Flexible photonic metastructures for tunable coloration: supplementary material

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Published 12 March 2015

This document provides supplementary information to “Flexible photonic metastructures for tunable coloration,”

<http://dx.doi.org/10.1364/optica.2.000255>. © 2015 Optical Society of America

<http://dx.doi.org/10.1364/optica.2.000255.s001>

S. 1. Fabrication Process

The fabrication process for the flexible metastructure consists of two parts. The first part is the fabrication of the HCM on a rigid substrate with nanofabrication techniques, as illustrated in Fig. S1(a). The process starts from an SOI substrate purchased from SOITEC, with a 220 nm thick silicon device layer and 3 μm thick buried oxide (BOX) SiO_2 layer. In order to achieve precise control of the HCM thickness, thermal oxidation is used to thin down the device layer thickness to the target value. For a 180 nm target thickness, dry oxidation is performed at 1000 $^{\circ}\text{C}$ for 2 hours and 40 minutes providing a target accuracy within 5 nm. The resulting ~ 90 nm thick thermal oxide doubles as a dry etching hard mask in the subsequent silicon etching step. The selectivity of SiO_2 is greater than DUV resist for silicon dry etching, and results in a steeper sidewall slope of the HCM. The DUV lithography is done with an ASML300 DUV stepper. The lithography resolution is 248 nm. The pattern on the DUV resist is then transferred to the SiO_2 hard mask by oxide dry etching. The DUV resist is stripped before the silicon etching step. A transformer coupled plasma (TCP) process is used to etch the silicon with the oxide hard mask. In the last step, the HCM structure is released with HF vapor. An etching rate of ~ 30 nm/min keeps the HCM suspended with support from folded beams. The release time is controlled to ensure the HCM is released but the large frame is still connected to the substrate with the BOX.

The nano-printing transfer process is shown in Fig. S1(b). The PDMS stamp-based process is used to pick up the HCMs from the SOI substrate. The PDMS stamp is made of SYLGARD 184 silicone elastomer.

A 10:1 mixing ratio between the base and the curing agent is used in this process. The mixing ratio can be adjusted for the specific mechanical property requirement of the PDMS membrane. After mixing the base and the curing agent, air bubbles are created. They can be removed by applying a vacuum to the mixture. The stamp is then solidified by baking the mixture at 75 $^{\circ}\text{C}$ for 1 hour. To pick up the HCMs from the SOI substrate, the PDMS stamp uniformly covers the surface of the SOI chip.

Fig. S1. (a) The nano-fabrication process of the HCM on the SOI substrate (b) The nano-printing transfer process.

In order to achieve high transfer yield (minimizing defective pixels), each individual HCM pixel needs to have good adhesion with the PDMS stamp. Small air bubbles can easily form between the stamp and the SOI chip surface, especially for thin PDMS stamps, making the transfer challenging for large sample sizes. This yield issue is solved by performing a second vacuum step. The SOI chip is fixed on the glass slide with double-sided scotch tape. Next, a PDMS stamp seals the perimeter of the SOI chip with an air film isolating the PDMS stamp from the SOI chip surface. The assembly is then put into a vacuum chamber. The pressure within the air film will be much lower than atmospheric pressure. Finally, by rapidly venting the vacuum chamber, the atmospheric pressure uniformly presses the PDMS stamp to the chip surface providing

excellent contact between each individual HCM pixel and the stamp. Because the HCM pixel is connected to the substrate with very weak folded beams, peeling off the stamp from the substrate will easily break the beams without affecting the HCM structures. Finally, the HCMs on the stamp are encapsulated by a thin PDMS layer by repeating the PDMS stamp preparation process.

S. 2 Simulations for the color designs

The color palette is designed to have a -1st diffraction order enhancement effect for four colors (green 540 nm, yellow 570 nm, orange 590 nm, and red 630 nm) with the same input and output condition. A variety of simulation techniques can be used to calculate the optical spectrum, such as an analytical formulation [S1-S3], rigorous coupled wave analysis (RCWA) method [S4], finite difference time domain method (FDTD) [S5], etc. In this design process, a numerical calculation with the analytical formulation is used for its fastest speed to find the required geometries. The simulation code is run in a Matlab environment with 224-core parallel computing. The PDMS refractive index used in the calculation is $n_{PDMS} = 1.41$. The silicon index is a complex number for visible wavelengths from reference [S6]. For a given input and output angle, the geometry of the HCMs is solved to have the strongest -1st diffraction order enhancement. Fig. S2 shows the simulated R_{-1} value by varying the bar and gap width, which are defined by the photolithography. For each designed color, the dimension can be found to have R_{-1} approaching 90%.

Fig. S2. The simulated R_{-1} with varied bar and gap width for (a) green (b) yellow (c) orange (d) red designs.

The optical properties of the HCMs from structure deformation are also calculated, as shown in Fig. S3. Because the Young's modulus of silicon is magnitudes higher than PDMS, it is indeed the low index gap that is stretching when the deformation is introduced. The diffraction spectra for R_{-1} is simulated by varying the gap size. The received wavelength with the corresponding gap size under the designed angle is marked as the white line in the contour figure. For all of the color designs, R_{-1} has broad enhancement bandwidth in order to shift the colors. For example, the green color (540 nm) can shift to yellow (570 nm) by stretching the gap width from 250 nm to 278 nm while maintaining high diffraction efficiency.

Figure S3. The simulated R_{-1} spectra with the deformation of the HCMs in the PDMS membrane for (a) green (b) yellow (c) orange (d) red designs.

S. 3 Coloration property characterization

The characterization setup schematics are shown in Fig. S4. The diffraction power efficiency is measured with the setup in Fig. S4(a). The fiber coupled laser source illuminates the sample through a collimator. A free space polarizer is used to attain TE polarization. The collimator provides 0.39 mm output waist diameter and 1.75 mrad divergence angle. The diffracted light is measured with a photodetector. The collimator and the detector are fixed on the rotational arms and aligned to have the rotation center at the spot where the sample is illuminated. The reflected power from the gold mirror is measured as the reference where the reflectivity is obtained from the mirror specification. This setup is also used for the beam steering measurement. The sample can be stretched by the clamps and the deformation can be measured with micrometers. The angular response and the beam size are measured by the photodetector with a pin hole.

The diffraction spectra of the HCMs are measured with white light illumination, as shown in Fig. S4(b). The light is illuminated and received by the collimator with 0.8 mm output beam diameter and 0.049° divergence angle. The difference wavelengths in -1st order are measured by adjusting the angle of the receiving arm. The spectrum is measured by the Ocean Optics HR4000 visible light spectrometer. By performing the integral of the spectrum over the spatial angle for each order, the diffraction spectrum as shown in Fig. 3(e) are measured. The dispersion of the measurement system is ruled out by using the gold mirror reflection measurement as reference.

Fig. S4. (a) The setup schematic for the diffraction efficiency measurement with the laser source. (b) The setup schematic for the diffraction spectrum measurement with the white light source and the spectrometer.

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