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ABSTRACT

We present polarized emission from single hexagonal silicon–germanium (hex-SiGe) nanowires. To understand the nature of the band-to-band emission of hex-SiGe, we have performed photoluminescence spectroscopy to investigate the polarization properties of hex-SiGe core–shell nanowires. We observe a degree of polarization of 0.2 to 0.32 perpendicular to the nanowire c-axis. Finite-difference time-domain simulations were performed to investigate the influence of the dielectric contrast of nanowire structures. We find that the dielectric contrast significantly reduces the observable degree of polarization. Taking into account this reduction, the experimental data are in good agreement with polarized dipole emission perpendicular to the c-axis, as expected for the fundamental band-to-band transition, the lowest energy direct band-to-band transition in the hex-SiGe band structure.

I. INTRODUCTION

Silicon is the prime material system used in electronic communication technologies due to its advantageous physical, electronic, and technological properties. For further development in the efficiency of these computing technologies, the integration of photonics on chips is a major focus of research.[1] Monolithic integration of laser onto silicon will make it possible to fabricate full optoelectronic chips.[2] Thus, a group IV material like silicon which has a direct bandgap, will open up important pathways for the further development of optoelectronic chips.[3,4] Much research exists into group-IV lasers based on germanium (Ge) such as germanium–tin,[5] strained-Ge,[6,7] and heavily doped Ge.[8] Recently, a breakthrough has been made in the development of a direct bandgap group-IV material system based on the silicon itself, hexagonal silicon–germanium (hex-SiGe).[9] In a previous work, it is shown that this new silicon-based group-IV semiconductor material hex-SiGe has a direct bandgap optical transition.[9] The transformation of the crystal structure from the more common cubic phase to the hexagonal phase causes the conduction band minima at the L-point to fold back to the Γ-point, in such a way that the alloy obtains a direct bandgap for Ge-contents above 60%. These hex-SiGe alloys are very efficient light emitters, with strong

band-to-band emission up to room temperature and a radiative recombination rate comparable to many state-of-the-art group III–V optical materials, such as indium-phosphide (InP) and gallium-arsenide (GaAs).[9,10] Hexagonal silicon–germanium might also find applications in quantum wells and quantum dots, while two-dimensional honeycomb group IV materials might provide an interesting field[11–13] of future research. These properties and the compatibility with Si technology provide opportunities for hex-SiGe in silicon photonics for integrating light sources, detectors, and optical amplifiers.

While efficient recombination is observed and well documented, many of the optical and electrical material properties of hex-SiGe are still unknown or unverified. A property that has not been experimentally verified is the polarization selection rules of the fundamental band-to-band transition. Control over the polarization of the emitted light is important for many applications, such as the engineering of LEDs[14–17] and nanophotonic communication technology.[18,19] Thus, verifying the inherent polarization of the emission is important for the development of such devices.

Matrix elements for the different polarization directions in hex-Ge have been determined by *ab initio* density-functional theory (DFT) calculations.[20,21] These calculations show that the

lowest conduction band is the Γ_{8c}^--band, while the Γ_{9v}^+-band is the highest valence band and the Γ_{7v+}^+-band is the second valence band. The calculated optical transition matrix elements parallel and perpendicular to the c-axis are very different, where the c-axis corresponds to the (0001) crystal direction and the growth axis of the nanowires. The fundamental optical transition ($\Gamma_{8c}^- \rightarrow \Gamma_{9v}^+$) across the bandgap emits light polarized perpendicular to the c-axis ($\perp$c), while the second valence band transition ($\Gamma_{8c}^- \rightarrow \Gamma_{7v+}^+$) is expected to emit non-polarized light. Thus, the two transitions can be distinguished from the polarization of the emitted light. For hex-SiGe alloys, the polarization selection rules are the same, but the exact degree of polarization can be slightly different as compared to elemental hex-Ge. Hex-(Si)Ge, with a strong polarized emission $\perp$c-axis for the fundamental direct bandgap, is very different from the more commonly known cubic-germanium (cub-Ge). For cub-Ge (as well as for cubic silicon), direct transition at the Γ-point in the Brillouin zone will exhibit strong optical absorption, but will have no polarized selectivity with respect to the equivalent axis for hex-Ge due to isotropy of the material.[20,22,23]

In this work, we experimentally show that the polarization of the fundamental direct bandgap transition is polarized perpendicular to the c-axis. We study both the geometric and material contributions to the degree of linear polarization of the emitted photoluminescence (PL). Indeed, after considering geometrical effects from the diameter of the nanowire, a clear polarization-dependent emission that is fully dominated by dipole moments oriented perpendicular to the c-axis is observed from hex-SiGe. This result confirms the predictions made by DFT calculations on the optical transition of the lowest band-to-band recombination.[20]

II. SAMPLE PREPARATION

The hex-SiGe nanowires were grown using the crystal transfer method as first reported by Hauge et al.[24] and the growth conditions were comparable to the conditions used by Fadaly et al.[9]. First, wurtzite GaAs core nanowires are grown with an outer diameter of 180 nm, on a GaAs (111)B substrate in an MOVPE reactor using a gold-catalyzed vapor–liquid–solid growth mechanism. The c-axis direction of the wurtzite crystal coincides with the vertical nanowire axis. A hex-SiGe shell is grown around the core, copying the crystal structure of GaAs. In Fig. 1(a), an SEM image of an array of hex-SiGe nanowires is shown, with an average diameter of 620 nm. The figure shows the smooth surfaces of the side facets of the nanowires and the ball-shaped mixed phase top. This top forms through vapor–solid growth on the top [0001] surface of GaAs, which is equivalent to the cubic [111] surface. As the top is not expected to emit light because of its indirect bandgap, we do not expect a significant influence of the ball-shaped top on the collected average emission polarization. Three different batches of wires are investigated, having an average diameter of 620 ± 20, 980 ± 120, and 1610 ± 50 nm, which will be referred to as samples A, B, and C, respectively.

To compare the optical properties of the three different diameters, the nanowires are first optically characterized [see Fig. 1(b)] using Fourier Transform InfraRed (FTIR) spectroscopy using a 976 nm continuous wave laser as the excitation source. The measurements are performed at T = 4 K and an excitation density of

$\sim$5 kW/cm^2 with an excitation spot diameter of $40 \, \mu$m. The nanowires are measured in the as-grown orientation, as shown in Fig. 1(d). When comparing the PL spectra, the three samples have similar PL emission energy and shape. The bandgap of the hex-SiGe nanowires used in this study can be estimated to be around 0.54 eV for all three samples. From a previous work, we can estimate the average incorporated Ge-content to be around 80%, which results in a direct bandgap.[9] Since each sample has an identical crystal structure, the only effect that influences the polarization of the emission is the geometry of the nanowires.

The nanowires are mechanically transferred to a silicon substrate to be able to measure the polarized emission with respect to the c-axis. This substrate is covered with a 200 nm layer of gold (Au) and a top layer of 400 nm silicon oxide (SiO$_x$), which is shown schematically in Fig. 1(c). The gold layer acts as a reflector to maximize both the excitation of the nanowire and the collection efficiency. SiO$_x$ acts as a spacer layer to prevent direct contact between the nanowire and the Au layer. The nanowires are transferred at a low density, as shown in Fig. 1(d), to ensure that each wire is measured individually.

III. EXPERIMENTAL SETUP

The degree of polarization (DOP) is determined by measuring the intensity of PL emission at different linear polarization angles. We define polarization anisotropy as DOP $= (I_\perp - I_\parallel)/(I_\perp + I_\parallel)$, where $I_\parallel$ and $I_\perp$ are the parallel and perpendicular polarized emission intensities with respect to the nanowire axis. A positive (negative) DOP implies that the emission is polarized perpendicular (parallel) to the long nanowire axis. To obtain the DOP and the polarization angle of the emission, we use Malus' law as a fitting function of the polarization-dependent PL intensity measurement

$$I(\theta) = \frac{1 + \text{DOP}}{2} \cos^2(\theta - \theta_0) + \frac{1 - \text{DOP}}{2} \sin^2(\theta - \theta_0). \quad (1)$$

Here, θ is the polarization angle at which the PL intensity I is measured and θ_0 is the angle of maximum intensity.

Individual nanowires are characterized using the micro-PL setup shown in Fig. 2(a). Here, the nanowires are optically pumped using a mode-locked fiber laser (NKT ORIGAMI 10-40) ($\lambda_{\text{pump}} = 1032$ nm, a repetition rate of 40.0 MHz, pulse length <200 fs). The wires are pumped through a 36×/0.40NA Cassegrain reflective objective and the emission is collected through the same objective. The signal is collected by a Superconducting Nanowire Single-Photon Detector (SNSPD) system with a measurement window up to a wavelength of 2.35 μm (Single Quantum EOS110). The polarization-dependent measurements are performed at cryogenic temperatures (T = 7 K) and an excitation density of 60 μJ/cm^2 with an excitation spot size diameter of 4 μm. The collected light is spectrally filtered using bandpass filters with an optical transmission range from 1350 to 2400 nm. Care has been taken to completely filter out the excitation laser and only select the PL emission from hex-SiGe. A linear polarizer plate is placed before the detector to select only one polarization angle to be measured. By rotating this plate to a different angle, another polarization angle can be selected. However, the SNSPD has a polarization-
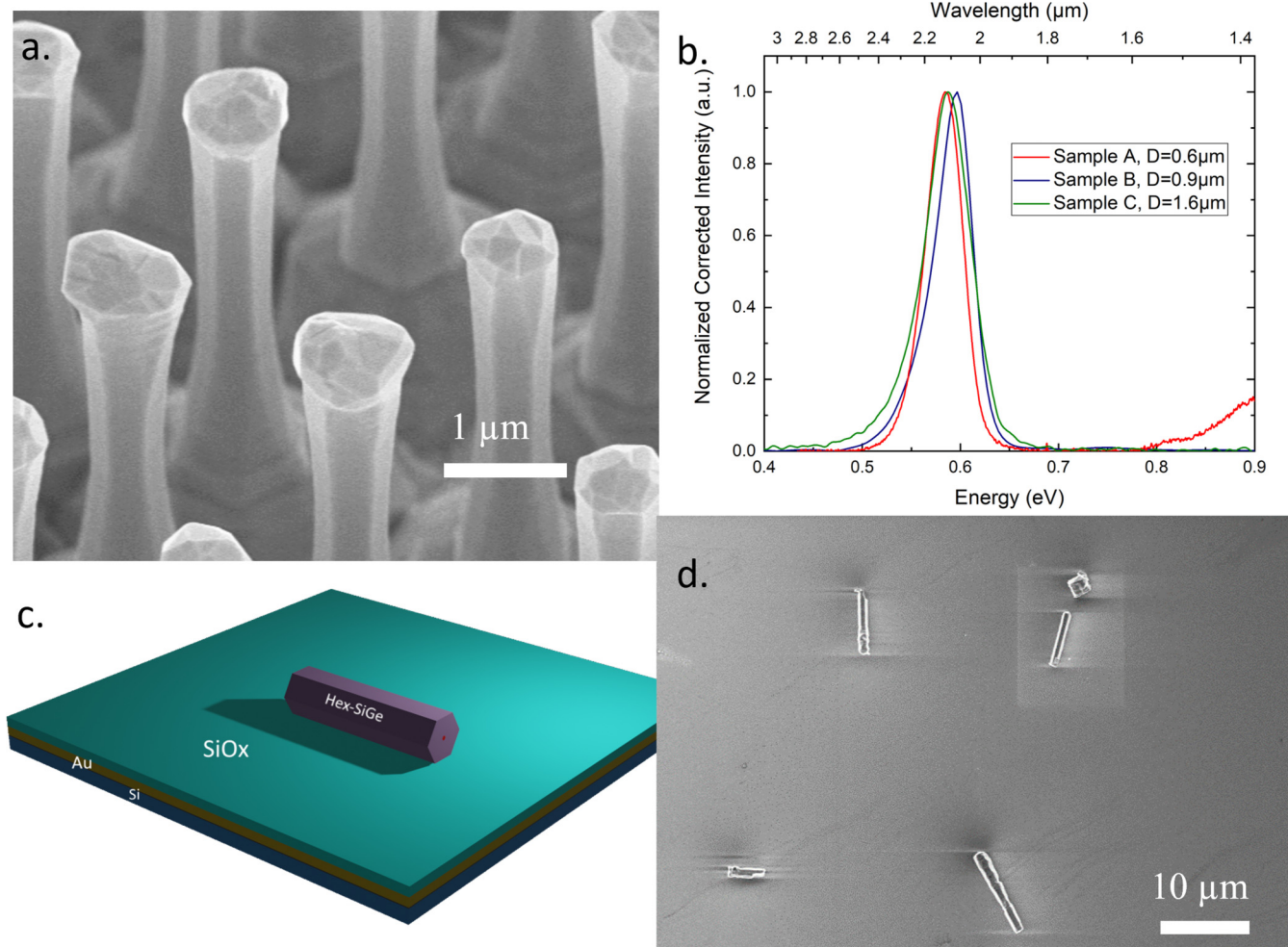
FIG. 1. (a) SEM image of as-grown hex-SiGe nanowires with an average diameter of 620 nm. (b) Photoluminescence spectra of an ensemble of wires from three different samples (P = 5 kW/cm², T = 4 K). All three samples emit at similar energies, confirming that the Ge-content of the samples is comparable. (c) Schematic of a single WZ-GaAs/hex-SiGe core/shell nanowire swiped on a SiOx substrate with a buried gold layer. (d) Scanning electron microscopy (SEM) image of nanowires transferred to the substrate. Low density of the transferred nanowires ensures that the nanowires are measured individually.

dependent sensitivity, which causes certain polarization angles to be measured more efficiently than others. Thus, after the linear polarizer selects the measured linear polarization, a half-wave plate is subsequently used, which rotates the selected polarization to the polarization angle for which the SNSPD is most efficient. This ensures that the detector is effectively polarization insensitive. The remaining polarization insensitivity of our detector is measured using a known non-polarized light source. This deviation is then used to adjust the data in post-processing to remove systematic errors in the measurement of the DOP, which is shown in more detail in the supplementary material. A CCD camera in a separate optical path in the setup is used to determine the orientation of the nanowires. From the collected image, the angle of the c-axis with

respect to the vertical axis is calculated before each measurement, with an accuracy of 5°.

IV. RESULTS AND DISCUSSION

The polarization-dependent measurement of one selected hex-SiGe nanowire from each sample is shown in Figs. 2(b)–2(d). Several wires are measured to average out any statistical variation between the wires. From samples A, B, and C, we measured 10 wires, 8 wires, and 15 wires, respectively, where the wires are selected on the quality of their morphology. The orientation of the wire is shown as a straight red line in the graphs, and a green dashed line shows the ⊥c-axis direction. For all three samples, the maximum PL intensity is measured when the polarizer is oriented
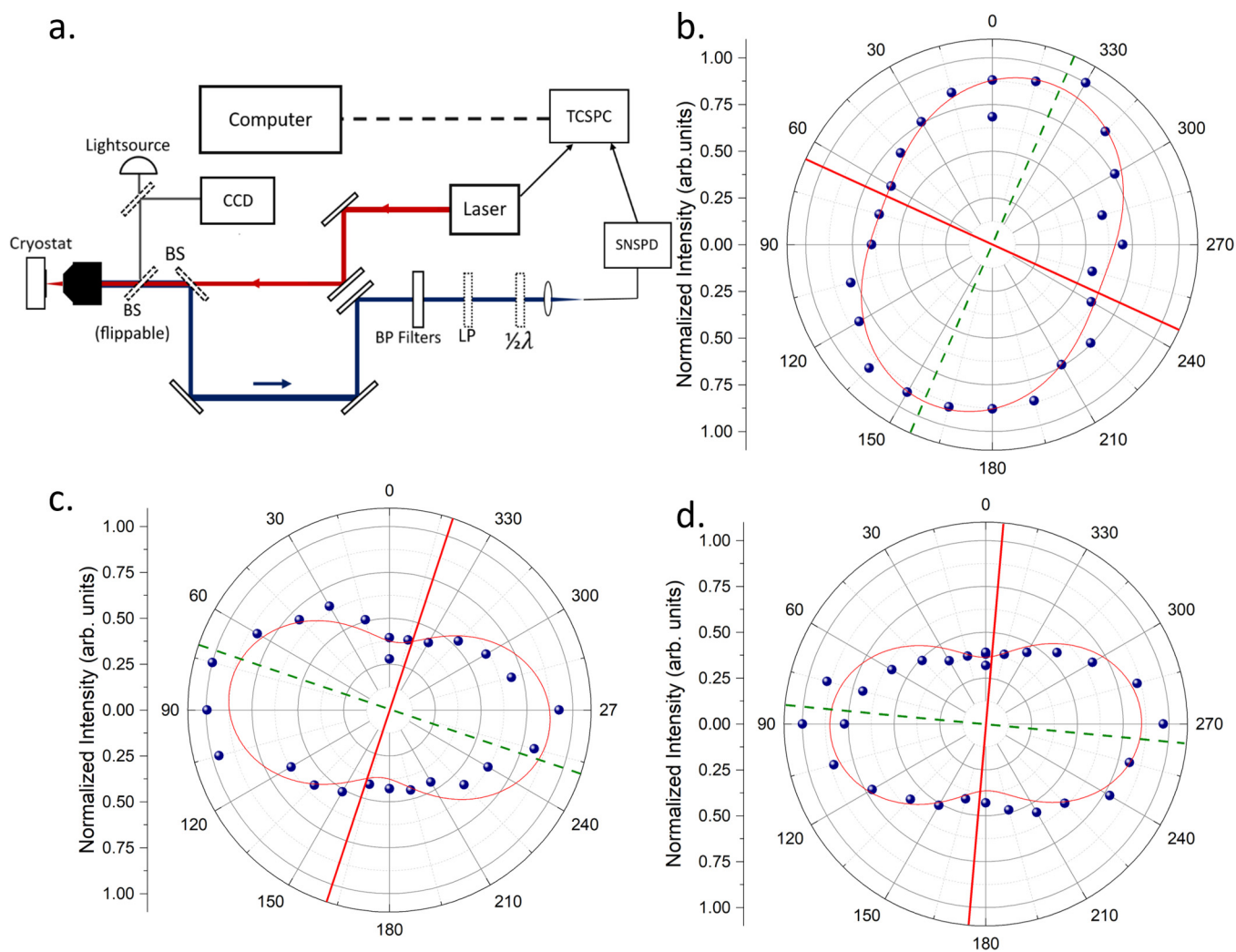
FIG. 2. (a) Schematic of the time-correlated single photon counting (TCSPC) optical setup for the analysis of polarized PL emission from a single hex-SiGe nanowire. A linear polarizer plate (LP) is used to select the polarization of the emission, which is collected by the superconducting nanowire single-photon detector (SNSPD). A half-wave plate ($1/2\lambda$) is used to rotate the polarization such that the polarization dependence of the detector is compensated. A CCD camera is used to determine the orientations of single nanowires. (b)–(d) Polar plots of the PL intensity as a function of the polarization angle for all three nanowire diameters, D = 0.6, 0.9, and 1.6 μm in panels (b)–(d), respectively. The intensity points (blue) are fitted with Malus' law (red), showing the strongest emission perpendicular to the nanowire axis. The orientation of the nanowire c-axis is given in red, and the dashed green line indicates the direction perpendicular to the c-axis.

perpendicular to the nanowire axis, which is the first indication that the lowest energy transition of hex-SiGe is the polarized $\perp$c-axis. The polarization data are subsequently fitted using Eq. (1) to determine the DOP as well as the polarization angle θ_0 of the emitted PL. The average DOP is plotted as a function of the wire diameter in Fig. 3(a), in which a weighted DOP is calculated by averaging the DOP for all wires transferred from each sample. In Fig. 3(b), the average polarization angle θ_0 is shown with respect to the c-axis. All three samples together show an average offset angle of $\theta_0 \approx 90°$, which means the strongest emission from the nanowires is the polarized $\perp$c-axis. Sample A shows an average

polarization angle of 80°, which deviates from the expected angle but has no clear physical explanation. The uncertainty in the estimation of nanowire orientation is likely the cause of the deviation.

The measured DOP increases slightly with increasing nanowire diameter. The DOP varies from DOP = 0.26 $\pm$ 0.03 for Sample A with 600 nm diameter to DOP = 0.312 $\pm$ 0.009 for Sample C with 1610 nm diameter. The measured increase in polarization anisotropy with diameter is expected from the dielectric contrast effect, which is a well-documented effect for 1D nanostructures like nanowires.[25–27] When the nanowire absorbs light with a wavelength larger than the diameter, not all linearly
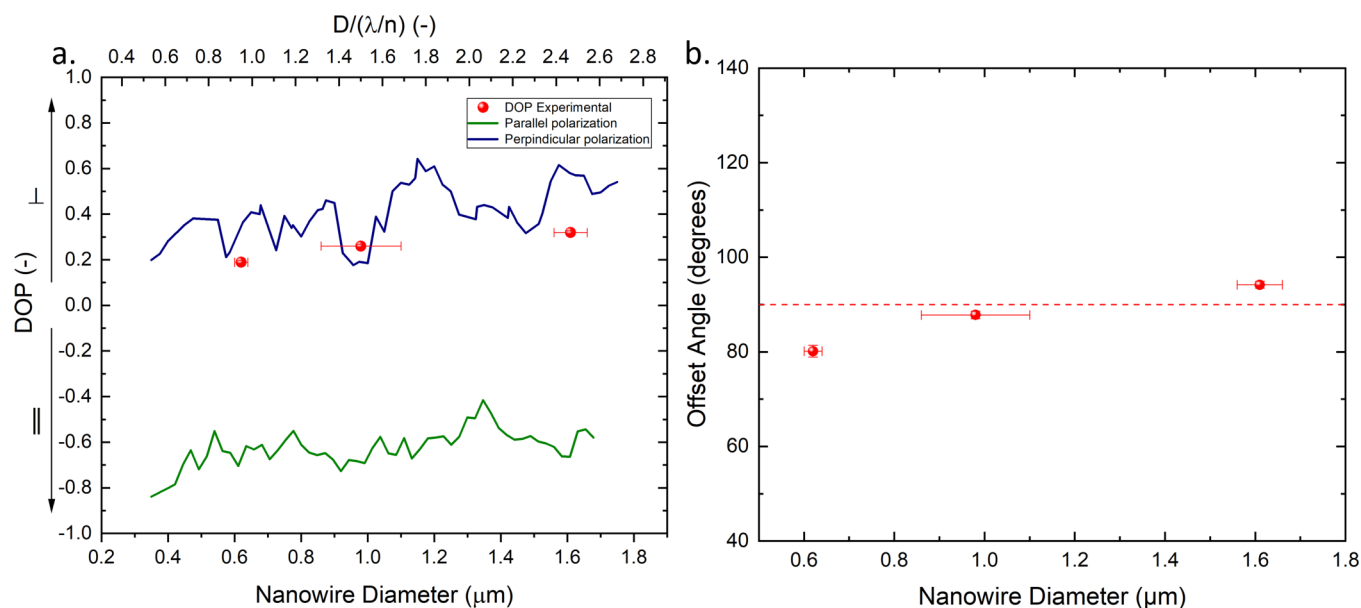
FIG. 3. (a) The red points represent the average degree of polarization (DOP) measured for each nanowire diameter using Malus' law fit on the experimental data in Fig. 2. The uncertainty in the nanowire diameter is due to variations between wires from the same sample. The data is averaged over 10, 8, and 15 wires for the data points at 0.62, 0.94, and 1.62 μm diameter. Results from FDTD simulations of the DOP from the far field of hex-$Si_{0.2}Ge_{0.8}$ NWs are included as a function of NW diameters. The blue (green) line represents the far field emission if the dipoles are polarized perpendicular (parallel) to the long nanowire axis. (b) The polarization angle θ_0 with respect to the long axis of the nanowires for different nanowire diameters, shows perpendicular polarization.

polarized components of the light are absorbed equally – this is due to the strong attenuation of light polarized perpendicular to the nanowire. For thicker nanowires, with diameters above 10% of the wavelength, the dielectric contrast effect becomes much less pronounced.[28] The contribution of the dielectric contrast has mainly been investigated for non-polarized dipole emission, like in cubic semiconductors. In our work, however, the hexagonal crystal structure of hex-SiGe is expected to yield a strong perpendicular polarization of dipole moments, changing the expected contribution of the dielectric contrast to the DOP compared to cubic crystal structure nanowires.[28] Thus, as the dielectric contrast effect is expected to influence the experimentally observed DOP, numerical simulations need to be performed to study the effect of nanowire geometry on the emission.

We also performed the polarization-dependent measurements on vertically oriented (standing) hex-SiGe nanowires as shown in Fig. 1(a). The excitation and collection of light are both done parallel to the nanowire axis, which is the c-axis. Now only polarization anisotropy caused by effects in the radial direction of the nanowire can be observed. From theoretical predictions, no polarized emission is expected from the selection rules of the bandgap of hex-(Si)Ge,[20] because the radial axes of the nanowires are synonymous due to the sixfold symmetry of the hexagonal crystal structure in the c-plane. In Fig. 4, we show the polarized emission of the vertically oriented nanowires of sample C. As expected from the theoretical predictions, no degree of polarization is observed. All polarization

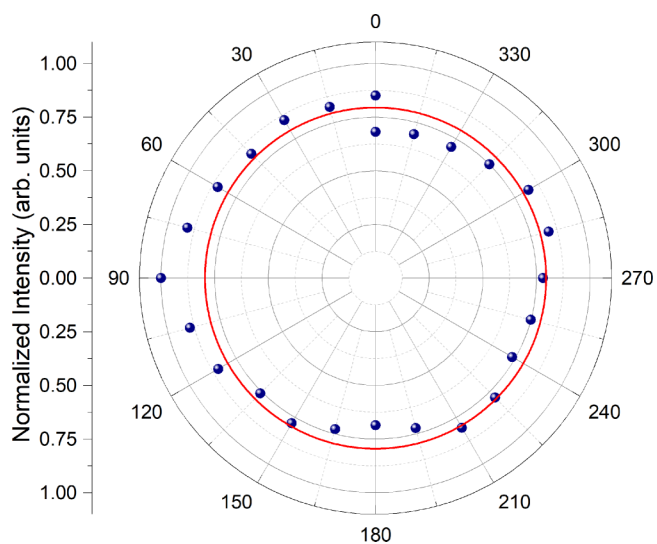**FIG. 4.** A polar plot of normalized PL intensity as a function of the polarization angle for a nanowire with $D = 1.6$ μm in a vertical orientation like Fig. 1(a). The intensity points (blue) are independent of the polarization angle, with no preferred polarization angle for the emission. The fit with Malus' law (red) is a perfect circle with a degree of polarization equal to zero.

angles show comparable PL intensities and the fit using Malus' law of Eq. (1) results in zero polarization anisotropy. It is reported that it is possible to observe polarized emission due to changes in the guided optical modes for small nanowire diameters.[25] The diameters of the nanowires of sample C are likely to be large to significantly influence polarization anisotropy in this direction. This confirms that in the c-plane of the hex-SiGe crystal, no polarization selection rules are present.

V. SIMULATIONS

We performed finite-difference time-domain (FDTD) simulations, using the commercially available Ansys/Lumerical software. In the simulation, a core/shell WZ-GaAs/hex-$Si_{0.2}Ge_{0.8}$ nanowire with a hexagonal morphology is positioned horizontally on a SiO_x slab. To appropriately simulate photoluminescence, four fields of 25 dipoles are positioned randomly throughout the nanowire to average out possible variations of the polarization anisotropy due to the dipole position. The emission wavelengths of the dipoles are closely matched to the spectra of the samples as shown in Fig. 1(b). The nanowire diameter is varied from $D = 0.35$ to $1.75\,\mu m$, while the GaAs core is kept at $D = 0.15\,\mu m$, to match the experiments. The far field of the polarization-dependent light emission from the nanowire is calculated at 1-m distance. This intensity is integrated over a full numerical aperture (NA) and split into electric field components in both lateral directions to extract linear polarization anisotropy.

To establish the effect of the dielectric contrast on the measured polarization anisotropy, we simulated the DOP for two extreme cases: (1) the dipole light emission is polarized completely perpendicular or (2) completely parallel with respect to the nanowire c-axis. The simulated amount of polarization anisotropy then directly reveals the influence of the dielectric contrast on the experimentally observable DOP. The simulated DOP is shown in Fig. 3(a) for both cases. The DOP for the dipole moments aligned perpendicular to the NW, in blue, is approximately 0.2 for the lowest diameter NW and rises to 0.54 for the largest diameters within the simulation range. The DOP increases gradually with only a slight oscillation, which can be caused by dielectric contrast effects[28] and due to residual effects of dipole positions inside the wire, which are potentially not fully averaged out. The magnitude of the DOP in a nanowire is significantly lower than the simulated DOP of 0.94 with the same dipole orientation in an infinite slab of hex-SiGe, showing that the contribution of the dielectric contrast effect is significant. These effects are due to the dielectric confinement of the electric field of light, originating from the difference in the dielectric constant of the nanowire and the vacuum. We also simulated the DOP for dipoles aligned parallel to the NW axis, which results in a strongly negative DOP, contrary to our experimental results. When we compare the experimental DOP to the simulated curves in Fig. 3(a), we observe a fair agreement between the experimental data and the simulated DOP for perpendicular polarized dipoles. This agrees with the selection rules for the lowest energy optical transition from DFT calculations,[20] which predict polarization in the ⊥c-axis direction.

The values of the experimental data are slightly lower than the simulation, which suggests that the optical strength of dipole emission in the ‖c-axis polarization direction is non-zero. A contribution from this polarization direction might be explained by a small contribution of the second valence band transition, ($\Gamma_{8c}^- \rightarrow \Gamma_{7v}^+$), which is non-polarized. Although the transition from the second valence band is symmetry-forbidden in hex-Ge, alloy disorder in hex-SiGe is expected to break the translational symmetry, at least resulting in a weakly allowed non-polarized contribution from the second valence band transition.[20] Apart from this minor deviation between theory and experiment, we conclude that the fundamental optical transition in direct bandgap hex-SiGe is a strongly polarized ⊥c-axis, confirming the DFT calculations for hex-(Si)Ge and giving more insight into the optical properties of this novel material.

VI. CONCLUSIONS

In summary, we confirmed the polarization selection rules of the lowest optical band-to-band transition in hex-SiGe as predicted by the DFT calculations. We measured the polarization-dependent PL intensity of hex-SiGe nanowires of different diameters to consider the dielectric contrast effect. This results in a DOP = 0.32 for a hex-SiGe nanowire with a diameter of $D = 1.6\,\mu m$. The contribution of the dielectric contrast was clarified by performing FDTD calculations of polarization anisotropy due to the dielectric contrast in nanowire geometry. The simulated DOP for perpendicular-oriented dipoles agrees well with the measured DOP in our hex-SiGe NWs. The polarization-dependent emission from the top facet of a vertically oriented nanowire shows no degree of polarization, which is expected from theoretical symmetry arguments. In contrast to the unpolarized emission from cub-Ge, we confirm that the emission of hex-SiGe is mainly dominated by dipole emission perpendicular to the c-axis of the crystal. Overall, this research provides an improved understanding of the optical properties of the novel material hex-SiGe. Understanding the polarization rules of the band-to-band transitions is very important for creating light detection devices from hex-SiGe and the development of a silicon-based light source for the realization of active silicon photonics.

SUPPLEMENTARY MATERIAL

See the supplementary material for the analysis of the correction factors for the polarization-dependent PL data.

AUTHOR DECLARATIONS
Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Marvin A. J. van Tilburg: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Writing – original draft (lead); Writing – review & editing (equal). **Wouter H. J. Peeters:** Investigation (supporting). **Marco Vettori:** Investigation (supporting). **Victor T. van Lange:** Formal analysis (supporting); Investigation (supporting). **Erik P. A. M. Bakkers:** Supervision (supporting); Writing – review & editing (supporting). **Jos E. M. Haverkort:** Supervision (lead); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are openly available in Zenodo.org at http:doi.org/10.5281/zenodo.7326144 (Ref. 29).

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