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Large-area porous alumina photonic crystals via imprint method

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Abstract

A perfect 2D porous alumina photonic crystal with 500 nm interpore distance was fabricated on an area of $4\ cm^2$ via imprint methods and subsequent electrochemical anodization. A 4" imprint stamp consisting of a convex pyramid array was obtained by modern VLSI processing using *DUV*-lithography, anisotropic etching, *LPCVD* Si_3N_4 deposition and wafer bonding. The optical properties of the porous alumina photonic crystal were measured with an infrared microscope in Γ-M direction. For both polarizations, a bandgap is observed at around 1 µm for r/a = 0.42. A reflectivity of almost unity for E-polarization in the region of the bandgap is a sign of the high quality of the structure, indicating almost no scattering losses. These experimental results could be correlated very well to the bandstructure as well as reflectivity calculations assuming a dielectric constant of ε = 2.0 for the anodized alumina.

Introduction

Periodic dielectric materials allowing to control the flow of light are classified as photonic crystals (PCs)[1]. After the concept of PCs was theoretically proposed, numerous papers have been published focusing on the near-IR spectral range. For extending this concept to the range of visible frequencies, new materials which are transparent in the visible and allow nano-structuring are required. Nowadays, 3-dimensional inverse opal structures [2], microperiodic polymeric structures via holographic lithography [3] and autocloning technique stacking alternatively two different materials on substrate with groove [4] are attractive to fabricate photonic crystals for visible ranges.

Due to their low absorption coefficient, excellent thermal stability and easy handling, porous alumina structures could be potential materials for photonic crystals. However, since a perfect arrangement of porous alumina channels is required for 2-D PCs, only few studies on porous alumina photonic crystals exist [5-7]. To fabricate mono-domain porous alumina structures, a method to pre-pattern the surface of aluminum before conventional anodization is a prerequisite. So far, three kinds of methods for pre-structuring have been developed: one is imprint by a dot-like stamp [8], another is e-beam lithography [9], and the other is 2-step imprint by commercial gratings [7]. For mass products in the future, imprint methods are considered to be more suitable than the e-beam lithography. However, the stamps, which have been developed for imprinting up to now, allow only small imprint areas.

Here, we will introduce a novel 4″ imprint stamp consisting of regular arrays of Si_3N_4 pyramids obtained by standard silicon processing techniques and discuss porous alumina photonic crystals as one of the possible applications.

Fabrication of a novel 4" imprint stamp.

For the imprint master stamp, a 4″ silicon wafer (100) was used as starting material. A 2D hexagonal array with a period of 500 nm on the silicon wafer was patterned by deep-*UV* lithography (248 nm). Afterward, the pattern was dipped in KOH for anisotropic etching, resulting in inversed pyramids in the silicon wafer. A replica of the Si inversed pyramids was obtained by *CVD*-deposition of Si_3N_4 layer on top of the patterned silicon. Another silicon substrate was bonded on top of the Si_3N_4 layer so that the Si_3N_4 was sandwiched between two silicon substrates. Subsequently, the initially patterned silicon substrate was removed by grinding and spin-etching. The pyramid of the master stamp with a height of about 260 nm and a lattice constant of 500 nm are shown in figures 1 (a) and (b).

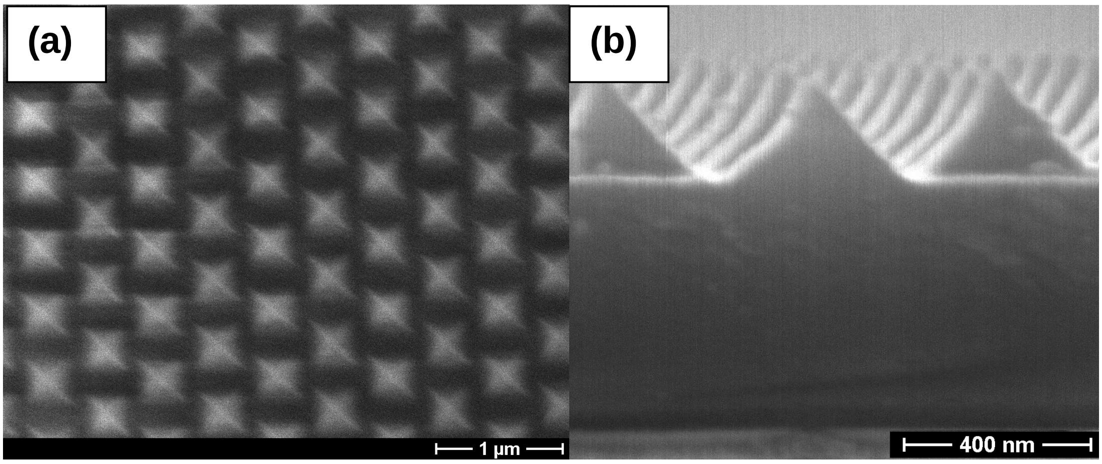


Figure 1 SEM images of a novel imprint stamp consisting of pyramids with 500 nm lattice constant and 260 nm height. (a) top view and (b) side view

The pyramidal shape of the master is very suitable to press it on electropolished aluminum surface. Typically, a pressure for indentation as low as $5kN/cm^2$ is used, which is 50 times lower than the dot-like stamp fabricated by Masuda's group [5, 10]. In figure 2, we can see inverse pyramidal holes in the aluminum surface after indentation. It indicates good pattern transfers from master stamp onto aluminum surface. The depths of the holes formed by imprinting with different pressures have been investigated by AFM depth analysis as shown in Fig. 3. In fact, a pre-patterned hole with the depth of 20 nm is sufficient to guide the position of a pore formed by subsequent anodization. From the *AFM* analysis, it can be observed that the surface roughness of pre-patterned area is much lower than that of just electropolished area. It could effectively lead to a perfect arrangement of pores at the beginning of pore formation.

Anodization of pre-patterned aluminum

The interpore distance of porous alumina is strongly influenced by the anodizing potential with a proportionality constant of around 2.5 nm/V [11]. For example, 500 nm periodic porous structures can be fabricated at 195V. Therefore, a pre-patterned aluminum with 500 nm interpore distance was subsequently anodized

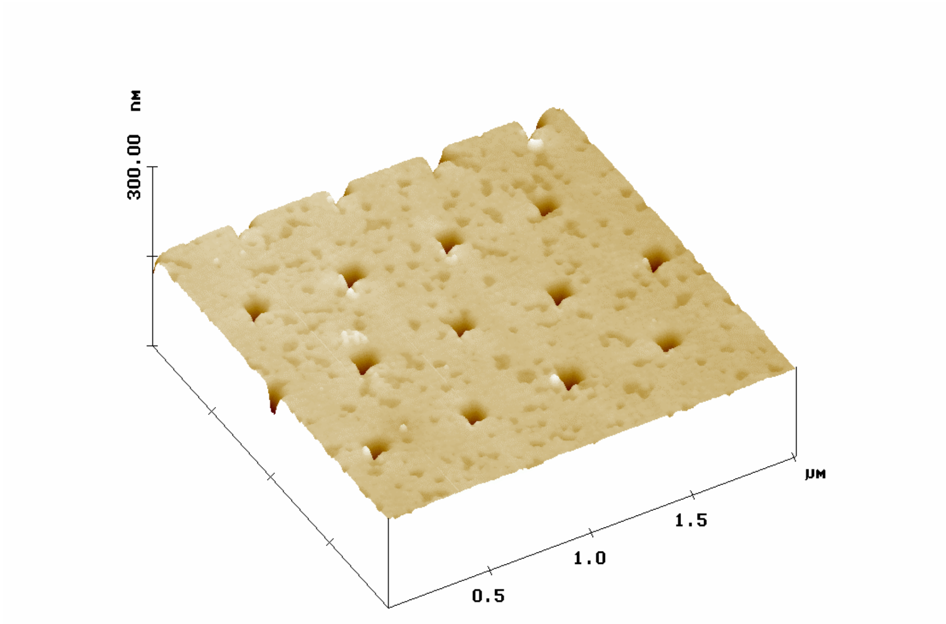


Figure 2 AFM image of inverse pyramidal holes in the surface of electropolished aluminum after imprint under 5 kN/cm^2.

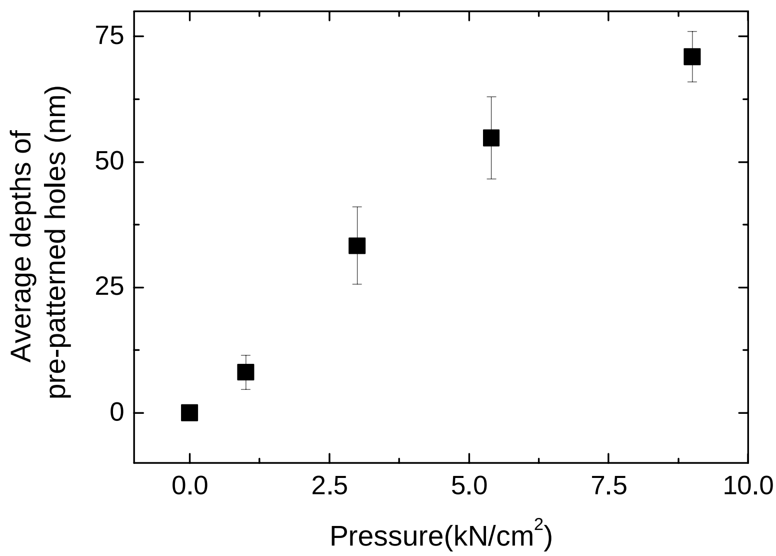


Figure 3 The relationship between imprint pressure and average depths of pre-patterned holes in the surface of aluminum.

in 1 wt% phosphoric acid at 195 V for 16h. The rate of growth of pore channels in phosphoric acid at 195 V is approximately 5 ~ 7 µm/h. In figures 4 (a) and (b), we can see perfectly arranged pores and straight pore-channels, respectively. In principle, a mono-domain of porous alumina with 4 cm^2-scale can be fabricated by our imprint method. In addition, notice that the inversed pyramidal holes shown in figure 2 change to a circular shape shown in figure 4 during the anodization. This means that the pore shape after anodization is independent of pre-pattern shape on the surface. Note that the pore shape can be control by the pre-patterning lattice such as rectangular and honeycomb [12].

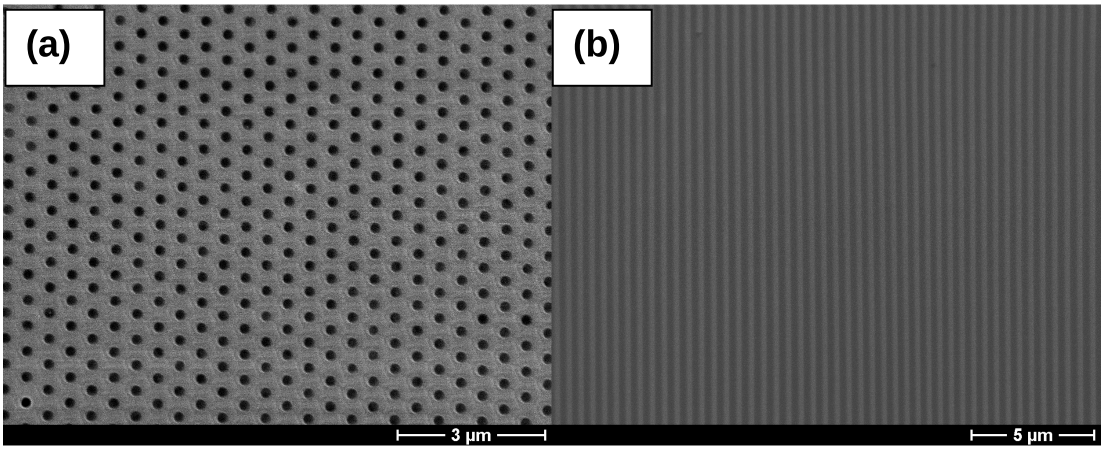
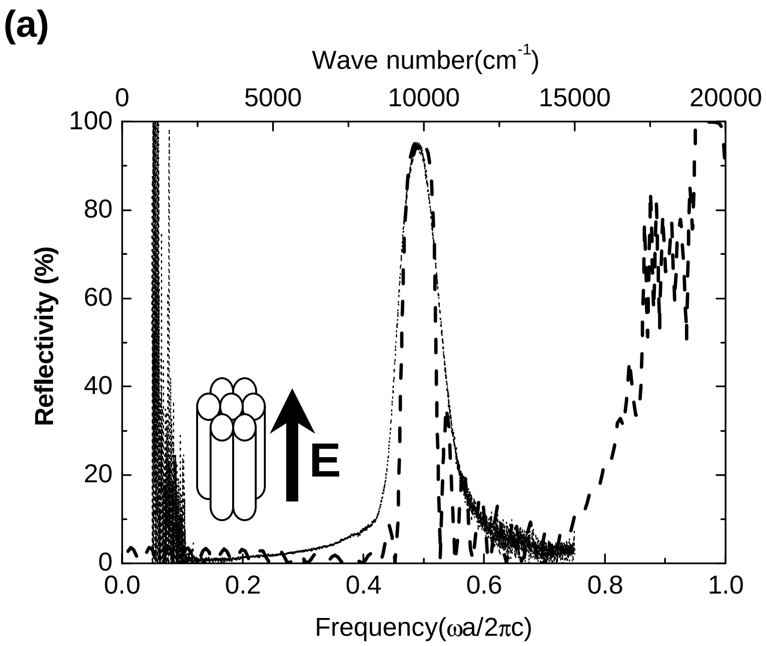


Figure 4 SEM images of a mono-domain porous alumina fabricated by anodization of pre-patterned aluminum. (a) top view and (b) side view

Optical characterization of porous alumina

For the optical characterization, the reflectivity of the porous alumina structure with r/a = 0.42 (radius / interpore distance) is measured along Γ-M direction with a FT-IR microscope. In figs. 5 (a) and (b), we compare the measured reflectivity (solid lines) and the theoretical calculation (dot lines) computed by the transfer-matrix method [13]. For both E- and H- polarization, the band-gap positions observed in the measurement are in good agreement with calculations assuming $\varepsilon = 2.0$. In addition, almost 100% reflectivity for E-polarization in the band gap is observed, indicating very low losses and high quality structures. The slightly stronger angular dependence of the H-polarization may lead to the lower reflectivity, since the IR-objective has an angular aperture of 30°.



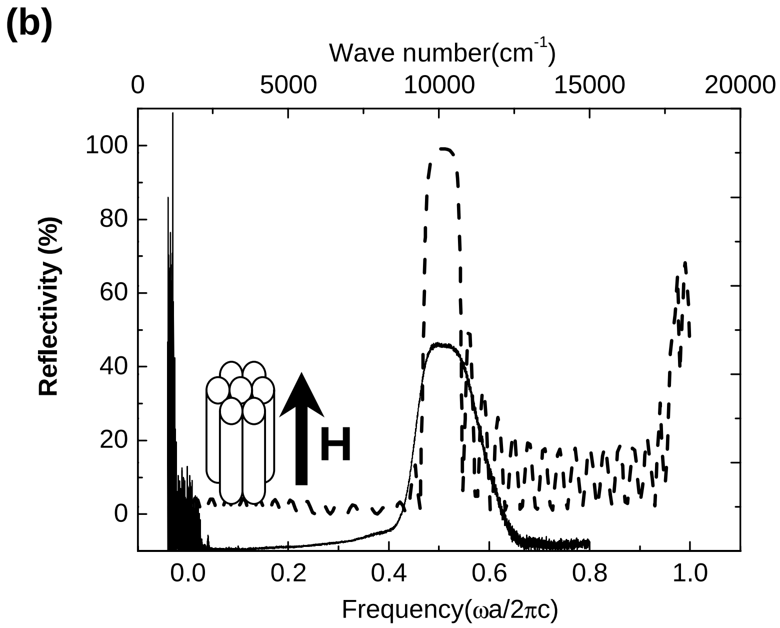


Figure 5 Reflectivity measured by FT-IR microscope (solid lines) and calculation computed by transfer matrix method assuming ε = 2.0 and 8 layers (dot lines). (a) E- polarization and (b) H- polarization.

The bandstructure of porous alumina with r/a = 0.42 is calculated by plane-wave method (figure 6). In Γ-M direction, a bandgap exists at the normalized frequency of around 0.5 in very good agreement with the experimental results and the computations of figure 5.

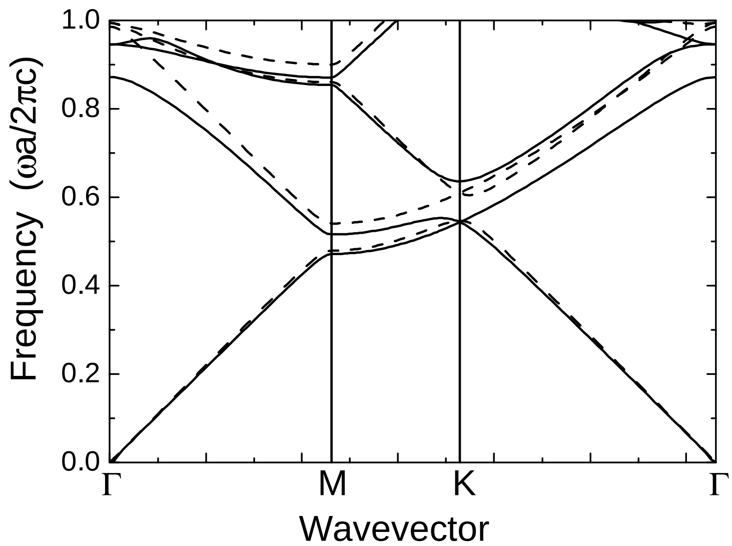


Figure 6 Bandstructure of porous alumina photonic crystals calculated by plane wave method assuming ε =2.0 when r/a = 0.42. Dash lines correspond to TE (H) polarization and solid lines to TM (E) polarization.

Conclusion

A novel 4" imprint stamp consisting of a pyramid array having a 500 nm lattice constant and 260 nm height was developed via VLSI processing. 2D porous alumina photonic crystals with 500 nm interpore distance were fabricated via imprint methods. For r/a = 0.42, a bandgap was observed at around 1 µm which was in good agreement with theoretical calculation assuming a dielectric constant of $\varepsilon = 2.0$.
In the case of E-polarization, a reflectivity of almost unity was observed, indicating almost no scattering losses and high quality structures.

Acknowledgements
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