Stress control of tensile-strained In$_{1-x}$Ga$_x$P nanomechanical string resonators

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(Dated: 30 August 2018)

We investigate the mechanical properties of freely suspended nanostrings fabricated from tensile-stressed, crystalline In$_{1-x}$Ga$_x$P. The intrinsic strain is a consequence of the epitaxial growth given by the lattice mismatch between the thin film and the substrate which is confirmed by x-ray diffraction measurements. The flexural eigenfrequencies of the nanomechanical string resonators reveal an orientation dependent stress with a maximum value of 650 MPa. The angular dependence is explained by a combination of anisotropic Young’s modulus and a change of elastic properties caused by defects. As a function of the crystal orientation a stress variation of up to 50 % is observed. This enables fine tuning of the tensile stress for any given Ga content $x$, which implies interesting prospects for the study of high Q nanomechanical systems.

Keywords: InGaP, GaInP, nanomechanical resonator, tensile strained crystal

Introducing strain in material systems enables the control of various physical properties. Examples include the improved performance of semiconductor lasers, enhanced carrier mobility in transistors, direct formation of quantum dots and increased mechanical quality factors (Q) in micro- and nanomechanical systems (M-/NEMS). In particular, tensile-strained amorphous silicon nitride has evolved to a standard material in nanomechanics in recent years. The dissipation dilution arising from the inherent tensile pre-stress of the silicon nitride film gives rise to room temperature Q factors of several 100 000 at 10 MHz resonance frequencies while additional stress engineering has been shown to increase Q by a few orders of magnitude. However, defects set a bound on the attainable dissipation and hence Q in amorphous materials. Stress-free single crystal resonators, on the other hand, feature lower room temperature Q factors but exhibit a strong enhancement of Q when cooled down to millikelvin temperatures as a result of the high intrinsic Q of single crystal materials. Combining dissipation dilution via tensile stress with high intrinsic Q of single crystal materials could open a way to reach ultimate mechanical Q at room temperature.

In the last years a few possible candidates for tensile-strained crystalline nanomechanical resonators have emerged. Those include, for example, heterostructures of the silicon based 3C-SiC and the III-V semiconductors GaAs, GaNA, and In$_{1-x}$Ga$_x$P. Advantages of ternary In$_{1-x}$Ga$_x$P (InGaP) are the direct bandgap (for $x < 63 %$) and the broad strain tunability. When grown on GaAs wafers, this alloy system may be compressively strained, strain-free or tensile strained, with possible tensile stress values exceeding 1 GPa, by varying the group-III composition $x$. The prospects of InGaP in nanomechanics range from possible applications in cavity optomechanics to coupling with quantum-electronic systems, such as quantum wells and quantum dots.

Here we explore freely suspended nanostrings fabricated from InGaP as nanomechanical systems. Our analysis reveals that even for fixed $x$ the tensile stress state of the resonator can be controlled by varying the resonator orientation on the chip. This implies that unlike for the case of silicon nitride NEMS, resonator orientation will be an important design parameter allowing to fine-tune the tensile stress for any given Ga content $x$.

We investigate crystalline string resonators from two differently stressed, MBE grown III-V heterostructures, illustrated in Fig. (a) and (b). Both structures consist of two 86 nm thick InGaP layers, each capped by 1 nm of GaAs. Both InGaP layers are situated atop a sacrificial layer of high aluminum content Al$_y$Ga$_{1-y}$As (AlGaAs), with $y = 92 %$. Note that only the top InGaP and AlGaAs layers were employed as resonator and sacrificial layer, respectively in this work.

By varying the Ga content of InGaP, the lattice-constant $a_0^\infty (x)$ changes by up to 7 %. Since the substrate lattice constant of AlGaAs changes by only 0.1 %, as a function of its Al content, we assume the lattice constant of AlGaAs to equal that of plain GaAs $a_{GaAs}$. The difference in lattice constants result in a lattice mismatch $\delta^\infty = (a_0^\infty (x) - a_{GaAs})/a_{GaAs}$ between the InGaP and the AlGaAs lattice. This mismatch induces an in-plane strain $\varepsilon^\parallel (x)$ in the InGaP layer and is defined by the ratio.

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\[ \varepsilon^\parallel(x) = \frac{a^\parallel(x) - a^\infty(x)}{a^\infty(x)}, \quad a^\parallel = a_{\text{GaAs}} \]  

(1)

with the distorted in-plane lattice constant \( a^\parallel \) of the strained InGaP layer, which in case of a 100% pseudomorphic layer equals the lattice constant of the substrate \( a^\parallel = a_{\text{GaAs}} \). An InGaP layer grows strain-free (lattice-matched) on a GaAs substrate for \( x = 51\% \) Ga content, i.e. \( a^\parallel = a_{\text{GaAs}}(0.51) \). The layer is grown tensile (compressive) strained for a higher (lower) Ga content. With this heterostructure it is thus possible to adjust and tailor the strain in a film up to a critical thickness determined by \( x_{\text{HS}} \). In this work we investigate In\(_{1-x}\)Ga\(_x\)P with Ga contents of \( x_{\text{HS}} = 58.7\% \) (high-stress) and \( x_{\text{LS}} = 52.8\% \) (low-stress). The resulting strain values are \( \varepsilon^\parallel(x_{\text{HS}}) = 5.34 \times 10^{-3} \) and \( \varepsilon^\parallel(x_{\text{LS}}) = 0.95 \times 10^{-3} \), for InGaP on GaAs, respectively.

String resonators were defined by electron-beam-lithography followed by a SiCl\(_4\) inductively coupled plasma etch, using negative electron-beam-resist ma-N 2403 as an etch-mask, before releasing them with a buffered HF wet etch. The resonators are additionally cleaned via digital wet etching. In the end we critical-point dried the samples, to avoid stiction and destruction of the structures. Examples of free standing string resonators are shown in Fig. 1(c).

The samples are mounted inside a vacuum chamber (pressure \( < 10^{-3} \) mbar) to avoid degradation of the AlGaAs sacrificial-layer under ambient conditions as well as gas damping. We measured the fundamental resonance frequency of the out-of-plane flexural mode of resonators of different length and orientation on the substrate, using piezo-actuation and interferometric detection. Figure 2 shows measured frequencies as a function of the resonator length \( L \) for two different resonator orientations on the chip. Resonators with an angle of 0° are oriented parallel to the cleaved chip edges, see inset of Fig. 2, which correspond to the <110> crystal directions for III-V heterostructures on (001) GaAs substrate wafers. Hence, the strings point along a <110> direction. For comparison, we also discuss resonators which are rotated clockwise by 45°, and hence are oriented along a <100> direction of the crystal.

\[ f_n = \frac{n^2\pi}{2L^2} \sqrt{\frac{EI}{\rho A}} \sqrt{1 + \frac{\sigma AL^2}{n^2\pi^2EI}} \]  

\( f_n \approx \frac{n^2\pi}{2L^2} \sqrt{\frac{\sigma}{\rho}} \frac{AL^2}{n^2\pi^2EI} \gg 1, \]  

(2)

where \( E \) is Young’s modulus, \( I \) is the area moment of inertia, \( \rho \) is the mass density, \( A \) is the cross-sectional area, and \( \sigma \) the stress. This formula reduces to \( f \approx (1/2L)\sqrt{\sigma/\rho} \), for the case of high tensile stress and the fundamental mode \( (n = 1) \) considered here. The resonance frequencies shown in Fig. 2 are fitted with Eq. (2a) and clearly follow the expected \( 1/L \) dependence. Being in the high tensile stress regime, a change in frequency for a given resonator length can only originate from a different tensile stress \( \sigma \). The frequency mismatch between the 0° and 45° data indicates that the stress depends on the resonator’s orientation. Solving Eq. (2a) for \( \sigma \) and calculating the weighted mean from all data points yields \( \sigma(x_{\text{HS}}, 0^\circ) = 642.3(3.3) \text{MPa} \) and \( \sigma(x_{\text{HS}}, 45^\circ) = 440.2(2.6) \text{MPa} \), indicating that the tensile stress varies by almost 50% with crystal direction.

For anisotropic materials, stress and strain are related by the fourth rank compliance \( S \) or stiffness \( C \) tensors:

\[ \sigma = C \varepsilon \quad \text{and} \quad \varepsilon = S \sigma, \]  

Those tensors simplify to 6 \times 6 matrices with three independent components, \( c_{11}, c_{12} \) and \( c_{44} \), for the case of the cubic symmetry of e.g. the
zincblende crystal structure:

$$C(x) = \begin{pmatrix} c_{11} & c_{12} & c_{12} & 0 & 0 \\ c_{12} & c_{11} & c_{12} & 0 & 0 \\ c_{12} & c_{12} & c_{11} & 0 & 0 \\ 0 & 0 & 0 & c_{44} & 0 \\ 0 & 0 & 0 & 0 & c_{44} \end{pmatrix}. \tag{3}$$

For In$_{1-x}$Ga$_x$P, each component $c_{ij}$ depends on the Ga content $x$.

By applying matrix rotations and transformations (see supplementary material), one can calculate the angle dependent Young’s modulus $E(x, \theta)$ of an ideal and defect free system from Eq. 3. Figure 3 shows $E(x, \theta)$ for the two different Ga contents $x_{HS} = 58.7\%$ and $x_{LS} = 52.8\%$. The Young’s modulus displays a similar behavior for both Ga contents, and varies between 80 GPa and 125 GPa, between the $<100>$ and $<110>$ crystal directions, respectively. In addition, Fig. 3 clearly reveals the 90° rotation symmetry of $E(x, \theta)$. To calculate the tensile stress we multiply the Young’s modulus by the strain from Eq. 1 according to Hooke’s law:

$$\sigma(x, \theta) = E(x, \theta)\varepsilon(x, \theta). \tag{4}$$

The resulting stress values for both angles, $\sigma(x_{HS}, 0^\circ) = 655.3$ MPa and $\sigma(x_{HS}, 45^\circ) = 454.9$ MPa, coincide well with the experimental results.

![FIG. 3. Crystal orientation in wafers and angle dependent Young’s modulus in In$_{1-x}$Ga$_x$P. (a) Schematic crystal orientations of a (001) GaAs wafer. In this case, unrotated (0°) resonators point along a $<110>$ crystal direction. The resonator angles are changed clockwise, e.g. from [110] towards [001]. Inset: Definition of the resonator angle such that 0° resonators are parallel to the chip edge along a $<110>$ direction. (b) Orientation dependent Young’s modulus inside the (001) wafer plane, showing a 90° rotation symmetry. Solid line for $x_{HS} = 58.7\%$ and dashed line for $x_{LS} = 52.8\%$. (c) Close-up of angle dependent Young’s modulus, showing the first quadrant of the polar plot (b).](image)

To further investigate the angular stress dependence of InGaP, we fabricated the same resonator arrays with angles changing in $\Delta\theta = 11.25^\circ$ steps. For each orientation the tensile stress is extracted using the procedure described in Fig. 2. The top plots of Fig. 4 show the resulting angular stress dependence for two different Ga contents. In both cases, local stress maxima are observed at 0° and 90°, i.e. along $<110>$ crystal directions. Accordingly, the minima are found at 45° and 135°, which correspond to $<100>$ directions.

![FIG. 4. Angular stress and strain dependence of tensile strained In$_{1-x}$Ga$_x$P string resonators. (a) High-stress InGaP with Ga content of $x_{HS} = 58.7\%$. Stress varying between 430 MPa and 640 MPa (top). Dashed gray line: Theoretically calculated stress , using Eqs. 1 and 4. Blue line: Taking a change of elastic properties due to defects into account by a cos(2\theta) angle dependent change of the Young’s modulus (bottom). (b) Low-stress InGaP with $x_{LS} = 52.8\%$. Showing a similar change of the Young’s modulus as in (a). Error bars represent the uncertainty from the weighted mean calculation.](image)
along the [110] direction have a higher density than lines along [100].

![Reciprocal space maps depicting the asymmetric reflections of the HRXRD measurement. On the left the beam is oriented along the [110] and on the right along the [100] direction. The circle indicates the substrate peak. The layer peak position for a 100 % pseudomorphic layer is indicated by a diamond, and for a fully relaxed layer by a triangle. These reciprocal space maps confirm, that the unstructured InGaP is 100 % pseudomorphic. (b) The cathodoluminescence measurement shows dark dislocation lines along the <110> crystal directions, having different defect densities.]

It has been shown, that defects can influence the elastic properties of crystalline materials, and can lead to a softening as well as a hardening of the elastic constants.\[^{11-14}\]

This change of elastic properties can be treated as an effective Young’s modulus \( \sigma(x, \theta)/\varepsilon^x(x) = E(x, \theta) + \Delta E(\theta) \). We extract the deviation \( \Delta E(\theta) \) from the experimentally obtained stress, the strain using Eq. 4 and the theoretically calculated Young’s modulus determined in Fig. 3. The extracted values are shown in the bottom plots of Fig. 4 and clearly reveal an angular deviation from the theoretical Young’s modulus. Both the softening and hardening of elastic constants can be seen for our two different InGaP compositions, softening for the high-stress InGaP and also hardening for the low-stress. Fitting a \( \cos(2\theta) \) function to the data, leads to the deviation functions \( \Delta E_{HS}(\theta) = (-5.53 + 5.13 \cos(2\theta)) \) GPa and \( \Delta E_{LS}(\theta) = (2.44 + 23.40 \cos(2\theta)) \) GPa, respectively. Adding those functions to the theoretical Young’s modulus to calculate the angular stress (Eq. 4), we obtain the solid blue lines in Fig. 4 which yield good agreement with the measured data.

In conclusion, we have explored tensile-strained nanomechanical string resonators fabricated from crystalline In\(_{1-x}\)Ga\(_x\)P. The initial InGaP thin film is pseudomorphically strained for a thickness of 86 nm and a Ga content of \( x_{HS} = 58.7\% \). For the given composition we extracted an angle-dependent tensile stress of up to 650 MPa. The observed angular stress dependence with respect to the crystal orientation is explained by a combination of anisotropic Young’s modulus and a change of elastic properties caused by defects. This enables control over the stress of a nanomechanical resonator for a given heterostructure with fixed Ga content, which in turn could be optimized to enable maximum tensile stress. In addition, angular stress control opens a way to investigate the influence of tensile stress on the dissipation of nanomechanical systems. Stress control and further characterization of strained crystalline resonators will help to gain a deeper understanding in pursuit of ultimate mechanical quality factors.\[^{11-13}\] Finally, InGaP is a promising material for cavity optomechanics, as two photon absorption is completely suppressed at telecom wavelengths.\[^{20,22}\] Moreover, tensile strained InGaP could open a way to combine a high Q nanomechanical system with a quantum photonic integrated circuit on a single chip.\[^{15,26}\]

**SUPPLEMENTARY MATERIAL**

See supplementary material for detailed description of fabrication, calculation of Young’s modulus and comments on critical thickness of InGaP, as well as more details on the HRXRD measurements.

**ACKNOWLEDGMENTS**

Financial support by the Deutsche Forschungsgemeinschaft via the collaborative research center SFB 767, the European Unions Horizon 2020 Research and Innovation Programme under Grant Agreement No 732894 (FET Proactive HOT), as well as the QuantumERA ERA-NET Cofund in Quantum Technologies (project QuaSeRT), as well as the Horizon 2020 Programme is gratefully acknowledged.

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SUPPLEMENTARY MATERIAL

S1. Fabrication

The employed heterostructures were grown by molecular beam epitaxy on 150 nm diameter, 675 µm thick (001) GaAs wafers. Both \( \text{In}_{1-x}\text{Ga}_x\text{P} \) (InGaP) layers on each wafer have a thickness of 86 nm, and are capped by 1 nm GaAs on both sides, respectively. The two investigated Ga contents are \( x_{\text{HS}} = 58.7\% \) (high-stress) and \( x_{\text{LS}} = 52.8\% \) (low-stress).

From bottom to top, the structure consists of a GaAs buffer layer followed by a GaAs/Al\(_y\)Ga\(_{1-y}\)As distributed Bragg reflector. The bottom sacrificial Al\(_y\)Ga\(_{1-y}\)As (AlGaAs) layer has a thickness of 1065 nm. The following two InGaP layers are separated by the 265 nm thick top sacrificial Al\(_y\)Ga\(_{1-y}\)As layer. The aluminum content of all Al\(_y\)Ga\(_{1-y}\)As layers is \( y = 92\% \). We only use the top InGaP and sacrificial AlGaAs in this work.

String resonators are defined by electron-beam-lithography. The negative electron-beam-resist ma-N 2403 serves as etch-mask. To avoid delamination, we apply an adhesion promoter TI Prime before spin-coating the resist. The roughly 240 nm thick resist features good resistance against dry and wet etches. Etch-resistance is further increased by an additional hard bake after development, for 10 min at 120°C in a convection oven. We pattern the string resonators with an inductively coupled plasma (ICP) etch, using a SiCl\(_4\):Ar (1:3) gas mixture. Etching is carried out at a pressure of 1.7 mTorr with 250 W ICP power and 60 W RF power and at a temperature of 30°C. To remove possible chlorine residues of the ICP etch, we soak the samples for 10 min in DI water. With an oxygen plasma cleaner we remove the resist etch-mask. The following buffered HF etch releases the string resonators, by etching the sacrificial AlGaAs with an etch rate of 50–90 nm/s, depending on the crystal direction. After thoroughly rinsing the samples, we use a digital wet etch to remove the GaAs cap layers and possible etch residues. In the end we dry the samples via critical point drying. To avoid degradation of the AlGaAs sacrificial-layer under ambient conditions, the samples are quickly mounted inside the vacuum chamber of the measurement setup.

Under ambient conditions the AlGaAs sacrificial layer quickly degrades and swells, fracturing the suspended InGaP resonators, see Fig. S1 (a), (b). Alternatively the AlGaAs surface can be passivated by rapid thermal oxidation (RTO) as shown in Fig. S1 (c)-(e). This is done by a 5 min long rapid thermal anneal at 550°C in an oxygen atmosphere. The AlGaAs surface is stable for at least three weeks. To what extent this treatment changes the mechanical properties of InGaP string resonators, remains a topic of further investigation.

![FIG. S1. AlGaAs degradation and rapid thermal oxidation (RTO).](image-url)

(a) InGaP nanoresonators after a few days in ambient air. Cracks at the InGaP clamping points are already visible due to the degradation of the underlying AlGaAs. (b) The same clamping points as in the image above are completely destroyed after 8 weeks in ambient air. (c) Temperature profile of the 5 min long RTO, with a 100 sccm O\(_2\) flow. (d) Scanning electron micrograph of the heterostructure, right surface treated with RTO and left untreated side produced by a fresh cleave right before imaging. (e) Heterostructure after 21 days in ambient conditions. Untreated surface shows severe degradation from swollen AlGaAs layers while the treated surface remains unaffected.
S2. Calculating Young’s modulus

The Young’s modulus relates stress and strain in the one-dimensional case of isotropic, uniaxial materials via Hooke’s law $\sigma = E \varepsilon$. For anisotropic materials, stress and strain are related by the fourth rank compliance $S$ or stiffness $C$ tensors, $\sigma = C \varepsilon$ and $\varepsilon = S \sigma$. Those tensors are simplified to $6 \times 6$ matrices with three independent components, for the case of the cubic symmetry of e.g. the zincblende crystal structure. For the [100] crystal direction, Young’s modulus simply equals the inverse first component of the compliance matrix $S$:

$$s_{11} = \frac{1}{E} . \quad (S1)$$

However, since we know the elastic constants $c_{ij}(x)$ of In$_{1-x}$Ga$_x$P, we start with the stiffness matrix to calculate the Young’s modulus for any desired crystal direction:

$$C(x) = \begin{pmatrix} c_{11}(x) & c_{12}(x) & c_{12}(x) & 0 & 0 & 0 \\ c_{12}(x) & c_{11}(x) & c_{12}(x) & 0 & 0 & 0 \\ c_{12}(x) & c_{12}(x) & c_{11}(x) & 0 & 0 & 0 \\ 0 & 0 & 0 & c_{44}(x) & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{44}(x) & 0 \\ 0 & 0 & 0 & 0 & 0 & c_{44}(x) \end{pmatrix} . \quad (S2)$$

Using the rotation-matrices Eqs. $(S6)$, we can perform clockwise rotations through an angle $\theta$ about a desired major crystal axis, initially $X=[100], Y=[010]$ and $Z=[001]$:

$$C(x, \theta) = (\text{Rot}_i(\theta) \cdots \text{Rot}_j(\theta) C(x)) \text{Rot}_j^T \cdots \text{Rot}_i^T , \; i, j = X, Y, Z . \quad (S3)$$

For example the application of Rot$_Z(45^\circ)$ to a [100] direction, produces a vector pointing along the [110] direction. By inverting the rotated stiffness matrix, we calculate the compliance matrix:

$$S(x, \theta) = (C(x, \theta))^{-1} . \quad (S4)$$

As in Eq. $(S1)$, the Young’s modulus is the inverted first component $s_{11}$ of the compliance matrix. But now it is along the rotated [100] direction, thus pointing in any desired direction:

$$E(x, \theta) = s_{11}^{-1}(x, \theta) \quad (S5)$$

A full three dimensional plot of the Young’s modulus is shown in Fig. $S2$. The red line represents the area cut through the (001) plane depicted in Fig. 3.
FIG. S2. 3D plot of Young’s modulus of InGaP. Arrows indicate major crystal directions. Red line represents the cut through the (001) plane as depicted in Fig. 3.
S3. Critical thickness

This section provides an overview of the existing models to calculate the critical thickness of strained epilayers, and is a summary of Refs. 32 and 33.

There are two different approaches to treat the generation of dislocations, and thus yield a different thickness when a strained epilayer starts to relax. The first approach is the force-balancing model by Matthews. This model considers the forces on dislocation lines. Those are misfit strain (as driving force), which is opposed by the tension in the misfit dislocation line. An epilayer starts to relax when the force exerted by the misfit strain becomes larger than the tension in a dislocation line. The other approach to calculate the critical thickness is the energy-balancing model by People and Bean. It compares the homogeneous strain energy density with the energy density associated with the generation of dislocations. If the surface strain energy density exceeds the self-energy of an isolated dislocation, dislocations are introduced which lead to a relaxation.

The formulas to calculate the critical thickness $h_c$ are as follows:

- Matthews:

$$ h_c = \frac{b}{4\pi \varepsilon^\parallel} \frac{(1 - \nu \cos^2(\Theta))}{(1 + \nu) \cos(\alpha)} \ln \left( \frac{h_c}{b} \right) $$

- People & Bean:

$$ h_c = \frac{(1 - \nu)}{1 + \nu} \frac{b^2}{16\pi \sqrt{2} a_\infty^\parallel \varepsilon^\parallel} \ln \left( \frac{h_c}{b} \right) $$

$a_\infty^\parallel$: lattice constant of In$_{1-x}$Ga$_x$P

$b = a_\infty^\parallel / \sqrt{2}$: magnitude of Burgers vector of dislocation

$\nu = c_{12}/(c_{11} + c_{12})$: Poisson ratio of InGaP; $c_{ij}$ elastic constants.

$\varepsilon^\parallel(x) = \frac{a_\parallel^\parallel - a_\infty^\parallel(x)}{a_\infty^\parallel(x)}$: in-plane strain, $a_\parallel^\parallel$ distorted lattice constant due to lattice mismatch, as in Eq. 1.

$\Theta$: angle between dislocation line and Burgers vector (60° for most III-V semiconductors)

$\alpha$: angle between slip direction and direction in epilayer plane which is perpendicular to the line of intersection of the slip plane and the interface (60°)

By solving these formulas we obtain the curves of Fig. S3, which demonstrate that the model of Matthews gives a more conservative estimate of the critical thickness than the one by People & Bean. The horizontal line marks the thickness of the investigated InGaP film, whereas the vertical lines indicate the two Ga contents. Clearly, the model of Matthews is not able to describe the pseudomorphic high stress material which would greatly exceed the critical thickness. We thus conclude that the MBE growth process is better described by the model of People & Bean.
FIG. S3. Critical thickness, calculated for $\text{In}_{1-x}\text{Ga}_x\text{P}$ on GaAs. Dotted line is the model based on the work of Matthews and solid line of People and Bean. The vertical lines show the Ga contents investigated in this work. The horizontal line indicates the employed thickness 86 nm.
S4. High resolution x-ray diffraction measurements on the high-stress InGaP wafer

X-ray reciprocal space mapping (RSM) is used for strain and structural characterization of crystalline heterostructures. The x-rays scatter from the electronic density of the crystal, reproducing the lattice planes of the crystal. Measuring the angle of the scattered x-rays and using Bragg’s law it is possible to calculate the distance between lattice planes, which can be related to the lattice constant of the crystal. Thin, mismatched layers distort tetragonally and therefore the lattice parameter parallel to the wafer surface differs from the perpendicular one, \( a_{\parallel} \neq a_{\perp} \).

With symmetric RSMs, the 002 and 004 reflections of Fig. S4 and S5, one can extract the perpendicular lattice constant. The following symmetric RSMs show a strong GaAs substrate peak at wave-vectors of about \( Q_{S[001]}^{002} \approx 2.223 \text{ Å}^{-1} \) and \( Q_{S[001]}^{004} \approx 4.446 \text{ Å}^{-1} \), for the 002 and 004 reflection respectively. This corresponds to the substrate lattice constant of \( a_S = 5.653 \text{ Å} \). The layer peak of InGaP can be seen at \( Q_{L[001]}^{002} \approx 2.246 \text{ Å}^{-1} \) and \( Q_{L[001]}^{004} \approx 4.493 \text{ Å}^{-1} \) and corresponds to the strained, perpendicular lattice constant \( a_{\perp} = 5.595 \text{ Å} \).

When additionally measuring asymmetric RSMs, the 224 reflections of Fig. S4 and S5 and the 404 reflections of Fig. S6, it is possible to extract the parallel lattice parameters from the peak position on the \( Q_{[110]} \) axis. Since both the substrate and layer peak have the same \( Q_{[110]} \) component, their parallel lattice constants equal: \( a_S = a_L = 5.653 \text{ Å} \), this is true for all the scan directions ([110], [101], [010], [100]).

We can clearly see with these findings, \( a_S = a_L \neq a_{\perp} \), that the high-stress InGaP wafer is 100% pseudomorphic.

| Component | Value |
|-----------|-------|
| \( Q_{[110]} \) (Å⁻¹) | \( Q_{[001]} \) (Å⁻¹) |
| \( Q_{S[001]}^{002} \) | \( Q_{S[001]}^{004} \) |
| 2.223 | 4.446 |

| Component | Value |
|-----------|-------|
| \( Q_{L[001]}^{002} \) | \( Q_{L[001]}^{004} \) |
| 2.246 | 4.493 |

**FIG. S4.** HRXRD reciprocal space maps depicting the symmetric 002 and 004 and asymmetric 224 reflections for a x-ray beam oriented along the [110] direction.
FIG. S5. HRXRD reciprocal space maps depicting the symmetric 002 and 004 and asymmetric 224 reflections for a x-ray beam oriented along the [110] direction.

FIG. S6. HRXRD reciprocal space maps depicting 404 reflections. On the left the beam is along the [100] and on the right along the [010].
In addition to the RSMs, to get further insight on the strain, one can look at a reflection curve as a function of scattering angle to extract structural information from the epitaxial structure. Figure S7 shows a HRXRD curve of the 002 reflection in blue. The substrate peak can be seen at a scattering angle of about 31.6°, and the InGaP layer peak at about 32°. The smaller, regularly distributed peaks arise from the GaAs/AlGaAs super-lattice. By simulating and fitting the slow and rapid oscillations of the curve, it is possible to extract the thicknesses and compositions of the different layers in a heterostructure. The simulation, red line in Fig. S7, is done with the heterostructure of Tab. S1. The nominal heterostructure composition is shown in Tab. S2. From the measurement one can see, that the two InGaP layers have different compositions and also point to an In gradient along the growth direction. We have not taken into account the compositional gradients in our calculations in the main paper. This will be subject to follow-on work.

**FIG. S7.** HRXRD curve of the 002 reflection for the high-stress InGaP wafer. Fitting the simulation (red) to the signal (blue) leads to the layer-compositions and -thicknesses shown in Table S1. Inset: Zoom-in on the InGaP peak at a scattering angle of about 32 degree.

**TABLE S1.** Heterostructure of the high-stress InGaP wafer. Layer-compositions and -thicknesses are extracted by fitting the HRXRD scan of Fig. S7. One can clearly see deviations from the nominal heterostructure in Tab. S2. The composition of both InGaP layers differs and they also both have gradient in their In content along the growth direction.

| Layer | Repeat | Material   | Al content     | In content     | Thickness (µm) |
|-------|--------|------------|----------------|----------------|----------------|
| 0     | 1      | GaAs substrate | —              | —              | —              |
| 1     | 1      | AlGaAs     | 0.99536        | —              | 0.01449        |
| 2     | 41     | GaAs       | —              | —              | 0.07801        |
|       |        | AlGaAs     | 0.96612 (top)  | 0.98612 (bottom) | 0.09002        |
| 3     | 1      | GaAs       | —              | —              | 0.07670        |
| 4     | 1      | AlGaAs     | 0.96032        | —              | 1.05490        |
| 5     | 1      | GaAs       | —              | —              | 0.00100        |
| 6     | 1      | InGaP      | —              | 0.46721 (top)  | 0.07762        |
|       |        |            |                | 0.42233 (bottom) |                |
| 7     | 1      | AlGaAs     | 0.98550        | —              | 0.26441        |
| 8     | 1      | GaAs       | —              | —              | 0.00100        |
| 9     | 1      | InGaP      | —              | 0.42227 (top)  | 0.08643        |
|       |        |            |                | 0.38401 (bottom) |                |
| 10    | 1      | GaAs       | —              | —              | 0.00100        |
TABLE S2. Nominal heterostructure composition of the high-stress InGaP wafer.

| Layer | Repeat | Material | Al content | In content | Thickness (µm) |
|-------|--------|----------|------------|------------|---------------|
| 0     | 1      | GaAs substrate | —          | —          | —             |
| 1     | 1      | AlGaAs | 0.92       | —          | 0.2720        |
| 2     | 41     | GaAs   | —          | —          | 0.0780        |
|       |        | AlGaAs | 0.92       | —          | 0.0906        |
| 3     | 1      | GaAs   | —          | —          | 0.0780        |
| 4     | 1      | AlGaAs | 0.92       | —          | 1.0653        |
| 5     | 1      | GaAs   | —          | —          | 0.0010        |
| 6     | 1      | InGaP  | —          | 0.41       | 0.0859        |
| 7     | 1      | GaAs   | —          | —          | 0.0010        |
| 8     | 1      | AlGaAs | 0.92       | —          | 0.2646        |
| 9     | 1      | GaAs   | —          | —          | 0.0010        |
| 10    | 1      | InGaP  | —          | 0.41       | 0.0859        |
| 11    | 1      | GaAs   | —          | —          | 0.0010        |