Polarization dependent structural colors from tilted metalo-dielectric nanopillars

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Abstract

The structural color from self-assembled metalo-dielectric (In/InP) nanopillars is shown to be polarization sensitive when the axial symmetry is broken. The characteristic dip in the reflection spectra due to resonant absorption is shifted by 90 nm as the polarization of incident light is altered from TE to TM at an incidence angle of 40°. We also show wafer-scale, mask-less fabrication of pillars that are tilted with respect to the substrate, a fast and cost effective method of creating the asymmetrical structures required for polarization sensitivity at normal incidence. A dip shift of 100 nm is observed for 40° tilted nanopillars of average height 380 nm, resulting in a smooth range of colors with changing polarization. FDTD simulations confirm the polarization dependent dip-shift in the resonant absorption wavelength. Furthermore, the field and intensity profiles obtained from the simulations indicate that the resonant absorption dips are due to HE1m-like modal excitations and their shift with respect to the incident angle and polarization leads to the change in perceived color from the tilted nanopillar system.

1. Introduction

Modifying the morphology, spatial arrangement and material composition of nanostructures enables engineering of optical functions such as subwavelength light localization [1–4], anti-reflection [5, 6], photodetection [7], harmonic generation [8], bio sensing [9–11], and structural colors [12–14]. In the last decade, structural color generation using nanostructures has gained interest as a novel technological solution to high-resolution color printing, displays, filters and camouflages [15–18]. One way to realize structural colors from nanostructures is through size dependent resonant optical phenomena such as Mie- and localized surface plasmon resonances (LSPR) [13, 18–20]. Such wavelength specific resonances are highly sensitive to the geometry of the structures interacting with the light. By breaking the spatial symmetry of the structures it is therefore possible to tune the optical response with the polarization state of the incoming light.

T Ellenbogen et al demonstrated polarization sensitive tunable color filters made from arrays of metallic nano-cross structures with varying arm lengths [21]. Each arm supports LSP resonances at frequencies determined by the arm’s length. Therefore, polarization sensitive excitation is possible as each arm responds to fields oscillating parallel to the arm’s axis. At the LSP resonance, a dip in the transmission spectra is observed due to increased absorption/scattering and the dip position shifts with changing polarization. Similar cross structures have been fabricated using dielectric materials, for example silicon, where Mie resonances are utilized to avoid the high losses of plasmonic structures [22]. Recently it has been shown that similar effects can be achieved with Al/SiO2 gratings [23], which rely on a combination of surface plasmon and guided mode resonances. Electrically tunable and polarization sensitive transmission color filters have been demonstrated by utilizing asymmetric lattice nanohole array on Al thin films [24]. The nanostructures discussed above are realized through traditional fabrication techniques such as patterning, development, deposition, lift off and
etching. These fabrication techniques are time consuming, limited to small areas (mm²) and comparatively expensive. In this work we show a maskless, single-step wafer scale method of fabricating InP nanopillars with controllable tilt with respect to the substrate that exhibit polarization sensitive structural colors.

2. Experimental

The nanopillars are created through a self-masked process under low energy nitrogen ion beam sputtering (IBS) of plain InP. Spontaneous pillar formation in binary III–V materials under IBS is generally understood to be due to a difference in sputter yield between the two species [25]. In the case of InP, preferential etching of phosphorous creates an In rich surface. The excess In atoms diffuse on the surface and congregate in clusters to minimize surface energy. The In rich clusters act as etch masks which are continually supplied with new In from within a diffusion length as sputtering continues. The spatial arrangement of the pillars is found to be disordered with a characteristic inter-pillar distance of two diameters (see figure S1 (available online at stacks.iop.org/MRX/8/046202/mmedia)) [26]. The elemental analysis on the InP/InP pillars has shown that the formed pillar is In rich at the top and consists of a crystalline InP stem covered with amorphous In-InP jacket [25]. The height and radius of the pillars are controlled through sputtering duration, though not independently, a longer pillar has a larger radius. For certain heights (200–600 nm) the nanopillar-arrays display vivid colors. Moreover, as IBS is highly directional it is possible to fabricate tilted pillars by positioning the substrate at an angle with respect to the incoming ion beam. The details of the fabrication of self-assembled nanopillars using IBS are provided in our earlier work [25]. Cross-sectional SEM images of the fabricated tilted pillars of height ~350 nm are presented in figure 1(a), where we see a gradual shift in the pillar orientation. Also shown, in figure 1(b), is the difference in color of reflected light from the samples of different pillar tilts when the analyzed light is filtered through a linear polarizer, for two polarizations with a 90° change. A photo of the full samples can be found in the supplementary information, figure S2.

3. Results and discussion

Polarization sensitivity was first observed from the upright pillars in off normal incidence reflectance measurements. The measurements were performed on upright pillars with an average height of 340 nm, average diameter of 100 nm and with a nearest neighbor distance of 100–200 nm (figure S1). The polarization- and incidence angles, φ and θ respectively, are defined in figure 2(a). All the reflectance measurements were performed using a Lambda 900 UV–vis-NIR spectrometer with an integrating sphere. To avoid reflected light leaking out of the entrance aperture of the sphere, the minimum incidence angle is set to 8°. The off-normal incidence angle measurements were performed by placing samples on a central rotation mount inside the integrating sphere. The total reflectance at θ = 8° (figure 2(b)) has a characteristic dip at ~470 nm. As expected, due to the symmetry of the pillar with respect to the axis of incidence, there is little difference in the spectra for the two polarizations (φ = 0° and 90°). In contrast, at an incidence angle of 40°, the position of the reflectance dip for φ = 90° is blue shifted and red shifted for φ = 0° w.r.t to the dip position for θ = 8°, with ~90 nm of overall difference in the dip positions. Furthermore, as shown in figure 2(c), it is possible to tune spectral dip position with increasing φ between the two polarization extremes. The variation in the spectral position of the reflectance dip alters the perceived color of the sample and is represented on the standard CIE 1931 chromaticity diagram in figure 2(d). The color changes from purple to orange while rotating from φ = 0° to φ = 90°, where all colors that are a mix of the two extremes can be achieved by intermediate polarizations. Figure 2(e) shows reflectance as a function of polarization angle at three wavelengths selected to demonstrate the variation in polarization response. At 380 nm the reflectance decreases with increasing φ, while at 500 and 620 nm it increases. In our previous study [26], we reported the shifting of the absorption dips with changing pillar height. However, increasing pillar height not only shifts the dip position but also reduces the specular reflectance. This is a result of finite absorption occurring in the pillars for all the incident wavelengths corresponding to energies above the bandgap, including the resonant absorption. By introducing polarization as a tunable parameter one can vary colors without reducing the surface reflectance.

Tilted nanopillars have been explored to demonstrate novel optical properties which otherwise are not observed in upright pillars. Metasurfaces consisting of self-assembled tilted gold nanopillars on silicon, have been demonstrated to show circular dichroism property in both linear and non-linear (SHG) optical regime [27]. Tilted silicon pillars have been reported to show anti-reflection properties that lead to a higher absorption efficiency compared to upright pillars [28]. Therefore, it is interesting to investigate tilted pillars for unique optical properties, especially in the structures whose resonances are sensitive to the pillar geometries and the angle of incidence of light. Due to the tilt, the symmetry of the pillars with respect to the axis normal to the substrate is broken and the pillars show polarization sensitive response to the normally incident light. A major
benefit of using tilted pillars attached to the substrate is that all pillars are oriented in the same direction, which is difficult for other pillar/wire systems such as grown nanowires where the random tilt directions evens out polarization sensitive signals. For demonstration of polarization sensitivity at normal incidence we use pillars tilted at an angle of 40°, seen in the bottom SEM image of figure 1(a). The average height along the pillar axis and diameters of the tilted pillars are 380 and 100 nm, respectively, and are comparable with the upright pillar dimensions. For the tilted pillars, the reflectance measurements were conducted at an 8° incidence, as shown in the schematic of figure 3(a). The reflectance spectra of the tilted pillars (figure 3(b)) show a similar shift in dip position with polarization angle as the upright pillars at off normal incidence angle, the dip is red shifted for light polarized in plane with the tilt (TM) and blue shifted for out of plane polarization (TE). The reflectance response with increasing polarization angle varies across the visible spectrum, illustrated by the three wavelengths seen in figure 3(c). At a wavelength of 420 nm the reflectance decreases with increasing φ, while at 500 nm the response is flat and at 620 nm it increases. Furthermore, the degree of reflection at the two dips can be mixed through intermediate polarizations to obtain colors in a straight line between the extremes, seen in the chromaticity diagram in figure 3(d). The color span attainable with tilted pillars is wider than that of the upright pillars. The color difference between the two polarization extremes is clearly visible in the bright field images (figure 3(e)) of the sample obtained from an optical microscope. The shift in color, purple to yellow, can be seen with the naked eye (figures S2, SI) for the tilted pillars as they do not require a specific incidence angle to show polarization sensitivity. Furthermore, the color span increases with tilt angle (figures S2, SI), similar to the behavior seen with increased incidence angle for upright pillars.

The dips in the reflectance at normal incidence for upright pillars is understood to originate from size dependent resonant absorption within the individual a-InP pillars [26]. The pillars have a high refractive index

**Figure 1.** (a) Cross-sectional SEM images of In/InP pillars with tilt angles from 0° to 40°. The pillars have an on axis average height of 340 nm for upright and 380 nm for 40° tilt, with a diameter of 100 nm. All samples have been fabricated with the same etch time but with varying sample tilts with respect to the ion beam, as indicated by the degrees in the SEM images. Scale bars in each image represent 200 nm. (b) The shiny color reflected from the samples for TM, φ = 0°, and TE, φ = 90°, polarized light. The pillar tilt increases in correspondence with the SEM images in (a). The images were taken with a digital camera with a linear polarizer in front of the objective. Each image is ~3 × 3 mm in size, full samples are ~5 × 5 mm and can be seen in figure S2 in the supplementary information.
and a cylindrical shape and thus act as waveguides which support guided and leaky modes. The presence of a high-index substrate might also couple the light into the substrate. As InP is absorptive in the visible spectrum, modes confined in the interior of the pillar are absorbed. Resonant absorption of guided/leaky modes at normal incidence has previously been shown in finite semiconductor pillars, where the spectral location of the absorption dip(s) is tuned by changing the diameter of the pillar [29–31]. Furthermore, the spectral position of the mode can be tuned with the incidence angle [32, 33]. To confirm and explore the mechanism behind the dip shift and polarization dependence in our pillar system, we perform electromagnetic simulations using the commercial FDTD solver Lumerical [34].

Figure 2. The effect of incidence and polarization angle of light on the reflectance spectra from upright In/InP pillars with an average height of 340 nm and diameter of 100 nm. (a) Definition of incidence- (θ) and polarization angle (ϕ), dashed lines indicate 0° in respective case. Also defined is the coordinate system that is used throughout the article. (b) Experimentally measured spectral reflectance for two different incidence angles and polarization configurations. (c) Gradual spectral blue shift (500 nm to 400 nm) of the dip position with varying polarization angle from ϕ = 0° to 90°, at θ = 40°. (d) Corresponding chromaticity diagram for the spectra in (c), showing a linear relationship between the generated color and the polarization. Also marked are the corners of the sRGB triangle. (e) Change in reflectance with polarization angle at three selected wavelengths.
The upright pillars are modelled as In balls placed on top of a InP cylinders that are standing on an InP substrate, as shown in figure 4(a). The total height of the pillar is 350 nm with a diameter of 100 nm. An In sphere of diameter 100 nm is on top of the pillar. The permittivity values for In, InP and a-InP were obtained from Adachi database \[35\]. Pillar dimensions were obtained through cross sectional SEM images and average closest neighbor distance was calculated by mapping the radial distribution function from positions collected by top view SEM images (figure S1). Pillars were placed in a square periodic lattice to reduce computational load, as we have previously shown that the positional ordering has effect only on the diffuse component of the reflectance, not on the resonant absorption dip \[26\]. Periodic boundary conditions were used in the lateral directions (x, y) with a period of 200 nm, while the vertical boundaries were capped with perfectly matching layers. A broadband
fixed anglesource (350–800 nm) was injected with an angle $\theta$ off of the $z$ axis, towards the substrate plane. A cubic mesh element with 2 nm side was used in the pillar and its boundaries. A frequency domain power monitor placed above the source measured the reflectance, while frequency domain field profiles were used to record the electric field in the $x$, $y$ plane and the $z$ locations specified in figure 4(c). Reflectance spectra obtained from simulations on upright pillars with $\theta = 40^\circ$, shows the gradual red shift in the absorption dip position from 400 to 500 nm with changing polarization angle from 0° to 90°. Grey dashed line represents reflectance at normal incidence. (c) $|E_x|$ intensity of the upright pillar with $\theta = 40^\circ$ and $\phi = 0^\circ$. (d)–(f), (g)–(i) Cross-section contour maps of the electric field intensity $|E_x|$, $|E_y|$ at different heights of the pillar for the specified incidence- and polarization angles, at the respective reflectance dip position. (g)–(i) Power absorbed per unit volume in the pillar at height $z_2$ for the three cases considered (c).

Figure 4. FDTD simulations on upright pillars. (a) Schematic of the simulation set-up. The pillar consists of a 300 nm high a-InP cylinder with an In ball of radius 50 nm placed on top of InP substrate. The total height of the pillar is 350 nm. (b) Reflectance spectra obtained from simulations on upright pillars with $\theta = 40^\circ$, shows the gradual red shift in the absorption dip position from 400 to 500 nm with changing polarization angle from 0° to 90°. Grey dashed line represents reflectance at normal incidence. (c) $|E_x|$ intensity of the upright pillar with $\theta = 40^\circ$ and $\phi = 0^\circ$. (d)–(f), (g)–(i) Cross-section contour maps of the electric field intensity $|E_x|$, $|E_y|$ at different heights of the pillar for the specified incidence- and polarization angles, at the respective reflectance dip position. (g)–(i) Power absorbed per unit volume in the pillar at height $z_2$ for the three cases considered (c).
pillar is calculated from the relation $P_{\text{abs}} = 0.5 \omega \varepsilon'^0 |E|^2$, where, $\omega$ is the angular frequency of the incident light, $\varepsilon'^0$ is the imaginary part of the permittivity of the material and $|E|^2$ is the electric field intensity. The absorption profiles were obtained using the built-in analysis tool of the Lumerical software. The absorption and intensity profiles (and vector plots of $E_{x,y}$, figure S3, SI) at the central dip position ($460\text{ nm}$) for normal incidence (figures 4(d), (g), (j)), corresponds remarkably well with the mode profile of the HE$_{11}$ mode, considering the short height of the pillars [31, 32, 36]. The HE$_{11}$-like mode, seen in figures 4(e), (h), (k), red shifts to $300\text{ nm}$ at $\theta = 40^\circ$ and $\phi = 0^\circ$, as predicted by analytic mode calculations of an infinitely long InP pillar of comparable radius in ref [33]. Furthermore, at $\theta = 40^\circ$ and for the TE polarization ($\phi = 90^\circ$) a resonant mode is excited at $400\text{ nm}$ resulting in a reflectance dip. The field and absorption profiles at $400\text{ nm}$, seen in figures 4(f), (i), (l), are similar to the profiles for $\theta = 0^\circ$, except for the polarization direction, indicating a possibility of another HE$_{1m}$-like mode. Furthermore, the vector plot of the profile (figure S3(b), SI) is indeed similar to the HE$_{12}$ mode for infinitely long cylinders [37]. It should be further noted that at higher angles we expect the leaky mode field profiles to resemble the Mie resonances, as it has been shown that Mie resonances and leaky modes are closely related at higher angles [32, 33].

4. Conclusion

We have demonstrated a polarization dependent coloration from In/a-InP nanopillars fabricated in a wafer scale, single step ion beam sputtering process. The upright pillars show polarization sensitive reflectance at off-normal incidence angle due to the breaking of the symmetry of the pillar as seen by the incident light. At an angle of incidence of $40^\circ$, the difference between the dip positions in the reflectance for the in and out of plane polarizations are ~$90\text{ nm}$.

Placing an InP substrate at an angle with respect to the direction of ion beam results in tilted nanopillars that exhibit polarization sensitive reflective colors at normal incidence of light. In the case of tilted pillars, at an angle of incidence of $40^\circ$, the polarization sensitive reflective colors range from purple to yellow and span a larger region in the chromaticity plot compared to the colors from the upright pillars (with off-normal incidence). The perceived color of the tilted pillar system changes with an overall shift in dip position of ~$100\text{ nm}$. FDTD simulations show that the polarization sensitivity stems from resonant absorption of HE$_{1m}$-like modes and the color response can be tuned gradually between the two extremes.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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Disclosures

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