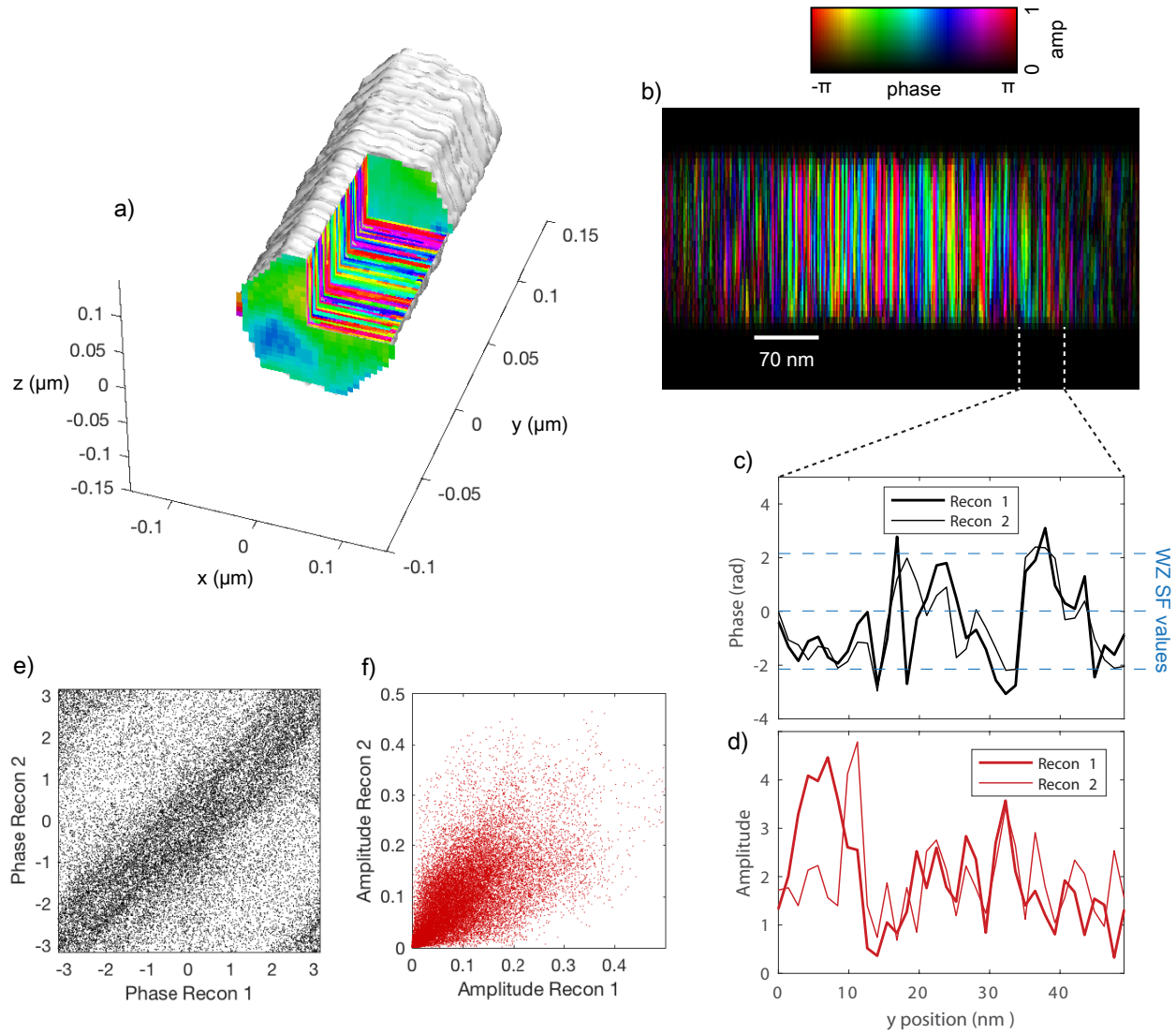


Figure 4: maBPP reconstruction of the $01\bar{1}0$ peak. A cut into the reconstruction volume (a) and a 2D slice (b) reveal rapidly varying phase features. A line cut of phase (c) and intensity (d) from the center of the nanowire compares two independent reconstructions with different starting guesses to test reproducibility. Correlation tables for every point in the two independent reconstructions show strong phase correlation. Note that the wrapping in phase around 2π results in a concentration of points at the top left and bottom right corners which should fall along the correlation axis (e) The two reconstructions show a lesser degree of amplitude correlation (f). The red arrow identifies the NW facet which was adhered to the Si substrate.

257 In the analysis discussed thus far, nanodiffraction mapping and maBPP have been used
 258 to map lattice variations and strain across length scales from a few microns to a few tens
 259 of nanometers, but shorter range structure variations in the nanowire can be accessed that
 260 have a direct impact on electrical properties. Figure 4(a) shows the 3D reconstruction of the
 261 $01\bar{1}0$ Bragg peak that is sensitive to lattice stacking order and strain in the nanowire. The
 262 maBPP image contains closely spaced regions of alternating phase and amplitude separated
 263 by planar boundaries normal to the growth direction. This morphology is consistent with
 264 TEM observations of stacking faults and phase boundaries in closely related nanowire sys-
 265 tems.³⁸ A 2D cut of the phase and amplitude variations along the growth direction is shown
 266 in 4(b). The phase color oscillations across stacking fault boundaries (shown as a line cut in
 267 Figure 4(c)) correspond roughly with the $[-2\pi/3, 0, 2\pi/3]$ phase shifts expected in the $01\bar{1}0$
 268 Bragg peak structure factor.⁴³ The $01\bar{1}0$ structure factor is sensitive not only to stacking
 269 disorder and crystal phase, but also to changes in lattice orientation and strain. Thus, in
 270 this nanowire additional variations in phase beyond those associated with stacking disorder
 271 are expected due to the substantial twists in lattice orientation.

272 The structural information in the $01\bar{1}0$ reconstruction includes multiple components,
 273 contains very high spatial frequency information, and thus requires careful consideration.
 274 As shown in Figure 2, a typical $01\bar{1}0$ coherent nanodiffraction pattern scatters to very high
 275 q_y . Such broad “barcode” interference patterns from stacking faults in nanowires have been
 276 observed previously with unfocused coherent beams, and offer the possibility of very high
 277 spatial resolution because of scattering to high q_y . In this work, photons were detected
 278 to $q_y = 0.48 \text{ \AA}^{-1}$, corresponding to an image pixel size in the y direction of 1.3 nm. (A
 279 pixel size of 6.5 nm was used in x and z due to the much more limited extent of scattering
 280 observed along q_x and q_z .) However, to date, efforts to invert such diffraction patterns to
 281 form an image via standard Bragg coherent diffraction phase retrieval methods have failed
 282 due to issues of uniqueness (multiple reconstructions initialized with random numbers yielded
 283 different local structures).^{20,43} Here, we address this issue in two ways. First, we utilize a

284 nano-focused beam such that only a few tens of stacking fault boundaries are illuminated
285 per exposure rather than several thousand, as done in previous studies. Second, we use a
286 ptychography approach that more strictly constrains the solution due to the overlap of the
287 beam positions. Both of these factors help to enable reproducible image reconstructions of
288 stacking faults via maBPP (see Figure 4(c-f)).

289 However, in this particular NW, stacking defects can only be reliably characterized over a
290 limited distance along the growth direction (<50 nm) because of the lattice twist/bend that
291 evolves over the length. Figure 2(c) shows that the Bragg peak maximum (θ_{Br}) changes with
292 position. In maBPP, these variations of θ_{Br} from the prescribed reference angle result in
293 additional phase change in the reconstruction. Figure 3(b) demonstrates how the long-range
294 twist modifies the phase for a given reconstruction of the $2\bar{1}\bar{1}0$ condition. The $01\bar{1}0$ Bragg
295 peak is sensitive to a different component of the same displacement field shown in 3(b).
296 Therefore a phase gradient is present in the resulting reconstruction (Figure 4) in addition
297 to the phase variations associated with WZ stacking faults. Thus, interpreting local phases
298 in terms of stacking faults can only be done over length scales for which phase contributions
299 from other structural phenomena are relatively constant (*e.g* lattice orientation gradients,
300 strain), which is ~ 55 nm for the nanowire shown here.

301 The reconstruction shown in Figure 4(b) demonstrates extraction of nanoscale structure
302 in the presence of these additional contributions. The Bragg condition in this reconstruction
303 was set to correspond to the rocking curve maximum in the region of the nanowire near the
304 white dashed vertical lines. The left half of the image therefore contains rapid pixel-to-pixel
305 phase oscillations due to the superposition of phase contributions from lattice twist, stacking
306 defects, as well as variations due to noise contributions,⁵⁰ making direct image interpreta-
307 tion difficult. In envisioning an *in-operando* maBPP study on SF characteristics in such
308 a nanowire, more advanced analytical tools are needed that can decouple the components
309 of lattice strain from stacking defects using multiple maBPP images of the same volume,
310 enabling larger fields of view to be interrogated.

311 Nevertheless, within a 50 nm field of view along the wire axis, as shown, these com-
 312 plicating factors are minimized, and several stacking fault boundaries can be reproducibly
 313 imaged. To demonstrate this, phase and amplitude from two different randomly initiated
 314 reconstructions (Recon 1,2) are compared for the region denoted in Figure 4(b). Line-outs
 315 from this region (4(c,d)) reproduce well, and a strong correlation is seen for all voxels in the
 316 volume bounded by planes parallel to the dotted lines (4(e,f)). Within this field of view we
 317 can identify ~ 10 WZ stacking fault boundaries that result in $[-2\pi/3, 0, 2\pi/3]$ phase values.
 318 The amplitude in this reconstruction is sensitive to ZB phase, but because the ZB inclusions
 319 are expected to persist over very small distances (< 1 nm), they will be under-resolved in
 320 this image. Given the observation of realistic features expected for these nanowires³⁸ on the
 321 scale as small as 2 pixels, we conservatively estimate an upper bound resolution along the
 322 wire axis of 2.6 nm ($2 \times$ pixel size). (SI Figure S3 shows reconstructions from simulations
 323 of a lower stacking fault density nanowire in which this spatial resolution estimate is more
 324 clearly demonstrated.) We note that many if not most III-As nanowires can be grown with
 325 a much lower density of stacking defects than the nanowire imaged here, suggesting that
 326 the maBPP methodology can be usefully applied to correlate defect density and electronic
 327 properties in many nanowire systems of interest. Finally, as in the $2\bar{1}\bar{1}0$ reconstruction, the
 328 average resolution along the x and z directions was found to be ~ 50 nm consistent with
 329 the limited angular extent of scattering along q_x and q_z . As is in any ptychography exper-
 330 iments, improvements in resolution can be obtained with improved signal-to-noise ratios of
 331 the diffraction signal, especially in regions that extend beyond the beam-limited annulus in
 332 reciprocal space.

333 In conclusion, we demonstrated the ability to image a single InGaAs nanowire on many
 334 length scales with sensitivity to multiple nanoscale lattice features. In analyzing and recon-
 335 structing diffraction patterns from the $2\bar{1}\bar{1}0$ Bragg peak, we found that the lattice orientation
 336 varied along the length of the wire at micron length scales and that the strain field along the
 337 wire cross-section was largely unaffected by this long range lattice rotation. Using diffraction

338 patterns measured from the same wire at the $01\bar{1}0$ Bragg peak, we could reproducibly re-
 339 construct images of stacking defects. This reconstruction evidenced sharp planar boundaries
 340 between different crystal phases of WZ structure, as expected. In both cases, phase retrieval
 341 was made possible by a multi-angle Bragg projection ptychography approach that accom-
 342 modates coherent nanodiffraction patterns measured at arbitrary overlapping positions at
 343 multiple angles about a Bragg peak, eliminating the need for scan registration at different
 344 angles which is impractical with nanobeams. In combination with coarse scanning nan-
 345 odiffraction measurements, maBPP allowed for structural investigation of a nanowire over
 346 three decades of length spanning from several microns to tens of angstroms. By enabling
 347 such a capability, maBPP can contribute significantly to our understanding of nanowires
 348 and other nanostructures by correlating structure and properties. This capability will be
 349 especially complementary to electron microscopy of nanowires on transparent supports and
 350 post-operando atom probe tomography of nanowires embedded in devices, and improvements
 351 in maBPP spatial resolution and strain sensitivity can be achieved with further development
 352 of the method, for example, by implementing simultaneous probe and sample reconstruction.

353 **Algorithm Description:** maBPP is predicated on a description of coherent scattering
 354 from a nanoscale crystal that equates the far-field diffracted intensity pattern measured with
 355 an area detector to a general probe position and measurement angle relative to the Bragg
 356 peak. These two degrees of freedom, position and angle, are illustrated in Figure 1(b,c). A
 357 monochromatic beam illuminating a crystal will satisfy a Bragg condition when the scattering
 358 vector $\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$ coincides with a Bravais lattice point \mathbf{G}_{HKL} of the illuminated crystal.
 359 (Here, $|\mathbf{k}| = 2\pi/\lambda$ where λ is the x-ray wavelength.) Small angular deviations from this
 360 condition can be expressed in terms of $\mathbf{Q} = \mathbf{q} - \mathbf{G}$. As described in other work, this
 361 vector \mathbf{Q} encodes changes in a coherent diffraction pattern due to angular variations along a
 362 Bragg rocking curve.^{51,52} A focused-beam nanodiffraction experiment also allows the incident
 363 beam to scan a given region of interest in a crystal by scanning the probe position relative
 364 to the sample (in this case, using sample stage motors x_{mot}, y_{mot}). Thus, a general Bragg

365 ptychography data set for a given field of view comprises of $j = 1 \cdots J$ two-dimensional
 366 coherent diffraction intensity patterns I_j measured as a function of different probe positions
 367 (\mathbf{r}_j) at various angles relative to the Bragg peak (θ_j).

368 Each of these intensity patterns is the squared modulus of the diffracted wave field at the
 369 detector, $I_j = |\psi_j|^2$. The quantity ψ_j can be generally expressed in a maBPP experiment as:

$$\psi_j = \mathcal{F}\mathcal{R}\mathcal{Q}_{\theta_j}P_{\mathbf{r}_j}\rho. \quad (1)$$

370 Here, $P_{\mathbf{r}_j}$ is the 3D wave field of the focused x-ray probe positioned to illuminate the crystal
 371 ρ according to the translation of the sample stage motors (x_{mot}, y_{mot}). The term $\mathcal{Q}_{\theta_j} =$
 372 $\exp[i\mathbf{r} \cdot \mathbf{Q}_{\theta_j}]$ (where $i = \sqrt{-1}$) is a 3D real-space complex-valued phase term that encodes
 373 spatial frequencies corresponding to angular deviations from θ_{Br} , where θ_{Br} is the angle that
 374 satisfies the Bragg condition of the crystal. \mathcal{R} is a 3D \rightarrow 2D projection along the \mathbf{k}_f direction,
 375 and \mathcal{F} is a 2D Fourier transformation. This construction of ψ_j leads to a general description
 376 of a Bragg ptychography data set in which the probe position and angle are arbitrary and
 377 need not be otherwise related so long as the typical degree of probe overlap ($\sim 50\%$) is
 378 enforced. In a manner similar to References 24 and 51, Equation 1 can be used to derive
 379 a gradient that minimizes the sum squared error $\epsilon^2 = \sum_j \| |\psi_j| - \sqrt{I_j} \|^2$ and that can
 380 be incorporated into phase retrieval algorithms such as the Ptychographic Iterative Engine
 381 (PIE) to reconstruct a 3D image, as was done in this work.

382 For the maBPP data sets measured at the two Bragg peaks featured in this work, diffrac-
 383 tion maps from only strongly scattering angles (indicated in red in Figure 2(a) and (b)) were
 384 used for image reconstruction. 25 iterations of maBPP with PIE were performed, and a
 385 hexagonal-shaped 3D support was used corresponding to the facet orientation of the SEM
 386 image in Figure 1(a). The diameter of the support for the $2\bar{1}\bar{1}0$ and $01\bar{1}0$ reconstructions
 387 was, respectively, 180% and 130% of the nominal wire diameter.

388 **Associated Content**

389 Supporting Information Available: Document describing 1) the effect of angular uncertainty
390 on a maBPP experiment, 2) simulation of strain induced by composition variations due
391 to random alloying, and 3) a maBPP reconstruction of a simulated nanowire with random
392 stacking faults as compared to the original structure.

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414 References

- 415 (1) Koblmüller, G.; Abstreiter, G. *Phys. Status Solidi RRL* **2014**, *8*, 11–30. DOI:
416 10.1002/pssr.201308207.
- 417 (2) Kim, H.; Lee, W.-J.; Farrell, A. C.; Morales, J. S.; Senanayake, P. N.;
418 Prikhodko, S. V.; Ochalski, T.; Huffaker, D. L. *Nano Lett.* **2017**, *17*, 3465–3470. DOI:
419 10.1021/acs.nanolett.7b00384.
- 420 (3) Shen, L.; Pun, E. Y.; Ho, J. C. *Mater. Chem. Front.* **2017**, *1*, 630–645. DOI:
421 10.1039/C6QM00279J.
- 422 (4) Li, N.; Liu, K.; Sorger, V. J.; Sadana, D. K. *Sci. Rep.* **2015**, *5*, 14067. DOI:
423 10.1038/srep14067.
- 424 (5) Mårtensson, T.; Svensson, C. P. T.; Wacaser, B. A.; Larsson, M. W.; Seifert, W.;
425 Deppert, K.; Gustafsson, A.; Wallenberg, L. R.; Samuelson, L. *Nano Lett.* **2004**, *4*,
426 1987–1990. DOI: 10.1021/nl0487267.
- 427 (6) Koblmüller, G.; Mayer, B.; Stettner, T.; Abstreiter, G.; Finley, J. J. *Semicond. Sci.*
428 *Tech.* **2017**, *32*, 053001. DOI: 10.1088/1361-6641/aa5e45.
- 429 (7) LaPierre, R.; Robson, M.; Azizur-Rahman, K.; Kuyanov, P. *J. Phys. D Appl. Phys.*
430 **2017**, *50*, 123001. DOI: 10.1088/1361-6463/aa5ab3.
- 431 (8) Spirkoska, D.; Arbiol, J.; Gustafsson, A.; Conesa-Boj, S.; Glas, F.; Zardo, I.;
432 Heigoldt, M.; Gass, M.; Bleloch, A. L.; Estrade, S. *Phys. Rev. B* **2009**, *80*, 245325.
433 DOI: 10.1103/PhysRevB.80.245325.

- 434 (9) Verheijen, M. A.; Algra, R. E.; Borgström, M. T.; Immink, G.; Sourty, E.; van
435 Enckevort, W. J.; Vlieg, E.; Bakkers, E. P. *Nano Lett.* **2007**, *7*, 3051–3055. DOI:
436 10.1021/nl071541q.
- 437 (10) Rudolph, D.; Funk, D., Stefan; Markus; Morkötter, S.; Hertenberger, S.; Schweick-
438 ert, L.; Becker, J.; Matich, S.; Bichler, M.; Spirkoska, D. *Nano Lett.* **2013**, *13*, 1522–
439 1527. DOI: 10.1021/nl3046816.
- 440 (11) Jeon, N.; Loitsch, B.; Morkötter, S.; Abstreiter, G.; Finley, J.; Krenner, H. J.;
441 Koblmüller, G.; Lauhon, L. J. *ACS Nano* **2015**, *9*, 8335–8343. DOI: 10.1021/ac-
442 snano.5b04070.
- 443 (12) Schroer, M. D.; Petta, J. R. *Nano Lett.* **2010**, *10*, 1618–1622. DOI: 10.1021/nl904053j.
- 444 (13) Irber, D. M.; Seidl, J.; Carrad, D. J.; Becker, J.; Jeon, N.; Loitsch, B.; Winnerl, J.;
445 Matich, S.; Dblinger, M.; Tang, Y.; Morkötter, S.; Abstreiter, G.; Finley, J. J.;
446 Grayson, M.; Lauhon, L. J.; Koblmüller, G. *Nano Lett.* **2017**, *17*, 4886–4893. DOI:
447 10.1021/acs.nanolett.7b01732.
- 448 (14) Jahn, U.; Lähnemann, J.; Pfüller, C.; Brandt, O.; Breuer, S.; Jenichen, B.; Ram-
449 steiner, M.; Geelhaar, L.; Riechert, H. *Phys. Rev. B* **2012**, *85*, 045323. DOI:
450 10.1103/PhysRevB.85.045323.
- 451 (15) Loitsch, B.; Winnerl, J.; Grimaldi, G.; Wierzbowski, J.; Rudolph, D.; Morkötter, S.;
452 Doblinger, M.; Abstreiter, G.; Koblmüller, G.; Finley, J. J. *Nano letters* **2015**, *15*,
453 7544–7551. DOI:10.1021/acs.nanolett.5b03273.
- 454 (16) Dzhigaev, D.; Stankevic, T.; Bi, Z.; Lazarev, S.; Rose, M.; Shabalina, A.; Reinhardt, J.;
455 Mikkelsen, A.; Samuelson, L.; Falkenberg, G. *ACS Nano* **2017**, *11*, 6605–6611. DOI:
456 10.1021/acs.nano.6b08122.

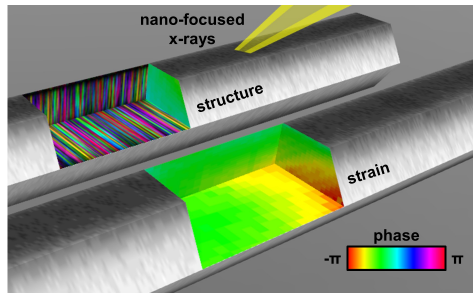
- 457 (17) Davtyan, A.; Krause, T.; Kriegner, D.; Al-Hassan, A.; Bahrami, D.;
458 Mostafavi Kashani, S.; Lewis, R. B.; Küpers, H.; Tahraoui, A.; Geelhaar, L. *J.*
459 *Appl. Crystallogr.* **2017**, *50*, 673–680. DOI: 10.1107/S1600576717004149.
- 460 (18) Diaz, A.; Mocuta, C.; Stangl, J.; Mandl, B.; David, C.; Vila-Comamala, J.;
461 Chamard, V.; Metzger, T.; Bauer, G. *Phys. Rev. B* **2009**, *79*, 125324. DOI:
462 10.1103/PhysRevB.79.125324.
- 463 (19) Labat, S.; Richard, M.-I.; Dupraz, M.; Gailhanou, M.; Beutier, G.; Verdier, M.; Mas-
464 tropietro, F.; Cornelius, T. W.; Schüllli, T. U.; Eymery, J.; Thomas, O. *ACS Nano*
465 **2015**, *9*, 9210–9216. DOI: 10.1021/acs.nano.5b03857.
- 466 (20) Davtyan, A.; Biermanns, A.; Loffeld, O.; Pietsch, U. *New J. Phys.* **2016**, *18*, 063021.
467 DOI: 10.1088/1367–2630/18/6/063021.
- 468 (21) Davtyan, A.; Lehmann, S.; Kriegner, D.; Zamani, R. R.; Dick, K. A.; Bahrami, D.;
469 Al-Hassan, A.; Leake, S. J.; Pietsch, U.; Holỳ, V. *J. Synchrotron Radiat.* **2017**, *24*,
470 981–990. DOI: 10.1107/S1600577517009584.
- 471 (22) Treu, J.; Speckbacher, M.; Saller, K.; Morkötter, S.; Döblinger, M.; Xu, X.; Riedl, H.;
472 Abstreiter, G.; Finley, J.; Koblmüller, G. *Appl. Phys. Lett.* **2016**, *108*, 053110. DOI:
473 10.1063/1.4941407.
- 474 (23) Treu, J.; Stettner, T.; Watzinger, M.; Morkötter, S.; Doeblinger, M.; Matich, S.;
475 Saller, K.; Bichler, M.; Abstreiter, G.; Finley, J. J.; Stangl, J.; Koblmüller, G. *Nano*
476 *Lett.* **2015**, *15*, 3533–3540. DOI: 10.1021/acs.nanolett.5b00979.
- 477 (24) Hruszkewycz, S. O.; Allain, M.; Holt, M. V.; Murray, C. E.; Holt, J. R.; Fuoss, P. H.;
478 Chamard, V. *Nat. Mater.* **2016**, *16*, 244–251. DOI: 10.1038/nmat4798.
- 479 (25) Godard, P.; Carbone, G.; Allain, M.; Mastropietro, F.; Chen, G.; Capello, L.;

- 480 Diaz, A.; Metzger, T. H.; Stangl, J.; Chamard, V. *Nat. Commun.* **2011**, *2*, 568. DOI:
481 10.1038/ncomms1569.
- 482 (26) Rodenburg, J. M.; Hurst, A. C.; Cullis, A. G.; Dobson, B. R.; Pfeiffer, F.; Bunk, O.;
483 David, C.; Jefimovs, K.; Johnson, I. *Phys. Rev. Lett.* **2007**, *98*, 034801. DOI:
484 10.1103/PhysRevLett.98.034801.
- 485 (27) Thibault, P.; Dierolf, M.; Menzel, A.; Bunk, O.; David, C.; Pfeiffer, F. *Science* **2008**,
486 *321*, 379–382. DOI: 10.1126/science.1158573.
- 487 (28) Godard, P.; Allain, M.; Chamard, V.; Rodenburg, J. *Opt. Express* **2012**, *20*, 25914–
488 25934. DOI: 10.1126/science.1158573.
- 489 (29) Holt, M. V.; Hruszkewycz, S. O.; Murray, C. E.; Holt, J. R.; Paskiewicz, D. M.; Fu-
490 oss, P. H. *Phys. Rev. Lett.* **2014**, *112*, 165502. DOI: 10.1103/PhysRevLett.112.165502.
- 491 (30) Ulvestad, A.; Clark, J. N.; Harder, R.; Robinson, I. K.; Shpyrko, O. G. *Nano Lett.*
492 **2015**, *15*, 4066–4070, DOI: 10.1021/acs.nanolett.5b01104.
- 493 (31) Munkholm, A.; Thompson, C.; Foster, C. M.; Eastman, J. A.; Auciello, O.; Stephen-
494 son, G. B.; Fini, P.; DenBaars, S. P.; Speck, J. S. *Appl. Phys. Lett.* **1998**, *72*, 2972–4.
495 DOI: 10.1063/1.121511.
- 496 (32) Takahashi, Y.; Suzuki, A.; Furutaku, S.; Yamauchi, K.; Kohmura, Y.; Ishikawa, T.
497 *Phys. Rev. B* **2013**, *87*, 121201. DOI: 10.1103/PhysRevB.87.121201.
- 498 (33) Hruszkewycz, S. O.; Highland, M. J.; Holt, M. V.; Kim, D.; Folkman, C. M.; Thomp-
499 son, C.; Tripathi, A.; Stephenson, G. B.; Hong, S.; Fuoss, P. H. *Phys. Rev. Lett.* **2013**,
500 *110*, 177601. DOI: 10.1103/PhysRevLett.110.177601.
- 501 (34) Pateras, A. I.; Allain, M.; Godard, P.; Largeau, L.; Patriarche, G.; Talneau, A.;
502 Pantzas, K.; Burghammer, M.; Minkevich, A. A.; Chamard, V. *Phys. Rev. B* **2015**, *92*,
503 205305–11. DOI: 10.1103/PhysRevB.92.205305.

- 504 (35) Pelz, P. M.; Guizar-Sicairos, M.; Thibault, P.; Johnson, I.; Holler, M.; Menzel, A. *Appl.*
505 *Phys. Lett.* **2014**, *105*, 251101. DOI: 10.1063/1.4904943.
- 506 (36) Clark, J. N.; Huang, X.; Harder, R. J.; Robinson, I. K. *Opt. Lett.* **2014**, *39*, 6066–6069.
507 DOI: 10.1364/OL.39.006066P.
- 508 (37) Hruszkewycz, S. O.; Holt, M. V.; Murray, C. E.; Bruley, J.; Holt, J.; Tripathi, A.;
509 Shpyrko, O. G.; McNulty, I.; Highland, M. J.; Fuoss, P. H. *Nano Lett.* **2012**, *12*, 5148–
510 5154. DOI: 10.1021/nl303201w.
- 511 (38) Morkötter, S.; Funk, S.; Liang, M.; Doeblinger, M.; Hertenberger, S.; Treu, J.;
512 Rudolph, D.; Yadav, A.; Becker, J.; Bichler, M.; Scarpa, G.; Lugli, P.; Zardo, I.;
513 Finley, J. J.; Abstreiter, G.; Koblmüller, G. *Phys. Rev. B* **2013**, *87*, 205303. DOI:
514 10.1103/PhysRevB.87.205303.
- 515 (39) Saxena, D.; Mokkaḡpati, S.; Parkinson, P.; Jiang, N.; Gao, Q.; Tan, H. H.; Jagadish, C.
516 *Nat. Photonics* **2013**, *7*, 963–968 DOI: 10.1038/nphoton.2013.303.
- 517 (40) Yan, H.; Nazaretski, E.; Lauer, K.; Huang, X.; Wagner, U.; Rau, C.; Yusuf, M.; Robin-
518 son, I.; Kalbfleisch, S.; Li, L. *Sci. Rep.* **2016**, *6*, 20112. DOI: 10.1038/srep20112.
- 519 (41) Nazaretski, E.; Lauer, K.; Yan, H.; Bouet, N.; Zhou, J.; Conley, R.; Huang, X.; Xu, W.;
520 Lu, M.; Gofron, K.; Kalbfleisch, S.; Wagner, U.; Rau, C.; Chu, Y. S. *J. Synchrotron*
521 *Radiat.* **2015**, *22*, 1–6. DOI: 10.1107/S1600577514025715.
- 522 (42) Nazaretski, E.; Yan, H.; Lauer, K.; Bouet, N.; Huang, X.; Xu, W.; Zhou, J.;
523 Shu, D.; Hwu, Y.; Chu, Y. S. *J. Synchrotron Radiat.* **2017**, *24*, 1113–1119.
524 DOI:10.1107/S1600577517011183.
- 525 (43) Favre-Nicolin, V.; Mastropietro, F.; Eymery, J.; Camacho, D.; Niquet, Y. M.;
526 Borg, B. M.; Messing, M. E.; Wernersson, L.-E.; Algra, R. E.; Bakkers, E. P.

- 527 A. M.; Metzger, T. H.; Harder, R.; Robinson, I. K. *New J. Phys.* **2010**, *12*, 035013.
528 DOI:10.1088/1367-2630/12/3/035013.
- 529 (44) Hruszkewycz, S. O.; Holt, M. V.; Allain, M.; Chamard, V.; Polvino, S. M.; Mur-
530 ray, C. E.; Fuoss, P. H. *Opt. Lett.* **2015**, *40*, 3241. DOI: 10.1364/OL.40.003241.
- 531 (45) Stankevič, T.; Hilner, E.; Seiboth, F.; Ciechonski, R.; Vescovi, G.; Kryliouk, O.; Jo-
532 hansson, U.; Samuelson, L.; Wellenreuther, G.; Falkenberg, G. *ACS Nano* **2015**, *9*,
533 6978–6984. DOI: 10.1021/acs.nano.5b01291.
- 534 (46) Dzhigaev, D.; Shabalin, A.; Stankevič, T.; Lorenz, U.; Kurta, R.; Seiboth, F.; Wal-
535 lentin, J.; Singer, A.; Lazarev, S.; Yefanov, O.; , M. N. S. L. S. G. F. C. G. S. A. M.
536 R. F., M Borgström; Vartanyants, I. A. *J. Opt.* **2016**, *18*, 064007. DOI: 10.1088/2040-
537 8978/18/6/064007.
- 538 (47) Wallentin, J.; Jacobsson, D.; Osterhoff, M.; Borgström, M. T.; Salditt, T. *Nano Lett.*
539 **2017**, *17*, 4143–4150. DOI: 10.1021/acs.nanolett.7b00918.
- 540 (48) Wallander, H.; Wallentin, J. *J. Synchrotron Radiat.* **2017**, *24*, 925–933. DOI:
541 10.1107/S1600577517008712.
- 542 (49) Ulvestad, A.; Singer, A.; Clark, J. N.; Cho, H. M.; Kim, J. W.; Harder, R.; Maser, J.;
543 Meng, Y. S.; Shpyrko, O. G. *Science* **2015**, *348*, 1344–1347. DOI: 10.1126/sci-
544 ence.aaa1313.
- 545 (50) Mastropietro, F.; Godard, P.; Burghammer, M.; Chevallard, C.; Daillant, J.; Dubois-
546 set, J.; Allain, M.; Guenoun, P.; Nouet, J.; Chamard, V. *Nat. Mater.* **2017**, *9*, 946–952.
547 DOI: 10.1038/nmat4937.
- 548 (51) Cha, W.; Ulvestad, A.; Allain, M.; Chamard, V.; Harder, R.; Leake, S. J.; Maser, J.;
549 Fuoss, P. H.; Hruszkewycz, S. O. *Phys. Rev. Lett.* **2016**, *117*, 225501–5. DOI:
550 10.1103/PhysRevLett.117.225501.

551 (52) Dzhigaev, D.; Stankevič, T.; Besedin, I.; Lazarev, S.; Shabalin, A.; Strikhanov, M. N.;
552 Feidenhans, R.; Vartanyants, I. A. *Proc. SPIE* **2015**, *9592*, 95920S–95920S. DOI:
553 10.1117/12.2190416.



TOC Figure