Enhanced light trapping by focused ion beam (FIB) induced self-organized nanoripples on germanium (100) surface

Bhaveshkumar Kamaliya,1,2 Rakesh G. Mote,3,a Mohammed Aslam,1 and Jing Fu4

1Department of Physics, Indian Institute of Technology Bombay, Powai, Mumbai 400076, India
2IITB-Monash Research Academy, Indian Institute of Technology Bombay, Powai, Mumbai 400076, India
3Department of Mechanical Engineering, Indian Institute of Technology Bombay, Powai, Mumbai 400076, India
4Department of Mechanical and Aerospace Engineering, Monash University, Clayton, VIC 3800, Australia

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In this paper, we demonstrate enhanced light trapping by self-organized nanoripples on the germanium surface. The enhanced light trapping leading to high absorption of light is confirmed by the experimental studies as well as the numerical simulations using the finite-difference time-domain method. We used gallium ion ($Ga^+$) focused ion beam to enable the formation of the self-organized nanoripples on the germanium (100) surface. During the fabrication, the overlap of the scanning beam is varied from zero to negative value and found to influence the orientation of the nanoripples. Evolution of nanostructures with the variation of beam overlap is investigated. Parallel, perpendicular, and randomly aligned nanoripples with respect to the scanning direction are obtained via manipulation of the scanning beam overlap. 95% broadband absorptance is measured in the visible electromagnetic region for the nanorippled germanium surface. The reported light absorption enhancement can significantly improve the efficiency of germanium-silicon based photovoltaic systems. © 2018 Author(s).
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Recently, the research on germanium-based nanostructures has gained significant attention due to the potential applications in optical trapping, tera-hertz signal emission, and plasmonic response in the mid-infrared region of the electromagnetic spectrum.1–4 The optical trapping is significantly enhanced by the nanostructures like nanopyramids1 and nanoporosity2 over germanium. The enhancement is attributed mainly to the multiple-reflections in the nanostructured domain. The nanostructured germanium is also shown to exhibit terahertz radiation emission3 even though germanium is an indirect band-gap semiconductor. Researchers have achieved three- to five-fold increase in the amplitude of terahertz radiation emitted from the nanostructured germanium surface as compared to the bare-germanium surface. Nanostructured germanium also has potential to be used as an efficient mid-infrared plasmonic detector due to enhanced local surface plasmonic resonance.4 The quasi-random nanostructures of silicon fabricated using the wrinkle lithography technique,5 which are morphologically similar to the germanium nanoripples, have been reported to achieve absorption enhancement by a factor of 1.6 in the 800–1200 nm wavelength range.

The focused ion beam (FIB) technique is extensively used by researchers to form nano- and sub-nanostructures via self-organization on different surfaces such as nanoripples on germanium,6,7 titanium,8 diamond,9 LaAlO3, SrTiO3, Al2O3,10 nanowires on InP,11 antimony;12 nanofins on platinum;13 and nanodots on InI3.11 When ions are bombarded on the surface, selective surface erosion

a Author to whom correspondence should be addressed: rakesh.mote@iitb.ac.in
occurs, and the atoms from concave regions erode faster than those from the convex regions.\textsuperscript{14} Competition between selective surface erosion and surface diffusion induced by ion beam sputtering leads to re-organization of the surface atoms, and thus the nanostructures are formed.\textsuperscript{14,15} The germanium pyramidal arrays were fabricated using FIB milling,\textsuperscript{1} whereas the nanoripples can be formed by self-organization of the surface atoms due to the bombardment of the focused ion beam. The self-organizing operation using focused ion beam is relatively fast compared to the FIB milling giving the advantage of large throughput. The controlled orientation of self-organized nanoripples is useful as a template for functional nanostructure-based applications like the growth of plasmonic metal nanoparticles,\textsuperscript{16} aligned nanowires on the nanoripples surface,\textsuperscript{17} and to align anisotropic liquid crystal molecules.\textsuperscript{18} When nanoripples are used as a template for aligning metal nanorods and nanowires, the plasmonic properties would be dependent on the polarization of the incident light, and the differently oriented nanoripples on the same surface could lead to the multi-responsive plasmonic system. The optical absorption of ordered and periodic nanoripples could be expected to depend on the polarization of the incidence light. The formation of self-organized nanoripples on germanium induced by FIB bombardment was first reported by Zhou et al.\textsuperscript{9} They reported instantaneous formation of nanoripples for the ion dose of $-5.2 \times 10^{17}$ ion/cm$^2$ and revealed that the nanoripples are aligned in the direction of beam scanning. It is believed that the formation of nanoripples and its preferred orientation in the scanning direction is due to selective sputtering of the concave surface and propagating micro-explosions in the direction of scanning.\textsuperscript{6,19,20} However, the resulting topography prediction and its control are still less explored, and the effect of the beam overlap on the morphology of FIB induced germanium nanoripples has not been studied.

In this paper, we investigate the formation of self-organized nanoripples on the germanium (100) surface by raster scanning of FIB. We propose and establish a novel fabrication route for switching nanoripple alignment by changing the scanning beam overlap. The strong dependency of nanoripple’s orientation with the beam overlap is observed by fast Fourier transform (FFT) of the microscopic images obtained via scanning electron microscopy (SEM). Further, the optical behavior of the nanoripples like light trapping and reflectance is studied using finite-difference time-domain (FDTD) simulations and verified experimentally. The nanoripples are shown to exhibit enhanced light absorption.

The 30 keV Ga$^+$ FIB, with current 100 pA and beam diameter 30 nm, was scanned in raster scanning manner on the germanium (100) surface. The FIB was aligned with the surface normal and raster scanned on the sample as shown by the arrow in Fig. 1(a). In the present work, we studied the evolution of nanostructures on germanium (100) with the variation of Ga ion beam overlapping. We varied the beam overlap from 0% to −250% in order to investigate the evolution of ripple morphology as described in the supplementary material (Sec. A). Figures 1(b)–1(h) shows the SEM images of nanoripples formed instantaneously after Ga$^+$ FIB irradiation of $5.66 \times 10^{17}$ ion/cm$^2$ for different beam overlaps. Careful observation of SEM results reveals that for extreme negative beam overlap of −250% [as seen from Fig. 1(h)], the prominent ripples are aligned in parallel with the raster scanning direction. It can be noticed that on increasing the beam overlap from −200% to −25% [Figs. 1(c)–1(g)], the prominent nanoripples change their direction of orientation. Nanoripples are randomly oriented for −100%, −50%, and −25% beam overlaps [Figs. 1(c)–1(e)]. However, in the case of 0% beam overlap [Fig. 1(b)], most of the nanoripples are aligned perpendicular to the raster scanning direction.

The orientation pattern of the nanoripples is also confirmed by two-dimensional fast Fourier transform (FFT) of the corresponding SEM image as shown in the inset of Figs. 1(b)–1(h). The broad range of spatial frequencies are lying in the elliptical domain which has a major axis parallel to the raster scanning direction, as seen in the inset of Fig. 1(b). This suggests that the nanoripples are oriented perpendicular to the scanning direction for 0% beam overlap. However, for the −200% and −250% beam overlaps, the spatial frequency domains have a major axis perpendicular to the raster scanning direction, which implies that the prominent nanoripples are oriented parallel to the scanning direction. The literature on the FIB induced nanoripples reports that the nanoripples tend to align in the raster scanning direction.\textsuperscript{6} It is interesting to note here that even in the case of the non-linear scanning directions such as a spiral scanning direction the nanoripples are aligned in the spiral fashion.\textsuperscript{6,21} However, in this study, we observed a novel phenomenon that nanoripples are formed...
FIG. 1. Schematic of FIB processing at (a) 90° angle with surface and left to right scanning direction and raster scanning in the pixel method for 0% beam overlap; (blue arrow shows the direction of raster scanning); SEM images of nanoripples fabricated at fixed dose (∼5 × 10^{17} ion/cm^2) and different beam overlaps: (b) 0%, (c) −25%, (d) −50%, (e) −100%, (f) −150%, (g) −200%, and (h) −250%; [(b)−(h)] the inset images represent FFT for the corresponding SEM image.

parallel to the scanning direction only for the extreme negative beam overlap, whereas they are formed perpendicular to the scanning direction for zero beam overlap. In between the extreme negative and zero beam overlapping conditions, they are oriented randomly. This implies that the orientation of nanoripples is strongly dependent on the ion beam overlap and it is possible to manipulate the orientation of nanoripples by simply changing the beam overlap.

According to the mechanism proposed by Bradley and Harper, the formation of nanoripples is attributed to the fact that the sputtering of the concave region is higher than that of the convex region creating an imbalance on the contour. Bellon and Wilson observed that even a small dose of ion beam bombardment on germanium could create large microscale explosions. Such selective sputtering and the occurrence of micro-explosions are considered to be the mechanisms behind the formation of the nanoripples. The reason for the occurrence of aligned nanoripples in the direction of scanning is due to the overlapping and propagating microscale explosions, in the direction of scan, during the FIB bombardment. As discussed in the experimental section (Sec. A in the supplementary material), for the fixed dose of 5.66 × 10^{17} ion/cm^2, the number of pixels per patterned rectangle is 80 × 80 and 23 × 23 for 0% and −250% beam overlaps, respectively. This implies that the ion dose per pixel (i.e., ion dose for each spot of FIB bombardment) is 8.84 × 10^{13} ion/cm^2 and 1.07 × 10^{15} ion/cm^2 for 0% and −250% beam overlaps, respectively. Hence, the number of ions bombarded for each spot of FIB bombardment in the case of 0% and −250% beam overlaps is 7262 and 625 ions, respectively, giving about twelve times higher ion dose per pixel for −250% beam overlap compared to 0% beam overlap. Such a higher dose, in the case of extreme negative beam overlap, leads to greater microscale explosions, and the ripple orientation is largely decided by the propagation direction of microscale explosions. This enables alignment of the larger nanoripples in the raster scanning direction.

In order to study the detailed topography of nanoripples, SEM cross-sectional imaging is performed for the case of −250% beam overlap. The germanium nanorippled surface is first coated with a protective layer of platinum about 500 nm in thickness using the FIB-CVD method and is sectioned with the aid of FIB milling for SEM imaging. The nanoripped topography thus obtained is as shown in Fig. 2(a). Measurements from the cross-sectional image yield the maximum height of nanoripples to be 290 nm, whereas the maximum ripple angle to be 60°. The average ripple angle is about 38.7°. The nanoripples orientation parallel to the scanning direction can be seen in the tilted cross-sectional SEM image in Fig. 2(c).
The nanorippled surface is then tested for its optical performance. The reflection measurements are carried out in the wavelength range of 350-900 nm. The obtained absorption spectra (100% - Reflection%) are plotted for the germanium surface with and without nanoripples in Figs. 3(a) and 3(b), respectively. The optical absorbance of the nanorippled area is 90%-95% in the wavelength range 430-720 nm. However, for the bare germanium, the light absorption is around 45% in the 400–900 nm wavelength range. It is worth noting here that the thickness of the germanium wafer used in the current study is 400 µm which is more than 400 times the maximum wavelength (900 nm). Though the absorbance is enhanced for nanorippled germanium, the bulk-germanium thickness does not influence the spectral behavior.

Electromagnetic field distribution throughout the nanostructures is simulated by the finite-difference time-domain (FDTD) method using a commercially available numerical solver (Lumerical, Inc., Canada). The nanoripples for ~250% beam overlap were modeled in FDTD simulations by generating three-dimensional (3D) topography [Fig. 2(b)] from the SEM image of the fabricated surface as described in the supplementary material (Sec. B). Optical simulation of the typical surface represented in Fig. 2(b) is carried out in the wavelength range 350–900 nm using the normally incident plane wave source. The spatial domain of the simulation region is discretized with a fine mesh of 5 nm. The periodic boundary conditions (PBCs) are set along the \( xy \)-plane, and perfectly matched layer (PML) boundary conditions are set along the propagation direction. Bulk germanium is used as a substrate for the nanorippled layer. The wavelength-dependent complex dielectric constants for germanium were taken from Ref. 22 for the wavelength range 350-900 nm.

The absorption spectra obtained from the FDTD simulations for the nanorippled and bare germanium surfaces are shown in Figs. 3(a) and 3(b), respectively. Higher absorbance of 95%-97% in the visible range for the nanoripples is close to the reported values for germanium nanopyramid arrays. As seen in Figs. 3(a) and 3(b), the experimentally measured absorption spectra for the nanorippled and bare germanium co-relate well with the simulated results. A comparison with morphologically similar silicon nano-wrinkles reveals that the measured absorbance for the nanorippled germanium
surface is 15%-20% higher than the measured absorptance for silicon nano-wrinkles. The simulated absorptance by nanoripped germanium is 20%-30% higher than the absorptance of silicon nano-wrinkles.\textsuperscript{5} The work on germanium nanopyramid arrays by Han \textit{et al.}\textsuperscript{1} reported high absorption around above 98% in the wavelength range 500–800 nm for the nanopyramid arrays. However, the absorption for the wavelengths lower than 500 nm has not been reported.\textsuperscript{1} The absorptance for the nanoripples presented in this work is above 90% in the broadband wavelength range of 350–900 nm.

When the light is incident on vertical nanostructures, the structure scatters the light at multiple instances, which increases the probability of photon absorption as depicted schematically in the inset of Fig. 3(c). The observed enhanced-light trapping could be attributed to the occurrence of multiple instances of light-scattering for the nonporous germanium surface.\textsuperscript{2} It is known that the bare germanium has a high refractive index (e.g., 5.9 at 600 nm wavelength). Thus, a high index contrast is attained at the interface between a free space ($n_{\text{air}} = 1$) and the bare germanium surface. Such a high refractive index contrast at the interface of results in high reflectance for the incident light. However, with the nanostructured germanium surface, a suppressed reflectance is observed. The suppressed reflectance can be explained by considering the graded refractive index along the thickness. It is noted that these nanostructures are narrow at the top and broad at the base. Such geometrical variation provides a graded refractive index along the height and leads to an averaged effective refractive index for the nanorippled layer of germanium. Estimation of the graded refractive index could be helpful in explaining the enhanced absorption in the nanorippled structures. The obtained nanoripples are quasi-random, and there are irregular pores in between adjacent nanoripples. Quantification of average porosity of the structure will enable calculation of the effective refractive index of the structure by utilizing Yoldas’s mixing rule for porous material.\textsuperscript{21} Yoldas’s mixing rule for porous material having air in pores is as given by

$$n_{\text{eff}} = ((n_2^2 - 1)(1 - 0.01P) + 1)^{1/2}, \quad (1)$$

where $n_{\text{eff}}$ is the effective (average) refractive index of structure, $P$ is the percentage porosity ($P\%$) of the structure, and $n_2$ is the refractive index of bulk germanium. The nanorippled structure is considered as a multilayer material with the total height of 290 nm. Each layer of the nanorippled region is set to 5 nm thickness. Subsequently, the porosity at each layer is calculated analytically as described in the supplementary information (Sec. B, Fig. S1 of the supplementary material) and plotted in Fig. 3(c). It is observed from Fig. 3(c) that for wavelength 600 nm the effective refractive index, calculated using Eq. (1), increases while going from top to bottom as the porosity decreases gradually. This provides a positive gradient of the effective refractive index for the incoming light entering from free space to the germanium substrate.

Once the effective refractive indices of nanoporous material for the entire wavelength range are obtained, the reflectance can be calculated for the nanoporous material\textsuperscript{24} using the transfer-matrix method\textsuperscript{25–27} (see Sec. C in the supplementary material). The theoretical absorption spectrum (100\% − Reflection\% − Transmission\%) thus obtained is plotted in Fig. 3(a). The theoretical absorptance is around 90\% from 350 to 700 nm wavelength range. However, the absorptance gradually decreases in the wavelength range 700–800 nm and reaches 80\% value for 800–900 nm wavelength range [see Fig. 3(a)]. The graded refractive index from top to bottom of the nanorippled surface provides a smooth variation of the index from free space to the substrate, and the penetration depth increases for the incoming light. With germanium being lossy media, the increased light penetration depth provides more chance of absorption of the light, and hence the reflectance gets suppressed. The phenomenon of the graded refractive index is wavelength independent for the wavelength range under consideration. Thus, the multilayer model validates the anti-reflection behavior which is also shown by the experimental measurements and the FDTD simulations.

We have also investigated the effect of incidence angle and thickness of the nanorippled layer using FDTD simulations, as described in the supplementary information (Sec. D of the supplementary material). It is clear from Fig. S3 (in the supplementary material) that the absorptance increases upto 15\(^{\circ}\) incidence angle and then decreases for further increase in the incidence angle. Thus, the acceptable incidence angle for the enhanced absorptance for the nanoripples structures is about 50\(^{\circ}\). The results suggest that the nanoripples exhibit wide angle broadband absorption enhancement. The wide angle (upto 50\(^{\circ}\)) absorptance behavior of the germanium nanoripples is attributed to the smooth refractive
index gradient for the light coming from free space to the nanoripped surface.\textsuperscript{28,29} For the incidence angle below 50\degree, the smooth refractive index gradient ensures gradual bending of light beam toward the surface normal resulting in multiple internal reflections leading to suppressed reflection.\textsuperscript{28,29} While for the glancing angle incidence, the increased scattering leads to low absorption. It is seen from Fig. S4 (in the supplementary material) that the absorptance is increased for the higher thickness of the nanoripped layer. However, with the FIB processing, it is difficult to get the nanoripples with the higher aspect ratios. In order to increase the depth of nanoripples, the ion dose and dwell time of the Ga beam need to be increased. The increased dose leads to sputtering of the material instead of nanoripples formation.

We have further investigated the field distribution in the nanostructures using FDTD simulations. The electric field intensity (|\(E|^{2}\)) distribution in the \(xz\)-plane (\(y = 0\) nm), \(yz\)-plane (\(x = 0\) nm), and \(xy\)-plane (near field at 1 nm above the nanoripple surface) is shown in Fig. 4. The electric field intensity (|\(E|^{2}\)) is plotted for 350, 500, and 900 nm wavelengths [see Figs. 4(a)–(c)]. Bright hot spots of the electric field intensity indicate that the electric field is trapped at positions such as on the tip of ripples and the gap between them [see Figs. 4(a)–(c)]. It should also be noted that the light trapping is wavelength independent and in addition to the graded effective refractive index, the enhanced absorption is also attributed to the enhanced light trapping. Further, optical power absorption per unit volume is calculated by measuring the spatial absorption for each volume element in the simulation domain at different wavelengths. The optical absorption density for the \(xz\)-plane is as shown in Fig. 5 for the nanoripples and bare germanium. The shorter wavelengths, 350 nm and 500 nm, are absorbed at the surface of bare germanium [see Figs. 5(a) and 5(b)]. However, for the nanoripped surface, most of the shorter wavelengths are absorbed at the tip of the nanoripples and rest of it gets absorbed inside the nanoripples. In the case of longer wavelengths (900 nm), after a small amount of power absorption at the tip of the nanoripples, considerable amount of light gets absorbed inside the nanoripples and the remaining passes to the bulk of the substrate getting absorbed up to 200 nm depth [see Fig. 5(c)], whereas for the bulk germanium only some part of longer wavelength (900 nm) light gets absorbed up to the depth of 490 nm. Hence, the enhanced absorption due to the nanoripped surface is wavelength independent.

In summary, we demonstrate the broadband light trapping for the FIB induced germanium nanoripples both experimentally and through FDTD simulations. The resultant light absorptance is about 95\% for the visible light range. Such broadband light absorption in nanostructured germanium can lead to a higher efficiency in the photovoltaic based devices.

FIG. 4. Two-dimensional (2D) electric field intensity (|\(E|^{2}\)) distribution at wavelengths of (a) 350 nm, (b) 500 nm, and (c) 900 nm for nanoripples across planes \(xz\)-plane (\(y = 0\)), \(yz\)-plane (\(x = 0\)), and \(xy\)-plane (1 nm above nanoripples). The dashed white line indicates the germanium surface profile, and the color bar shows electric field intensity (|\(E|^{2}\)).
with variant orientations such as parallel and perpendicular to the ion beam scanning direction is demonstrated. The results provide a novel approach to manipulate the orientation of nanoripples by simply changing the beam overlap and thus enabling the fabrication of varied nanoripples on a defined area. Using this approach, different nanoripple orientations can be achieved at the normal incidence of the focused ion beam itself, which is advantageous over the conventional approach of varying incident angle. The present study has potential to open new avenues for controlling the orientation of nanoripples that can aid in the design of high-efficiency germanium-based photovoltaic devices.

See supplementary material for detailed description on the (A) experimental process, (B) three-dimensional topography and porosity calculations for the multilayered nanorippled surface, (C) theoretical absorption calculations from porosity using the transfer matrix method, and (D) FDTD simulations for the effect of the incidence angle and nanorippled layer thickness on the absorption.

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