Fabrication of Thin-Shell Periodic Nanopillars with Near-Unity Refractive Index

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The refractive index contrast between two materials is a critical parameter in the design and performance of photonic structures and devices. In photonic crystals, the width of the photonic bandgap is directly dependent on the refractive index contrast, influencing the propagation and confinement of light within the structure [1]. In distributed Bragg reflectors (DBRs), the difference in refractive index between the constituent layers determines optical properties, such as reflectivity and penetration of the optical wave [2]. Various methods have been explored to achieve low-refractive-index materials, including oblique deposition, the sol-gel process for silica aerogel films, and chemical vapor deposition of carbon nanotubes (CNTs) [3,4]. Our research group has developed highly ordered, thin-shell low-index nanolattice materials with tunable refractive indices via nanolithography and shell thickness control [5]. However, structural collapse and defect formation have been observed during fabrication, leading to optical scattering.

In this work, a periodic thin-shell nanopillar array is employed as a platform to investigate the optical characteristics of nanolattice architectures. Laser interference lithography is utilized to fabricate highly ordered 2D templates, enabling improved control over processing conditions and uniformity. The optical properties of nanopillar arrays with varying geometries are characterized using spectroscopic ellipsometry. This study contributes to a comprehensive understanding of the design principles and optical characteristics of thin-shell nanolattice structures. The fabrication process is illustrated in **Figure 1(a)**. Periodic nanopillars with a height of 830 nm and a period of 650 nm are initially patterned using Lloyd's mirror interference lithography. A conformal Al_2O_3 thin film is subsequently deposited via atomic layer deposition (ALD), consisting of 220 cycles, resulting in a thickness of 20 nm. The underlying polymer template is then removed through thermal annealing at 550° for 4 hours. The fabricated thin-shell nanopillars are characterized using scanning electron microscopy (SEM), as shown in **Figure 1(b)**.

The refractive index of the fabricated structure was characterized using spectroscopic ellipsometry, as illustrated in **Figure 2**. Data acquired at an incident angle of 70° was selected for analysis due to its low fitting error and high measurement accuracy. The optical response of the thin-shell nanopillars was modeled using a homogeneous Cauchy layer using a nanopillars/SiO_2/Si multilayer. **Figure 2(a)** presents the measured and fitted Δ and Ψ values across the wavelength range of 500-1200 nm. The mean-squared error (MSE) of the fitting was determined to be 85.3, which is primarily attributed to noise in the experimental data. The broadband refractive index of the hollow nanopillars is plotted in **Figure 2(b)**, with the refractive index of air included as a reference. The lowest index obtained was 1.028, significantly lower than that of bulk Al_2O_3 ($n \approx 1.65$). In contrast with previous work on low-index nanolattice materials, the proposed approach using interference lithography can yield full-substrate area with low defect density.

We have demonstrated the fabrication of highly uniform and periodic thin-shell nanopillar arrays with an ultralow refractive index using Lloyd's mirror interference lithography. To achieve a more comprehensive understanding of the design principles, the influence of geometrical parameters, such as periodicity and shell thickness, will be further explored. Additionally, the optical anisotropy arising from the periodic structural arrangement will be systematically analyzed.

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Figures:

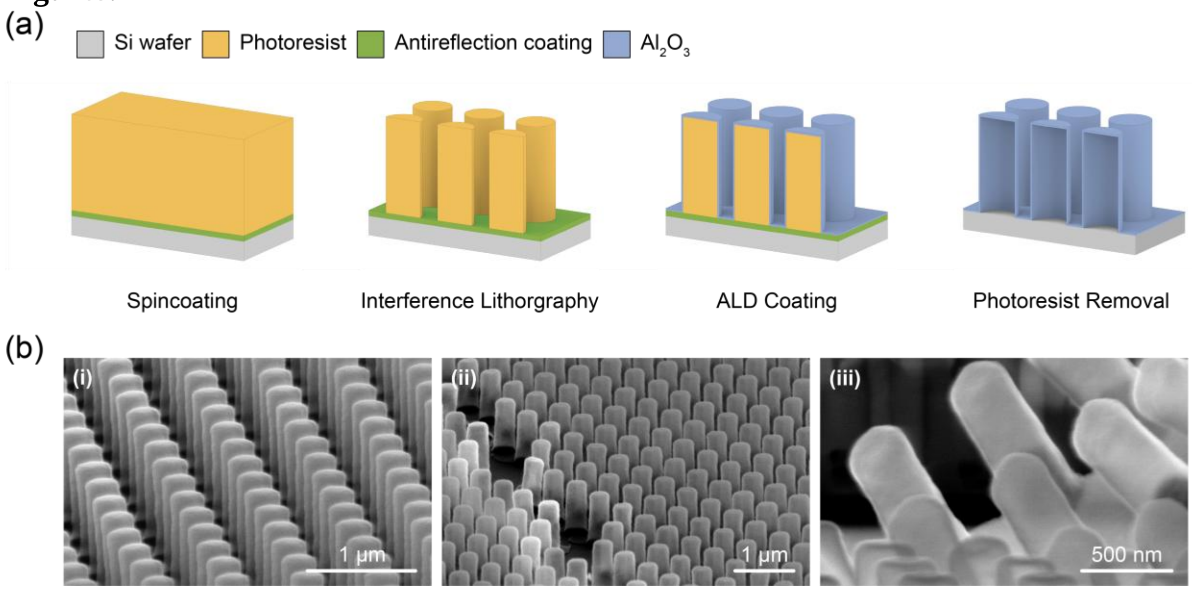

Figure 1. (a) Schematic illustration of the fabrication process for the thin-shell nanopillar array. (b) SEM image of the fabricated thin-shell nanopillar array, demonstrating high uniformity and periodicity. (i) Before template removal. (ii,iii) After template removal.

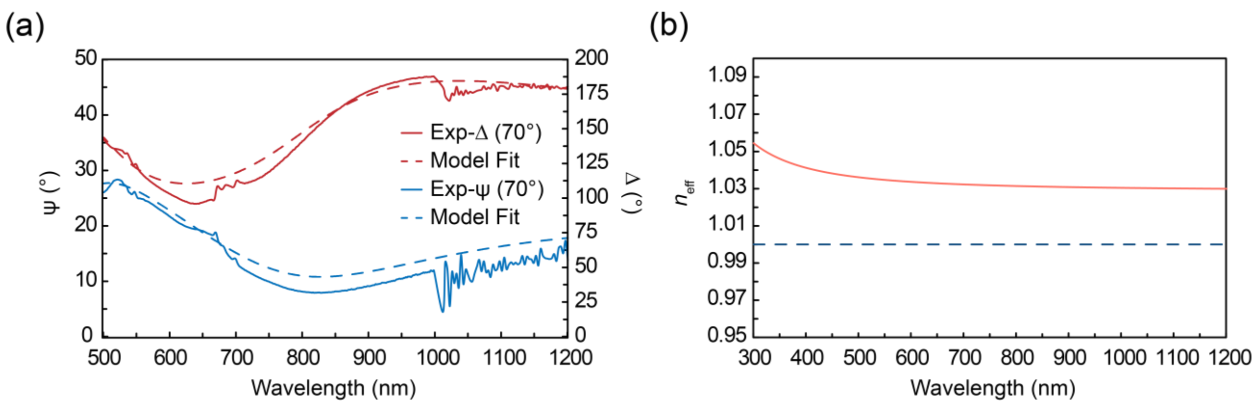

Figure 2. (a) Comparison of experimental and modeled Δ and Ψ values as a function of wavelength in the range of 500-1200 nm, measured at an incidence angle of 70°. (b) Effective refractive index (n_{eff}) of the thin-shell nanopillar structure over spectrum.

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