Fabrication of sub-micrometer periodic nanostructures using pulsed laser interference for efficient light trapping in optoelectronic devices

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ABSTRACT

Surface nano-texturing can play an important role for efficiency enhancement of light emission and absorption in optoelectronic devices through reduced surface reflection or enhanced broadband absorption. Periodic and uniform semiconductor nanostructures are highly applicable in bandgap tuning applications but are quite challenging to realize through conventional techniques. We present the fabrication of large area and uniform square lattice based periodic nanostructures with 300 - 400 nm spatial periodicity on a GaAs substrate using pulsed laser interference. Single pulses from a plane-polarized pulsed laser working at 355 nm with 20-50 mJ energy and 7 ns pulse duration are used in a conventional four beam interference geometry at an incidence angle of 36.3˚ to realize square lattice patterns on photoresist coated over the GaAs substrate. The optical properties of the proposed designs are studied using FDTD simulations and show more than 95% of electromagnetic energy trapping over a broad optical wavelength range. This semiconductor based nanostructuring technology can find applications in improving the efficiency of solar cells or light emitting devices.

Keywords: Laser interference lithography, pulsed laser, solar energy harvesting

1. INTRODUCTION

Photonic bandgap tunability and modification are important aspects in photonic design of semiconductor materials for enabling unique optoelectronic properties [1]. Periodic nanowires made using surface nanostructuring are an active area of semiconductor research with potential applications in solar cells and optoelectronic components [2-4]. Photonics integration is advantageous in semiconductors to utilize a broad solar spectrum for photovoltaic applications [5]. A large surface to volume ratio and reduction in refractive index contrast between the semiconductor and air are achieved through photonic-based wave guiding and confinement of modes in ordered 2D nanowires, where there is a confinement along XY plane and a free space propagation along the third direction.

Nanowires based on III-V semiconductors, specifically GaAs have been widely studied for solar cells [6], light emitting diodes [7], and photodetectors [8]. In general, semiconductor nanowire structures with typically a few hundred nm diameters and one to few micrometers of axial lengths can allow electromagnetic wave localization along the XY plane with the channelization of electromagnetic waves in the axial direction. Furthermore, the lifetime of the semiconductor devices is another important aspect which is limited by the surface recombination alongside the nanowire structures. These structures can be grown using various growth techniques such as selective area metalorganic vapor phase epitaxy [9] and molecular beam epitaxy [10]. Although these techniques are well-established for semiconductors, uniform and regular periodic growth of nanowires are still a challenge. In addition, the growth of such structures is very much dependent on the crystal orientation of the substrate. Fabricating nanowires in different crystal orientations are still a challenge using the existing bottom-up techniques. Therefore, a regular and uniform fabrication technique with proper scalable design parameters over large area is important to semiconductor photonics. A periodic patterning without the dependence of crystal orientations would add versatility to the semiconductor-photonic integration exploring wide applications.
Uniform integration of photonic and optoelectronic devices in semiconductors are also possible through laser interference lithography (IL) as a large area, low-cost and rapid fabrication approach [11-15]. In this work, we propose a single-step, large area pulsed laser-based interference lithography of four beams to realize sub micrometer scale periodic photonic structures over GaAs substrate towards nanoscale structuring or the growth of uniform nanowires. The current study is aimed at design and fabrication of interfaces for enhanced photonic wave guidance and light trapping towards absorption enhancement. We present square lattice based photonic GaAs nanowire designs, simulations and interference lithography-based fabrication approaches towards enhance light trapping mechanism. FDTD based simulation studies are carried out to show the optical absorption properties of GaAs nanowires in photonic lattices. Single-pulsed laser interference lithography is used to achieve nanostructuring on GaAs substrates. In future, this kind of study can find application in solar photovoltaics, where the device fabrication can be initiated with a positive electron beam resist to obtain similar periodic holes in GaAs for growth of nanowires resulting in pn or p-i-n type of solar cells.

2. SIMULATION STUDIES

2.1. FDTD Simulation

We have carried out FDTD based simulation using Lumerical’s FDTD solution module for studies on the optical properties of the patterned GaAs nanowire arrays in a square lattice as per the model shown in Figure1(a). The diameter of the nanowires (d) is 160 nm, periodicity of square lattice is 300 nm and the axial height (Z span) is varied from 1000 to 2500 nm for the maximization of optical absorbance. Absorbance is calculated as $A = 1 - (R + T)$. In case of a square lattice, we observe 94% absorbance for a Z span of 1µm of the nanowires that increases to 97% in case of a Z span of 2.5 µm as shown in Figure 1 (b). It is also inferred that although, the nanowires with maximum Z span of 2.5 µm show improved absorbance, the change in absorbance with respect to the Z span is not significant. Therefore, for a low-cost fabrication approach that uses less material, processing time and fabrication feasibility, we propose Z span of 1 µm as the preferred design for the solar cell applications. The electric field distributions through the GaAs nanowire photonic crystal showing electromagnetic energy trapping through photonic crystal-based wave guiding are presented in Figure1(c) and 1(d) at two different wavelengths 727 nm and 567 nm.

Figure 1. FDTD simulation model, (a) Square lattice with periodicity $a = 400$ nm, (b) broadband absorption through the designed square lattice based nanowires showing more than 94% absorption from 400-850 nm with an axial spacing of $Z = 1000$ nm that varies up to 97.7% for an axial variation of $Z = 2500$ nm, (c-d) electric field distribution in the YZ plane at two different wavelengths 727 nm and 567 nm showing light trapping due photonic crystal based wave-guiding.
2.2. FEM Simulation
We have carried our finite element method-based simulation using COMSOL Multiphysics on the designed square array-based GaAs photonic crystal with a lattice constant of 400 nm, diameter of 160 nm and height of 1µm. The surface normal plots of the electric field distribution in XY plane or the presence of different eigen modes are shown in Figure 2.

![Figure 2](image_url)

Figure 2. Surface normal plots of electric fields or eigen modes due to the GaAs nanowires arranged in a square lattice (a-i) presents at different eigen frequencies related to 788 nm, 762 nm 559 nm, 530 nm, 511 nm, 450 nm and 439 nm.

2.3. MATLAB Simulation using four beams interference
We have simulated the resultant irradiance profiles due to four plane-polarized interfering beams using MATLAB plane beams and have viewed the effect of interference angle on it. The irradiance profile due to the superposition of plane-polarized beams leading to the regular square lattice is given by:

\[
I = E_n \cdot E_n^* \\
E_n = P_n \cdot E_0 \exp(iK_n \cdot r + \varphi_n)
\]  

(1)

Where, \(P_n\) is the polarization vector, \(K_n\) are the propagation vectors associated with each interfering plane beam. \(\varphi_n\) is the initial phase or the phase of the interfering plane beams. The irradiance profile due to interference of the plane beams is presented in Figure 3 (a - b) for two different interfering angles showing different lattice spacing over same volume. Figure 3 (c) shows the intensity distribution over XY plane.
3. EXPERIMENT AND RESULT ANALYSIS

We have carried out conventional 4 beam laser interference lithography in a vertical geometry as shown in Figure 4 using a nanosecond pulsed laser (Spotlight 1000, INNOLAS Laser GMBH, Germany). We have considered single pulses for laser with powers varying from 15 - 40 mJ per pulse to examine the threshold of the photosensitive material. We have used the 3rd harmonic of an Nd: YAG laser at 355 nm, which is a TEM00 mode with horizontal polarization. The beam path in Figure 4 achieves vertical polarization in the plane of the beams impinging the substrate. Figure 5 shows a surface analysis study using SEM of the realized patterns on a photoresist (AZ1514H) coated crystalline substrate. A uniform square lattice of periodicity approximately 300 nm is realized through this technique for an interference half angle of 36.3°.

Initially, we have recorded the patterns on positive PR coated GaAs substrate and developed for 5 - 30 seconds for an exposure of 15 - 30 mJ to realize square array structures as shown in Figure 5 (a-d). We have carried out a wet-etching of GaAs using Hydrobromic acid (37%): Potassium dichromate: acetic acid (100%) in a ratio 1:1:1 followed by washing with DI water and the SEM image of the samples are presented in Figure 5 (e -f). However, after cleaning the samples with acetone, we could not obtain any patterned region. This may occur due to incomplete exposure of resist till the

Figure 3. Simulated irradiance profile due to multi-beam interference including four plane beams at (a-b) two different interference angles to show different periodicity of the photonic structure within the volume and (c) intensity distribution along XY plane.

Figure 4. (a) Geometry of the four beams interference and (b) schematic of the experimental setup. PM1 and PM2: Periscope mirrors, M1- M6: beam directional mirrors, BS1- BS3: 50: 50 beam splitters and III HM: 3rd harmonics of the Nd: YAG pulsed laser at 355 nm.
substrate interface. We have carried out electron beam lithography-based patterning on GaAs substrate and performed ICP etching to obtain nano-structured GaAs interface and performed optical characterization studies through spectroscopy for the sake of verification of simulation results on the absorbance of the nano-structured GaAs. SEM image of the EBL patterned, and etched GaAs interface is presented in Figure 6(a) and the optical reflectance (R) absorbance (1-R) properties measured through a visible spectrometer is presented in Figure 6(b). It is observed that a reverse patterning in GaAs has reduced the surface reflection up to 11% from 43% for the case of bare GaAs substrate. This shows enhanced optical absorption of 89% from 450 - 800 nm wavelength range for the nanostructured GaAs interface for light trapping applications towards solar photovoltaics.

Figure 5. SEM images of the patterned GaAs substrate due to 4-beams single-pulsed laser interference. (a-c) uniform patterning over large area and the magnified image at 15 mJ exposure, (d) magnified image at 30 mJ exposure, (e-f) wet-etched samples of GaAs and the magnified images.

Figure 6. (a) SEM image of the EBL patterned and ICP etched GaAs and (b) measured reflection and absorption of the patterned GaAs substrate.
4. CONCLUSION

We have carried out a single-pulsed interference lithography for rapid and large area nanostructuring over GaAs substrate. This study is intended towards the surface nano-texturing and ordered growth of nanowires for nanophotonics applications in semiconductor materials. To study the applications, we have carried out FDTD based simulation studies on the optical properties of the designed nanowire array photonic structure. We have observed through simulation and experiment that nanostructuring over GaAs have reduced the surface reflections and enabled high absorption up to more than 97% and 89% respectively over a broad wavelength range (450 - 800 nm). We believe, this kind of study is highly helpful towards semiconductor nanophotonics for solar energy harvesting.

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REFERENCES

[1] Joannopoulos J. D., Villeneuve P. R., Fan S., “Photonic crystals: putting a new twist on light”, Nature 386, 143 (1997).
[2] Yan, R. Gargas D. and Yang P., “Nanowire photonics” 569, 3 (2009).
[3] Barrelet C. J., Greytak A. B., Lieber C. M., “Nanowire photonic circuit elements”, Nano Lett. 4, 1981-5 (2004).
[4] Du Q. G., Kam C. H., Demir H. V., Yu H. Y., and Sun X. W., "Broadband absorption enhancement in randomly positioned silicon nanowire arrays for solar cell applications," Opt. Lett. 36, 1884-1886 (2011).
[5] I. Hochbaum and P. Yang, “Semiconductor nanowires for energy conversion” Chem. Rev. 110, 527-46 (2009).
[6] Czaban J. A., Thompson D. A., LaPierre R. R., “GaAs core–shell nanowires for photovoltaic applications”, Nano Lett. 9,148-54 (2008).
[7] Tomioka K., Motohisa J., Hara S., Hiruma K. and Fukui T., “GaAs/AlGaAs core multishell nanowire-based light-emitting diodes on Si”, Nano Lett. 10, 1639-44 (2010).
[8] Dai X., Zhang S., Wang Z., Adamo G., Liu H., Huang Y., Couteau C., Soci C., “GaAs/AlGaAs nanowire photodetector”, Nano Lett. 14, 2688-93 (2014).
[9] Zhang Y., Fonseka H. A., Aagesen, M. J., Gott A., Sanchez A. M., Wu J., Kim D., Jurczak P., Huo S., Liu H., “Growth of Pure Zinc-Blende GaAs (P) Core–Shell Nanowires with Highly Regular Morphology”, Nano Lett. 17, 4946-50 (2017).
[10] Ikejiri K., Noborisaka J., Hara S., Motohisa J., Fukui T., “Mechanism of catalyst-free growth of GaAs nanowires by selective area MOVPE”, J. Crystal Growth. 298, 616-9 (2007).
[11] C. Lu, RH. Lipson, “Interference lithography: a powerful tool for fabricating periodic structures”, Lase. Photon. Rev. 4, 568-80 (2010).
[12] C. K. Ullal, M. Mallovan, E. L. Thomas, G. Chen, Y. J. Han, S. Yang, “Photonic crystals through holographic lithography: Simple cubic, diamond-like, and gyroid-like structures”, Appl. Phys. Lett. 84, 5434-6 (2004).
[13] Behera S, Kumar M, Joseph J., “Submicrometer photonic structure fabrication by phase spatial-light-modulator-based interference lithography”, Optics Lett. 41, 1893-6 (2016).
[14] Behera S, Joseph J. “Design and fabrication of woodpile photonic structures through phase SLM-based interference lithography for omnidirectional optical filters”, Opt. Lett. 42, 2607-10 (2017).
[15] Behera S, Joseph J. “Metamaterial structures of variable and gradient basis orientations embedded with periodic linear defects: phase engineered design, single step optical realization, and applications”, Appl. Opt. 58, 50-5 (2019).