Reversible tuning of photonic crystal cavities using photochromic thin films

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We demonstrate reversible tuning of a photonic crystal cavity resonance using a thin photochromic film composed of spiropyran and polymethylmethacrylate that serves as a photosensitive cladding layer. Exposure of spiropyran to ultraviolet light results in smooth redshift of the cavity resonance that can be reversed by exposure to visible wavelength light. We achieve a reversible resonance shift of up to 2.7 nm, which can be performed locally on individual cavities. The resonance shift over multiple successive UV and visible light exposures is studied to determine the repeatability of the photochromic film. © 2010 American Institute of Physics. [doi:10.1063/1.3377910]

Integrated nanophotonic structures play an important role in development of high-speed and low-power optical devices. One of the most promising methods integrating optical components uses photonic crystal structures.1 Photonic crystals enable the development of high-Q cavities2–4 and low-loss waveguides5 that can be combined to form complex integrated structures on a semiconductor chip.5,6 These integrated structures are finding applications as efficient filters,7 optical logic components,8,9 and quantum optical devices10,11 on top of the device. SPs are a class of photochromic compounds whose absorption in the visible wavelengths can be modified reversibly through irradiation with UV and visible light.19 This change in absorption occurs because SPs can exist in two stable forms, spiropyran (SC), and merocyanine (MC). By irradiation with UV or visible light, SPs can be transformed reversibly between the colorless SC and colored MC forms. Accompanying the change in the visible absorption of the SP layer on top of the cavity, is a change in refractive index at near infrared (NIR) wavelengths, causing a shift in the cavity resonance. At the same time, SPs are highly transparent in the NIR wavelengths and illumination with NIR light has little effect on its absorption. These properties make SP an ideal photochromic material for gallium arsenide (GaAs) based photonic crystal devices.

The photochromic material used in our experiments consisted of 5 wt % 1,3,3-Trimethylindolino(naphthospirooxazin (TCI America) and 0.5 wt % 950 PMMA A4 dissolved in anisol. This particular SP was chosen because it was found to operate well at cryogenic temperatures, in contrast to other types of SP.20 The specific concentration of 5% SP was the maximum amount that could be dissolved in anisol, and was therefore selected in order to deliver the highest concentration of photochromic material to the sample. Adding a small amount of PMMA was found to provide a more uniform coat of the SP when spun on the sample surface.

Another method for tuning photonic crystals is to coat them with a photosensitive cladding layer, such as chalcogenide glass, whose index of refraction can be optically modified. Chalcogenide glass films have been used to tune both photonic crystal cavities16 and quantum cascade lasers17 by local optical excitation that induces a shift in the index of refraction of the cladding. However, for chalcogenide glass cladding this shift is irreversible, so once the cavity is shifted there is no way to undo the change.

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Here we introduce a technique for tuning of photonic crystal structures which is both local and reversible. This capability could be used for development of reconfigurable photonic devices that can be reversibly modified postfabrication. Reversible local tuning is achieved by spin coating a thin layer of photochromic material composed of a mixture of a spiropyran (SP) and polymethylmethacrylate (PMMA) on top of the device. SPs are a class of photochromic compounds whose absorption in the visible wavelengths can be modified reversibly through irradiation with UV and visible light.19 This change in absorption occurs because SPs can exist in two stable forms, spiropyran (SC), and merocyanine (MC). By irradiation with UV or visible light, SPs can be transformed reversibly between the colorless SC and colored MC forms. Accompanying the change in the visible absorption of the SP layer on top of the cavity, is a change in refractive index at near infrared (NIR) wavelengths, causing a shift in the cavity resonance. At the same time, SPs are highly transparent in the NIR wavelengths and illumination with NIR light has little effect on its absorption. These properties make SP an ideal photochromic material for gallium arsenide (GaAs) based photonic crystal devices.

In our experiments we first characterized absorption properties of a photochromic film spun on a glass slide at 500 rpm using a spectrophotometer. Figure 1 shows the results of the spectrophotometer absorption measurement from 400 to 1000 nm. The photochromic film right after spin coating, shown as a thick solid line, features little absorption indicating that the SP is initially in its SC form. The film is then exposed to UV light from a mercury xenon lamp with an intensity of 3 W/m² for 10 min. After UV exposure the absorption increases significantly in the wavelength range of 550 to 650 nm, as shown in the solid black curve, indicating that a sig-

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significant fraction of SP has been converted to its MC form. Next, the sample was illuminated with 532 nm light picked off from a frequency doubled neodymium-doped yttrium aluminium garnet (Nd:YAG) pump laser, for 10 min at an intensity of 12 W/m². The absorption was once again measured (solid light gray line) and found to return to near its original starting point, showing the reversibility of the process. When the film is not irradiated by UV or visible light, it is stable and does not degrade. The absorption in the 900 to 1000 nm region is small in both forms of SP. Thus, for cavities that are resonant at NIR wavelengths we can use the SP film to shift the cavity resonance without degrading the cavity $Q$.

After initial illumination with UV and green light, the procedure was repeated two more times on the same sample, as indicated by the dotted and dashed lines. It can be seen that the reversibility of the material degradations as it is cycled repeatedly between SC and MC forms. Although this degradation puts a limit on the number of times a film can be reversed, the film can be easily washed away with acetone and a new film deposited. We further characterized the refractive index of the photochromic film in its initial state to be 1.5 using an n-k analyzer, which estimates the refractive index of a thin film.

In order to demonstrate reversible tuning of photonic crystal structures, we spin-coated a thin film on a GaAs sample containing photonic crystal cavities. Figure 2(a) shows a scanning electron microscope (SEM) image of the photonic crystal cavity fabricated for the cavity shift experiments. The initial sample is composed of a 160 nm thick GaAs grown on a 500 nm thick sacrificial layer of Al$_{0.5}$Ga$_{0.5}$As. A single layer of InAs QDs of density 100–150 QDs/μm$^2$ was grown at the center of the GaAs slab. The two-dimensional photonic crystal with triangular lattice structure was patterned onto the sample using electron beam lithography and inductively coupled plasma etching. The sacrificial layer was then undercut by wet etching to create the photonic crystal membrane. We used an L3 cavity design formed by removing three holes from the photonic crystal lattice composed of 72 nm holes with a 230 nm lattice constant. The device was cooled to 4 K and photoluminescence (PL) measurements were performed using a confocal microscope.

To determine the appropriate SP thickness, the film was spun on the same sample at three different spin rates of 4000, 3300, and 2500 rpm. The film thicknesses corresponding to these spin rates, determined by independent SEM cross-sectional measurements, are 35 nm, 55 nm, and 100 nm, respectively. To obtain spectra of the cavity, the sample is mounted in a cryostat cooled to 4 K and illuminated at 780 nm from a Ti:sapphire laser using a 0.75 numerical aperture objective. The PL from the cavity is then captured using the same objective and focused onto the charge coupled device of the spectrometer for spectroscopy measurements. Figure 2(b) shows the measured PL cavity spectrum for these three different film thicknesses along with the spectrum of the uncoated cavity. The uncoated cavity has a center wavelength of 909.1 nm and a measured $Q$ of 5060. The InAs QDs act as a bright internal source, enabling us to measure the cavity resonance over the broadband luminescence of the QDs. After depositing the photochromic films of different thicknesses the cavity resonance was redshifted by 8.0 nm, 11.1 nm, and 16.5 nm, respectively. The cavity $Q$s measured at these three thicknesses were 4640, 4260, and 2640. For very thin films (30 nm), the effective index above the GaAs slab is still close to air so there is little effect on the cavity $Q$. As we make the film thicker the structure becomes more asymmetric and light is allowed to escape from the top more quickly, degrading the cavity $Q$. For the 100 nm film the degradation of the $Q$ was considered to be too severe, so all measurements were performed using either 35 or 55 nm films.

To locally shift a single photonic crystal on the sample, we placed the sample in the cryostat cooled to 4 K and illuminated it with either 375 nm light from a doubled Ti:sapphire laser or 532 nm light from a doubled Nd:YAG. The UV spot size was approximately 5 μm$^2$ in diameter, corresponding to a UV intensity of 1.2 W/cm². The green light was also focused to a spot size of 5 μm$^2$, corresponding to an intensity of 24 W/cm². Exposure of the sample surface created a visible change in the SP film that was localized to the illuminated spot. The intensity of visible light was chosen to be much higher than that needed for UV because the reversibility of SP degrades at cryogenic temperatures.

We initially illuminated with UV to convert the SP to its colorless SC form to its MC form. Figure 3(a) shows a series of PL spectra of the cavity after twelve 10 s exposures using UV light for 55 nm thick film. Each exposure created a redshift of approximately 0.25 nm. After twelve exposures a maximum shift of 2.7 nm was achieved. We note that no detectable change of cavity $Q$ was observed throughout the entire tuning range. Following UV exposure a set of exposures with the green laser was then used to convert the SP back to its SC form to reverse the tuning. The cavity was shifted back to near its original resonance but a residual shift of 0.5 nm remained. This residual shift is due to the fact that after green illumination the SP does not fully return to its original absorption spectrum. A similar experiment was carried out for the 35 nm film thickness which was found to
have nearly identical behavior but achieved only a 2 nm maximum cavity shift.

To explore the reversibility of the shift, we increased the UV intensity to 3.84 W/cm² and the green intensity to 76.8 W/cm². These intensities are sufficient to achieve the maximum shift with a single 10 s exposure. Figure 3(b) plots the cavity resonance after successive UV and green exposures for both the 35 and 55 nm thick coatings. The cavity can be shifted and reversed roughly three times before the tunability degrades too much for further significant shifts.

In conclusion, we have demonstrated a method for reversible tuning of photonic crystal cavity resonances using SP photochromic thin films. The film enabled reversible tunings as large as 2.7 nm by successive exposures of UV and visible wavelength light with little degradation to the cavity Q. The performance of the film could be improved by incorporating photochromic materials that exhibit greater index changes and reversibility. The photochromic films demonstrated in this work could add significant flexibility in nanophotonic design enabling reconfigurable optical chips with tunable components that be adjusted postfabrication. With the film, we can also tune a cavity in and out of resonance with a narrowband light source such as a single QD. This work can also be extended to other material systems like PbS QDs on AlGaAs membranes that have the potential to perform at room temperature. This work was supported by an Army Research Office Young Investigator Award (Grant No. W911NF0710427), NSF CAREER award, and the NSF Physics Frontier Center at Joint Quantum Institute (JQI@PFC). The authors would like to thank Rakesh Kumar for his helpful assistance.