Engineering of the Second-Harmonic Emission Directionality with III–V Semiconductor Rod Nanoantennas

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The ability to engineer nonlinear optical emission from nanostructures is a key challenge to create efficient and compact components for integrated devices. This paper shows a method to control and manipulate the directionality of second-harmonic generation emission by engineering geometry and position of rod nanoantennas. Single and dimer nanoantennas are fabricated by slicing III–V semiconductor nanowires with focused ion beam milling. The nonlinear optical response of nanoantennas is tailored by adjusting their length and position to achieve a targeted phase difference. The studied GaAs nanoantennas have a wurtzite structure that allows to achieve preferable directions for the second-harmonic emission compared to a typical bulk zinc blende structure from top-down fabricated nanostructures. Wurtzite nanoantennas provide a pure electric dipole response at the second-harmonic wavelength, which together with pi-phase control of emitted light is used for designing nonlinear emission patterns. The simulation results show how to redirect the second-harmonic beam up to 30° and how to tailor the emission profile by adding elements. This method of second-harmonic generation manipulation and phase array engineering can be applied to different types of nanowires and nanostructures. Nonlinear beam steering with structures from nanowires will foster the creation of compact optical components for integrated circuits.

1. Introduction

Optical beam steering and light redirection gained a lot of attention from the research community as a solution for free space optical communication. Control and manipulation of light at the nanoscale level by engineering arrays of optical resonators is one of the most important challenges to develop new types of compact optical components,[1,2] such as low-loss miniature laser sources,[3] tunable mirrors, and other integrated photonics devices.[4-5] The field of flat optics,[6] based on large scale arrays of resonant nanostructures, utilizes wavefront and polarization state engineering[7] for applications in all-optical switching,[8,9] polarization filtering,[10] optical holography and cryptography.[11-12] The development of flat photonic systems has been recently reinforced by dielectric and semiconductor Mie-resonant structures,[13-18] owing to their low losses and artificial magnetic optical response[19,20] in comparison to typically used plasmonic systems. Moreover, the strong nonlinear optical properties of some dielectric and semiconductor materials make them the most promising materials to create efficient structures for nonlinear light emission.[21] This gives more opportunities for wavefront engineering of higher-order harmonics.[22-24] Among these materials, III–V semiconductors such as GaAs or AlGaAs, due to their strong second-order nonlinear optical susceptibility,[25-27] have the highest potential of application in nonlinear photonics devices. Individual III–V nanoantennas supporting electric and magnetic multipolar resonances[26,28-32] have already demonstrated ultrahigh efficiency of second-harmonic generation (SHG) in subwavelength structures.[13-15] In addition to these advantages, semiconductor nanostructures can also be electrically doped and used for subwavelength active devices.[36,37]

The control of the nanoantennas’ geometry allows to manipulate the resonances excited in the structure. Thus we can enhance the generation efficiency, to engineer a directionality and a phase, and to control the polarization’s orientation for linear scattered light and nonlinear emissions. Some of these effects were recently demonstrated in single semiconductor nanodisks with low-radiative resonances, in nanoparticle dimers through...
One way to engineer the directionality of scattered light with all-dielectric antennas is based on the interplay between the electric and magnetic modes. In the case of lower-order multipoles such as electric (ED) and magnetic (MD) dipole modes, their interference may lead to a strong forward or backward light scattering also known as Kerker effect. [43] and such structures resemble artificial Huygens elements. [46] The manipulation of nonlinear emission within a single structure can also be performed by the excitation of higher-order multipoles at the frequency of higher harmonics. This interference between higher-order multipoles can satisfy the generalized Kerker condition. [47] Another way to achieve even higher control over the directionality of the light generation and emission is to use ensembles of nanoantennas assembled in a phase array. By controlling the geometry and position of each single nanoantenna, we can combine the emission fields and engineer the phase difference between neighboring elements composing a phase array ensemble. This allows steering the direction of the emitted nonlinear optical response. [22,48–50]

In reaching the final goal of effective light manipulation with nanoscale nonlinear sources the material system plays an important role. As an alternative to commonly used lithography methods for dielectric nanostructures and metamaterials fabrication, semiconductor nanowires provide a unique platform for nonlinear nanophotonics. First, they offer nonlinear optical properties coming from the crystal orientations that are hardly accessible in materials grown with common epitaxial technology. The crystal structure of III–V semiconductor materials may be hexagonal wurzite (WZ) or zinc blende (ZB), usually oriented in (111) direction instead of the (100) orientation typical for epitaxially grown films. [51–54] Recent research demonstrated that employing ZB materials with (111) or (110) crystal orientation leads to significant improvement of the nonlinear emission directionality. [25,55]

In III–V nanowires this ability to fabricate WZ or (111) ZB crystal structures induces a specific nonlinear optical behavior, which allows us to design and control the electromagnetic response of the nanoantenna. Moreover, nanowires can be structured with different methods of nanofabrication, such as lithography, focused ion beam (FIB) milling, to form individual nanostructures on various substrates. Finally, besides individual nanophotonic structures, arrays of nanowire can be used as a platform to create metasurfaces and metamaterials for nonlinear photonic devices.

Here, we used the advantages of GaAs nanowires to design and fabricate nonlinear nanoantennas with controllable emission of the second harmonic (SH) radiation. We demonstrated a new experimental approach to fabricate single rod, symmetric and asymmetric dipole nanoantennas, which are cut from bottom up grown WZ GaAs nanowires with focused ion beam milling technique. [56] The key advantage of this approach is that it enables fabrication of individual structures with precision up to tens of nanometers. We demonstrated that design of dimer nanoantennas allows to engineer the SH far-field radiation pattern caused by the strong dipolar polarization of WZ GaAs nanorods at the SH wavelength. With the clear matching between experimental and simulation results, we propose a method for engineering a 1D phase array of rod nanoantennas that provides a narrow emission profile of the SH as well as bent emission at larger angles with respect to the incident light direction. The approach of the phase array engineering demonstrated in our work is general and can be extended to other types of bottom-up grown III–V semiconductor nanowires, such as InAs, InP, GaP, AlGaAs, and InGaP. It can be applied to create various nonlinear nanophotonic devices compatible with complementary metal-oxide-semiconductor (CMOS) technologies.

2. Results and Discussion

2.1. Theoretical Model

The crystal structure of a nanomaterial defines its electromagnetic response at the SH wavelength along with the type of excited resonant modes. [56] Normally, if the nanowires are grown on (111) substrate they adopt (111) growth direction independently on ZB or WZ crystal structure. These crystal structures are different from epitaxially formed GaAs thin films, which are usually growing on (001) oriented GaAs substrates having pure ZB. We demonstrate the advantage of the crystalline phases in nanowires that allows to generate an ED response in rod nanoantennas formed from such nanowires. The WZ crystal structure belongs to the C∞v crystallographic point group of symmetry and provides a purely ED resonance excited at the SH wavelength in the direction of the nanorod’s long axis due to the particular selection rules (a more detailed comparison between ED resonances excited in ZB and WZ nanowires is presented in Section 3 in the Supporting Information). [56,57] The WZ crystal structure and dipolar SH response make GaAs nanowires attractive candidates to create phase nanoantennas for redirecting and steering the SHG emissions by fabricating structures with dominant ED responses.

In a first step, we developed a theoretical model that helps to design the nanoantennas and to achieve the desired far-field patterns of the linear scattering and SH emission. The finite element model calculates the linear scattering and nonlinear response with the undepleted pump approximation as well as the multipolar decomposition of the generated fields both at fundamental and SH wavelengths (more details given in the Supporting Information).

Using this model, we calculated the SHG from a single nanorod, with 200 nm diameter and different lengths, with WZ type of nonlinear tensor to analyze the contributions from different multipoles to the SHG signal. In the simulations, the rods were excited by an electromagnetic plane wave with the electric field polarized along the long axis of the rod (see schematic in Figure S1.1a, Supporting Information). As shown in Figure 1a, for a nanorod with 200 nm length (inset), the main contribution to the SHG spectrum in the spectral region from 800 to 1000 nm is coming from the ED mode only, while the contribution of other multipoles is negligible, which is typical for WZ crystal structure (see results of multipole decomposition for different lengths of rods in Section 2, Figure S2.1, Supporting Information). We have also performed a multipole decomposition of the SHG at different polarizations of the incoming excitation for the characterized single rod nanoantenna with length
Figure 1. a) Calculated SHG intensity with multipole decomposition for a single rod nanoantenna, b) calculated SHG intensity for the electric dipole (ED) contribution, and c) SHG phase of the electric dipole contribution from different single rod nanoantennas with the length in the range of 150–400 nm. d) Schematic of the combination of nanoantennas with length 200 and 350 nm at an excitation wavelength of 900 nm. The SHG intensity of the EDs is equal for both nanoantennas while the phase difference of the ED emissions is shifted by $\pi$, leading to destructive interference in the forward direction.

350 nm (the results are presented in Figure S2.2, Supporting Information). As can be seen from the results, the maximum of the total SHG intensity and ED contribution corresponds to 0° polarization. The purely ED response of the nanorod allows straightforward designing of the far-field pattern: by choosing the nanorods length and separation distance one can control the phase and the amplitude of the generated dipole modes in each nanorod. Thus, we simulated the behavior of the ED moment induced at SH frequency in rods with the length varying from 150 to 400 nm, which can be easily achieved experimentally with
the provided fabrication approach. Figure 1b,c shows respectively the amplitude and the phase of the induced ED in single structures with different lengths. The resonant wavelength shifts to higher values as the length of the rod increases. For structures with lengths up to 300 nm the ED contribution dominates in the SHG spectrum, while for longer lengths, the higher-order multipoles start to have a more significant contribution. Nevertheless, in the SHG spectrum we can find a region, where the ED is dominating over the other multipole components. As can be seen from Figure 1b, for the various nanorod lengths the phase of the SH response varies in a range larger than π, which allows us to obtain both constructive and destructive interference condition among different nanorod elements composing a 1D phase array. Figure 1d presents the schematic of two nanorods with length 200 and 350 nm at excitation wavelength 900 nm, for which the SHG intensity of the ED is equal and the phase difference is π, leading to destructive interference in the forward direction.

This nanodimer system is a basic example showing the feasibility of the far-field SH pattern manipulation with proper designing of the 1D phase array components. We have compared three types of nanoantenna: single nanorod antenna, symmetric and asymmetric nanodimer antenna. The results of this modeling are summarized in Table 1, they were calculated for normal illumination by a plane wave with an excitation wavelength of 900 nm. The far-field radiation from a single nanorod with different SHG intensities, for which the SHG intensity of the ED is equal and the phase difference is π, leading to destructive interference in the forward direction. This provides a constructive interference in the forward direction as it is shown in the Table 1.

At the same time, as can be seen in Figure 1c, the amplitudes of the electric dipoles generated at the SH wavelength in single nanorods of 200 and 350 nm lengths are equal while the phase shift between them is π. This gives an opportunity to combine both elements to effectively cancel the SH emission in the forward direction. The far-field diagram of the SH emission for the asymmetric dimer nanorod antenna has a two lobes pattern with the maxima of the SH emission at an angle of 30°. Table 1 shows the evaluation of the SH emission patterns for different configurations of nanoantennas.

### Table 1. The SH emission patterns for different configurations of nanoantennas. Schematic of the rod antennas and their corresponding simulation of the SHG far-field emission pump at 900 nm wavelength.

| Geometry                              | Single Nanorod | Symmetric dimer | Asymmetric dimer |
|---------------------------------------|----------------|-----------------|------------------|
| Schematic and SHG simulation          |                |                 |                  |
| for the far-field radiation           |                |                 |                  |

### 2.2. Experimental Results

Next, we confirm this model with experimental results by demonstrating the ability of such GaAs nanorods to achieve the control of the SHG emission directivity. Nanowires were first grown by molecular beam epitaxy (see the Experimental Section) and mechanically transferred on a glass substrate covered with indium tin oxide (ITO). Then, we sliced the nanowires with FIB milling into different configurations of single and dimer rod nanoantennas (more detailed illustration of the fabrication process is presented in the Experimental Section and in Section S4 in the Supporting Information).

We characterized different configurations of nanoantennas: single rods, symmetric and asymmetric dimers. The linear scattering spectra from these structures were measured in transmission mode with a dark-field microscope using a collection objective with a numerical aperture (NA) of 0.55 (see in the Supporting Information). The results of the measured and calculated linear spectra of the nanoantennas are presented in Table S5.1 in the Supporting Information. The multipole decomposition for the rod nanoantenna shows the domination of ED dipole contribution (Figure S5.2, Supporting Information). We also simulated the near field distributions for the linear scattering (see Figure S9.1, Supporting Information) to show field distributions in characterized nanoantennas.

To measure the SHG intensity we use a home built nonlinear microscope equipped with a tunable Ti:Sapphire laser, which covers the region from 700 to 1080 nm, combined with an optical parametric oscillator system that covers the region from 1080 to 1600 nm. The SHG responses from the structures were measured by sweeping the excitation wavelengths in the region from 800 to 1400 nm with steps of 10 nm (see more details in the Experimental Section). Figure 2 presents the experimental and simulation results for the SHG spectra from single nanoantenna, symmetric and asymmetric dimers, and insets with corresponding scanning electron microscopy (SEM) images of fabricated structures. The SHG spectra were measured at 0° polarization of incoming excitation, where the SHG intensity is maximal.

Figure 2 presents the experimental and simulation results for the SHG spectra from single nanoantenna, symmetric and asymmetric dimers, and insets with corresponding SEM images of fabricated structures. The measured SHG spectra, presented in
Figure 2. Measured and calculated SHG spectra for a) a single rod nanoantenna, b) a symmetric dimer, and c) an asymmetric dimer. Insets: Scanning electron microscopy (SEM) images of the corresponding structure.

Figure 3. a) Experimental results of SHG polar plots for a rod nanoantenna with a length of 350 nm. Calculated SHG polar plots for a similar structure with b) wurzite and c) zinc blende crystal structure.

Figure 2a–c, correspond with the simulation results regarding the position of the SHG peak and width. The peak broadening in Figure 2a, and shift in frequency of the peak in Figure 2b with extra plateau observed after the peak for a) and c) can be explained by some imperfections of the structures due to the FIB process which is not reflected in the perfect cylindrical shape of the simulations. As we can see from the SEM image in Figure 2b, the edges are not totally straight, and the substrate environment is not flat.

The SHG signal was collected using a 100X objective with an NA = 0.8, which corresponds to the collection angle ±53°. The NA of the objective is one of the factors that limit the collection of the SHG emitted beyond the solid angle. Additionally, for every 50 nm, we recorded the SHG polar plots as a dependence of the SHG intensity with respect to the polarization of the incoming excitation. The shape of the SHG polar response originates from the geometry of the nanoantennas, responsible of the Mie modes (see Figure S2.1, Supporting Information), and the material crystal structure. To identify the impact of the crystal structure, we consider the polar response far from dominating Mie resonances. The ED is dominating resonance, as can be seen in Figure 1a and Figure S2.1c in the Supporting Information. We performed the SHG polar measurements in the spectral range of 800–1400 nm (Figure 3). As we already showed in our previous works in the regions where the contribution of the ED (and MD as well) resonances to the emitted SHG is low, the shape of the SHG polar signal is defined by the crystal structure of the nanoantenna.

We compared the shapes of the measured SHG polar response and the corresponding simulations with pure WZ and pure ZB nanoantennas to identify their crystal structure (Figure 3 and Figure S6.2, Supporting Information). If we compare the SHG polar plots for a similar nanoantenna with WZ or ZB crystal structure, in Figure 3 the shape of the measured polar response is corresponding to WZ crystal structure at the wavelengths where the contribution from low order Mie resonances is negligible. For ZB crystal structure the simulated SHG polar signal has four lobes shapes at these wavelengths. The calculated SHG polar signals (Figure 3b) for WZ crystal structure always have two lobes that correspond to the experimental results for the nanoantennas presented in Figure 3a. The simulations for the nanoantenna with pure ZB, presented in Figure 3c, show nonzero SHG intensity at 90° excitation polarization. The experimental results and the simulations for pure WZ crystal structure for all wavelengths have two lobes shape and zero SHG intensity for 90° excitation polarization.

The SHG polar plots measured for other structures are presented in Section S6 in the Supporting Information. In Figure S6.1 in the Supporting Information the symmetrical dimer has a small contamination of ZB crystal structure. We have demonstrated that the ED is still the dominating resonance for structures with mixed WZ + ZB crystal structure (see Section S2 in
After we identified the crystal structure of the fabricated structures, we measured the directionality of the generated SH emission. For this we performed back-focal plane (BFP) measurements of the SHG far-field radiation pattern (see the experimental setup presented in Section S7 in the Supporting Information) to test the directionality of the SH emission. In the BFP setup, the SHG is collected using an objective with NA = 0.8, which corresponds to 53° of the generated angle. Figure 4 shows the results of the BFP measurements together with the calculations of the SHG far-field radiation patterns. We measure at 900 nm pump wavelength for the single rod and the symmetric dimer nanoantennas, and at 840 nm for the asymmetric nanoantenna, which corresponds to a phase difference approximately of \( \pi \) (see Figure 1c). For the simulation results we present 3D far-field radiation pattern and corresponding cross-section of the collected SHG. The SHG radiation patterns from the single rod nanoantenna show a domination of the ED in the far-field radiation. The widening of the far-field pattern in the direction along the nanowire axis is related to the additional weak mixing of the higher-order multipole components. These results for the symmetric dimer demonstrate the predicted far-field pattern with a sharper maximum
The presented results allow for further implementation of the proposed approach in order to engineer a 1D phase array consisting of multiple rod nanoantennas, cut from a nanowire, with designed sizes and spacing to achieve a specific phase shift. Using this approach, one can achieve the control over the directionality of the SH emission with longer and more complex 1D phase arrays.

We present several designs to achieve a specific directionality of the SH emission. In Figure 5, we plot the SH far-field diagrams at 830 nm pump wavelength for designed 1D phase arrays with two, three, and four components. In the design shown in Figure 5, we tilted the SHG lobe at an angle allowed by the material parameters and geometry of the nanoantenna. Here, we worked with the noninteracting particles, which mean that the interparticle distance cannot be too short providing that their resonances are not affected by the interaction with other elements in the array. However, one can go beyond that limitation and utilizes their interaction for achieving even larger directivity and tilt angle of the second-harmonic emission.

We simulated the nonlinear emissions with a finite element method and with the analytical dipole model. The observed discrepancy between the dipole model and the exact calculations is related to the additional interactions between the modes in closely placed nanoantennas, which is not taken into account in the dipole approximation. For closely placed nanoantennas with center-to-center distances smaller than the SH wavelength one can achieve a strong asymmetric field emission diagram relative to the ZY plane (Figure 5a). The asymmetry is provided by the phase difference between the dipole moments for nanorods of 165 and 190 nm length leads to a suppression of the SH emissions on one of the sides (see Figure 1b). This can be explained by the interference of two dipoles. In our dipole approximation, we took into account the difference in the magnitude for various nanoantenna sizes. This difference does not affect the directionality but affects the shape of the additional SHG emission lobes. In our model, we considered the influence of the difference in the amplitude and how it can affect the SHG emission lobes. Nanorods with such sizes have a phase

Figure 5. 1D phase array engineering concept. SHG far-field radiation pattern from phase array constructed from a) two nanorods, b) three nanorods, and c) four nanorods. Calculated with the dipole approximation radiation pattern model for d) dimer, e) trimer, and f) quadrimer phase arrays. The pump wavelength is 830 nm.
difference of approximately \( \pi/2 \), and by considering also the distance between them, which is close to \( \lambda/2 \), the SH emission field pattern consists of two similarly placed dipoles interfering destructively on one side and constructively on the other. A more detailed explanation is provided in Section S8 in the Supporting Information. Moreover, with additional nanorods in the array one can achieve much sharper lobes and directivity of the SH far-field pattern (see Figure 5b,c). These results are matching with the noninteracting dipole approximation as shown in Figure 5e,f.

3. Conclusion

We present a method to build a 1D phase array from III–V nanowires sliced with FIB milling for tailoring the nonlinear beam emission directionality. By placing the rod nanoantennas that have a dominant ED resonance and a phase shift between each other we can engineer the angle of the nonlinear response generation. With the slicing of III–V nanowires with FIB technique, we experimentally demonstrate the concept of building a phase array from rod nanoantennas by varying the geometry of the dimer nanorod antennas. Fine tuning the amplitude and phase of the dipole moments induced at the SH wavelength allows to reshape the far-field nonlinear emission diagram and, in particular, to suppress the forward nonlinear scattering. We demonstrated that only WZ crystal structure allows to excite a pure z-dipole (see Figure S3.3, Supporting Information). However, simulations show that the observed directional SHG is robust to inclusions of ZB phase in the material. Beyond that, our approach can be applied to different types of III–V nanowires such as InAs, InP, GaP, AlGaAs, and InGaP. Furthermore, the phase array engineering can be expanded to longer nanorods to implement other features, for example bending with simultaneous enhancement of the SHG. This approach can be applied to different types of nanowires, which makes such 1D phase arrays promising structures to create novel elements for integrated optical devices. The results of this work can be applied for the development of new types of nonlinear wave front shaping photonic components, which can be applied to integrated photonic circuits.

4. Experimental Section

Fabrication of GaAs/AlGaAs Nanorods: The nanorods were fabricated using FIB milling from GaAs nanowires, covered with thin AlGaAs shell, deposited on ITO glass substrate. First, the nanowires were grown by molecular beam epitaxy on Si (111) substrates using Au colloid droplets as a catalyst material.\(^{[60]}\) As a next step, the GaAs nanowires were grown, and \( \text{Al}_{0.5}\text{Ga}_{0.5}\text{As} \) shells were grown in situ to passivate the surface states.\(^{[61]}\) The nanowire sample was irradiated with a halogen lamp source. The scattered light was collected in a transmission geometry using a 50x objective with an NA = 0.55. Nonlinear Optical Characterization: Nonlinear optical measurements of the SHG spectra were performed with a nonscanning transmission optical microscope described in detail in the previous works.\(^{[52,59]}\) For the excitation of the samples, a tunable Ti:Sapphire laser (from 700 to 1080 nm with steps of 10 nm) was used. For the region from 1080 to 1400 nm, structures were excited with the optical parametric oscillator (OPO) system attached to the Ti:Sapphire laser. The laser light was focused with a 10x objective on the ITO covered glass with rod nanoantennas. The signal was then collected with a 100x objective (NA = 0.8) and focused onto a scientific CMOS camera. The polarization was controlled via a half-wave plate placed on the excitation beam path. For the nonlinear optical characterization, the measurements of SHG intensity were performed for different excitation wavelengths as well as different polarizations of the incoming excitation. Similar measurements for the directionality of the SHG were performed using the setup for BFP measurements (see more details in Section S7 in the Supporting Information). Numerical Finite Element Method (FEM) Simulations: The numerical simulations were performed using the COMSOL Multiphysics software package. For the numerical simulations of the scattering cross-section from the rod structures, a model was developed to calculate the linear scattering cross-section from the rod nanoantennas and the spectra of SHG for different geometries. This model considers the numerical aperture of the collection objectives (more details see in Section S1 in Supporting Information). The developed numerical model allows calculating: the linear scattering spectra, the SHG spectra, the multipole decomposition for linear and SHG spectra, and the far-field radiation patterns.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

1D phase array, electric dipole, III–V rod nanoantennas, second-harmonic generation, wurtzite crystal structure

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