Experimental verification of the applicability of the homogenization approximation to rough one-dimensional photonic crystals using a holographically fabricated reflection grating

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The theoretical reflectance spectrum of a one-dimensional photonic crystal with large amounts of interfacial roughness has been calculated using a previously proposed method, and compared to the actual experimental reflectivity of the structure. The photonic crystal was fabricated using a simple and fast method involving the holographic exposure of a liquid crystal/photosensitive prepolymer syrup via the self-interference patterns from two laser beams. The calculated reflectance spectrum for this structure matched the experimental one extremely well, giving very similar reflectivity peak positions and intensities. Slight discrepancies between the two reflectance spectra are attributed to either small variations in the microstructure of the reflection grating beyond that which is captured in the transmission electron micrograph, or the dispersion of the polymer which was not taken into account. These results serve as experimental verification of the theory for rough photonic crystals reported previously. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2336346]

One-dimensional periodic dielectric gratings, also known as one-dimensional photonic crystals (1DPCs), possess unique optical properties that make them desirable for many optical devices.[1] Recent research in 1DPCs has focused on advanced fabrication techniques, such as interference holography[2,3] or electrochemical etching,[4,5] that can generate large-area multilayer structures quickly and easily. Unfortunately, these techniques produce photonic crystals that have a large amount of interfacial roughness, causing them to be significantly different from their ideal analog. Until recently, the real optical behavior of such structures was not easily predictable using traditional techniques, such as the transfer matrix method.[6] However, in several recent papers,[7,8] we have proposed a method to predict the normal incidence reflectivity of rough 1DPCs using the homogenization approximation[9] in conjunction with the transfer matrix technique. As of yet, this theory has not been verified through comparison with experimental data. In this communication, we describe the fabrication of a rough reflection grating via interference holography, and use this 1DPC structure to validate the previously reported technique.

The 1DPC used in this study was designed to have three reflectivity peaks roughly corresponding to the red, green, and blue (RGB) portions of the visible spectrum. This was done in order to test the above mentioned theory with a more complicated structure, as well as to demonstrate the feasibility of fabricating a RGB reflector in a single step using a simple holography setup. Commercially available holographic photopolymers, such as Polaroid's DMP128 or DuPont's OmniDex, have already been used to create RGB reflectors that enhance the brightness and image quality of liquid crystal displays.[10] However, these reflectors are produced through a complicated process involving three holographic exposures, ultraviolet/visible light curing, and heating. Recently, a panchromatic ultrafine grain emulsion BBVpan, manufactured by Colourholographics Ltd., has been utilized to create a multiple band reflection grating in a single step.[11] However, this fabrication method involved a complicated setup using three separate laser wavelengths. Furthermore, the optical response of the device was poor due to the low index contrast in the starting polymer materials.

In contrast, the 1DPC created for this study was fabricated by a single-wavelength holographic exposure of a syrup. The syrup contained 57 wt % dipentaerythritol hydroxypenta acrylate (Aldrich) as a monomer, 1 wt % Rose Bengal (Spectral Group) as a photoinitiator, 2 wt % *N*-phenylglycine (Aldrich) as a coinitiator, 10 wt % *N*-vinylpyrrolidinone (Aldrich) as a cross-linking monomer, 15 wt % liquid crystal TL213 (Merck), and 15 wt % toluene (Aldrich) as a nonreactive solvent.[3] Several drops of the syrup were placed onto a glass slide, and a second slide was

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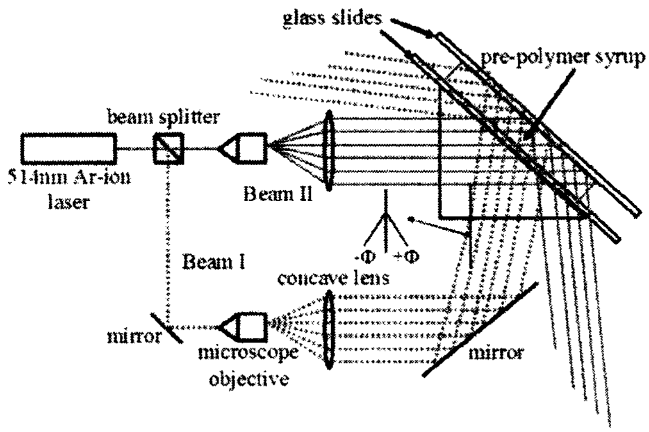

FIG. 1. Schematic of the optical setup for fabricating the multiplexed reflection grating.

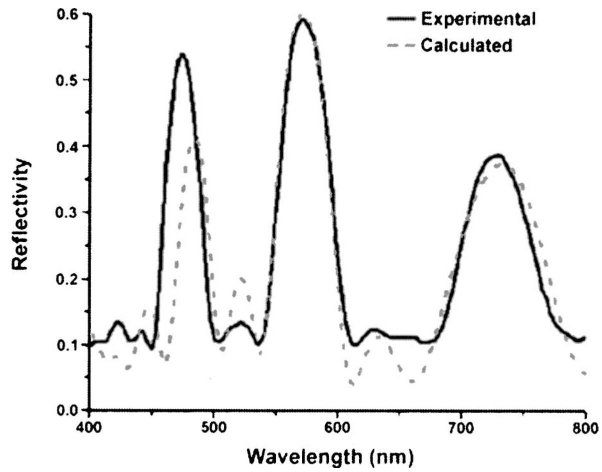

FIG. 2. The experimental (solid line) and calculated (dashed line) reflectivity spectrum of the fabricated rough multiplexed reflection grating.

pressed against the first one, causing the syrup to spread. The thickness of the syrup between the glass slides was controlled by 3 μm thick spacers.

The sample was introduced into the optical setup shown in Fig. 1. The 514 nm line of an Ar-ion laser operating at 200 mW was split into two separate beams (beams I and II), and each beam was then expanded with a 4× microscope objective and collimated with a 20 cm focal length concave lens. Each beam passed through different sides of a right-angle prism that was placed directly next to the sample. The beams then entered the sample and self-interfered with their own total internally reflected beams, creating the interference patterns which were used to expose the prepolymer syrup and write the grating pattern. During the photopolymerization process, the nonreactive solvent migrates to the low intensity regions which become polymer deficient due to the low rate of polymerization. Upon removal of the nonreactive solvent, lightly cross-linked polymer-rich bands and highly nanoporous void-rich bands develop.[3] The spacing in the interference patterns, and therefore the periodicity in the resulting grating structure, depend on the angle between the incident laser beam (Φ) and the normal of the prism. Positive angles produce finely spaced patterns (corresponding to bluer reflection gratings) with narrower void regions, while negative angles create patterns with larger spacings.[12] The angles of the two beams were varied independently to create a reflection grating that was tuned to both the blue ($\Phi=20°$) and green ($\Phi=-5°$) regions of the visible spectrum. A red reflectivity peak also appeared in the reflectance spectrum of the grating due to the combination of the blue and green periodicities in the written structure.

The prepolymer sample was exposed for approximately 1 min and subsequently postcured under a 75 W table lamp for 24 h. One of the glass slides was then removed, and the normal incidence reflection spectrum of the grating was obtained by measuring the transmission of the sample at normal incidence using a UV-3101 PC spectrophotometer and assuming zero absorption. This spectrum is shown in Fig. 2 (solid line). Red (729 nm), green (573 nm), and blue (474 nm) peaks are present with reflectivities of 39%, 59%, and 54%, respectively.

A bright-field transmission electron micrograph of the entire grating structure (i.e., all of the layers) is shown in Fig. 3(a). Although it appears that only one periodicity is present in this structure, a Fourier transform of this image reveals three distinct frequencies corresponding to the three periodicities, as shown in the inset of Fig. 3(a). It is clear from this micrograph that the grating structure deviates from its ideal analog quite substantially due to the extreme amount of interfacial roughness present. Therefore, traditional simulation techniques, such as one-dimensional transfer matrix,[6] cannot be used to accurately predict the real optical behavior of this structure directly. Thus, we have used our previously proposed method[7,8] to calculate the theoretical reflectivity spectrum of this structure at normal incidence. This was done by using a modified version (to account for the dispersion of the glass slide) of the code in Appendix A of Ref. 13.

Our theory[7,8] posits that the optical behavior of a rough 1DPC at normal incidence will be equivalent to the response of a 1DPC with a diffuse refractive index profile, where the inhomogeneity of the refractive index at the rough interfaces is averaged out. A three-step process was used to implement this theory on the fabricated structure. First, the micrograph in Fig. 3(a) was converted into a refractive index mapping using a simple MATLAB script. The lighter regions were mapped to the refractive index of the voids (1.0), while the darker regions were mapped to the refractive index of the polymer (1.52) measured at 632 nm. Second, the averaged index profile for the structure was obtained by averaging the dielectric constant across each pixel row in the mapping,

$$n_{\mathrm{av}}(j) = \left\{ \frac{1}{N} \sum_{i=1}^{i=N} [n_{\mathrm{map}}(i,j)]^2 \right\}^{1/2}. \qquad (1)$$

Here, N is the total number of pixels in the direction parallel to the layers, i and j indicate the pixel index in the mapping, and $n_{\mathrm{map}}(i,j)$ is the value of the refractive index at the i,j pixel position. Both the refractive index mapping and the averaged refractive index function for the structure in Fig. 3(a) are shown in Fig. 3(b).

Finally a one-dimensional transfer matrix calculation was performed at normal incidence on the homogenized

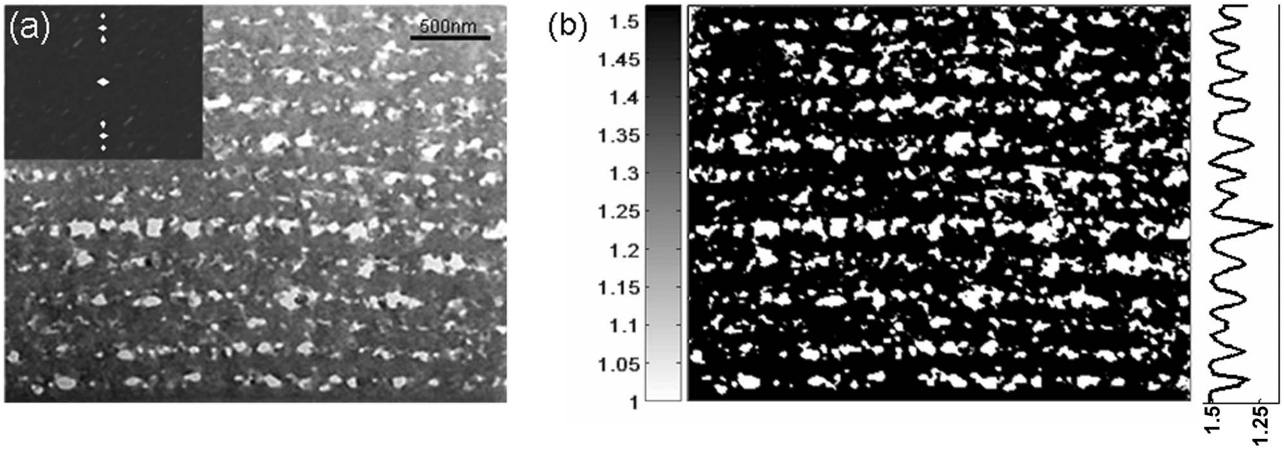

FIG. 3. (a) A transmission electron micrograph of the fabricated reflection grating. The inset shows the Fourier transform of the micrograph, revealing the three periodicities in the structure. (b) The refractive index mapping of the image shown in (a) and the averaged refractive index profile calculated from it.

structure to predict its reflectance spectrum in the relevant wavelength range. In addition, the extra reflection that occurs from the back surface of the glass slide was added to the calculated spectrum. To second order, this additional reflection at normal incidence was derived to be

$$\Delta R = \left(\frac{1 - n_{\mathrm{glass}}}{1 + n_{\mathrm{glass}}}\right)^2 (1 - R_1)(1 - R_2), \qquad (2)$$

where n_{glass} is the refractive index of the glass slide, R_1 is the normal incidence reflectivity of the 1DPC (with semi-infinite glass substrate) coming from the top, and R_2 is the normal incidence reflectivity of the 1DPC coming from the bottom (i.e., from the glass side). The total calculated reflectivity, including this offset, is shown in Fig. 2 (dashed line).

The calculated reflectance spectrum agrees very well with the experimental one for wavelengths above about 530 nm. However, the blue end of the calculated spectrum significantly deviates from the experimental spectrum. The blue reflectivity peak is still present in the calculated spectrum, but its position and height are shifted. There could be at least three reasons for this. First, the blue end of the spectrum is most sensitive to small variations in the structure. Therefore, if we mapped some of the pixels in the image to the wrong refractive index, then the blue end of the spectrum would begin to deviate from the real response of the structure. This could possibly be corrected with a better image quality. Second, the spot size in the spectrophotometer samples a much larger area than what is seen in the micrograph. Therefore, small variations in the microstructure of the grating, within the spectrophotometer spot size, from the structure that is represented in the micrograph will affect the blue end of the spectrum much more than the red end. This could be remedied somewhat by taking several images at different positions on the sample and averaging the calculated spectra from each image. Third, the dispersion of the polymer itself was not considered in the calculation. This could significantly alter the reflectivity of the grating at wavelengths below 530 nm, where the refractive index and absorption may become substantially larger than those measured at 632 nm.

In conclusion, a 1DPC tuned to reflect the red, green, and blue portions of the spectrum was fabricated through the holographic exposure of a prepolymer syrup. Our previously reported technique[7,8] for calculating the normal incidence reflectance spectrum of rough 1DPCs was employed to calculate the theoretical reflectivity of the fabricated grating. The calculated spectrum matched the experimental one extremely well for wavelengths above about 530 nm. The blue end of the calculated spectrum deviated somewhat from the experimental reflectivity, possibly due to the dispersion of the polymer. This work serves as experimental verification of the homogenization approximation method for calculating the reflectivity of rough 1DPCs that we proposed earlier.

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