Macroporous silicon: Homogeneity investigations and fabrication tolerances of a simple cubic three-dimensional photonic crystal

Sven Matthias, a Reinald Hillebrand, b Frank Müller, and Ulrich Gösele
Max Planck Institute of Microstructure Physics, Weinberg 2, Halle 06120, Germany

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Large area three-dimensional (3D) photonic crystals have been fabricated either by layer-by-layer methods, colloidal self-assembly, or by macroporous silicon etching. The last mentioned method has proven to be a versatile and fast technique to manufacture simple cubic 3D photonic crystals, having a complete photonic band gap with a width of 4.99% in the infrared spectral range. This report is focused on the investigation of their homogeneity and additional numerical simulations concerning the tolerable disorder allowing still for a complete photonic band gap. Fabry-Pérot resonators, which are realized by 3D photonic crystals containing planar defects, are characterized optically in spatially resolved transmission measurements by scanning infrared Fourier spectroscopy. The impact of the observed inhomogeneities on the complete photonic band gap is studied in detail by modeling the influence of structural parameters. Fabrication tolerances for the simple cubic arrangement of intersecting air spheres in silicon are deduced. © 2006 American Institute of Physics.

I. INTRODUCTION

Photonic crystals1–3 gained considerable interest in basic research and technology due to their unusual optical properties allowing, e.g., the superprism effect,3 lossless waveguiding,4 or supercontinuum generation.5 In particular, three-dimensional (3D) photonic crystal structures could prohibit light propagation throughout a specific frequency range in all directions and can be thought of as the material for optical components in the 21st century. Although various 3D photonic crystal structures have been proposed and some of them were realized,6–11 their almost disorder-free experimental manufacturing remains a major challenge. A lot of effort is directed on the experimental fabrication of structures with diamond symmetry using layer-by-layer methods or microassembly in order to benefit from the large complete band gaps. Although simple cubic 3D photonic crystals exhibit smaller complete photonic band gaps of 5% up to 12%,6,12–14 they are attractive due to the easy, low cost, and fast fabrication by very large scale integrated (VLSI) technology. Three kinds of large area approaches focusing on the manufacturing of simple cubic structures are known: layer-by-layer methods,15 conventional planar silicon micromachining,16 and complex macroporous silicon etching.17 The last two mentioned methods are suitable for the fabrication of simple cubic arrangements of intersecting spherical voids in a silicon matrix.17 Figure 1 shows a scanning electron microscopy (SEM) image of such nearly perfect photonic crystals. Though the interesting optical phenomena of photonic crystals are based on their perfect peri-

FIG. 1. Scanning electron micrograph (SEM) of a simple cubic 3D network of intersecting air spheres obtained by isotropic etching of a strongly modulated macroporous silicon sample.
odicy and shape, structural disorder affects the optical properties significantly. It is a matter of fact that measuring band gaps with high precision optically is not straightforward for nonperfect structures. Usually, a model for the type of imperfection is needed and then the modeled spectra are fitted to the measurements to deduce the model parameters.

In this report we use a different approach, which is more related to a real-world device application. A planar defect is embedded between two photonic crystals. For an appropriate design such a geometrical defect generates an optical state inside the forbidden gap. The structure acts as a resonator, which leads to an additional peak in the transmission spectrum. Within the fundamental stop gap this device can be considered as a Fabry-Pérot resonator. In resonance, the optical length $l_{\text{res}}$ of the resonator is $l_{\text{res}}=\lambda/2$, where $\lambda$ is the wavelength inside the medium. While the length of the pores does not significantly fluctuate, the pore shape and therefore the effective refractive index $n_{\text{eff}}$ do. Hence a geometrical variation of these parameters will influence the resonance frequency. The quality in the undisturbed layers mainly influences the reflectivity of the two Bragg mirrors around the defect and thus determines the $Q$ factor of the resonator. In this report we concentrate at first on the frequency of the resonance as a measure of the pore shape fluctuations.

An example of the fabricated Fabry-Pérot resonators is shown in Fig. 2(a) (inset). The defect plane is in between two 3D photonic crystals acting as Bragg mirrors as five layers each. The transmission spectrum of this resonator along the growth direction, i.e., perpendicular to the defect plane, is measured by Fourier transform infrared spectroscopy (FTIR). One example of such a FTIR transmission spectrum is displayed in Fig. 2(a).

For a large-scale inspection of the photonic crystals we mapped the resonance frequency for a squared area of 10 $\times$ 10 mm$^2$, with a squared spot size of 250 $\times$ 250 $\mu$m$^2$. A motorized stage control, which scans the sample in two dimensions, allows performing $40 \times 40$ measurements with an increment of 25 $\mu$m. In this way 1600 spectra [similar to Fig. 2(a)] were taken and the resonance frequency was automatically extracted. Figure 2(b) shows the resonance position of such a sample as a gray scale plot. The resonance frequency covers the range from 960 up to 1050 cm$^{-1}$. This is equal to a change of $\pm 4.5\%$, which means that the product of the effective refractive index $n_{\text{eff}}$ and the length of the defect $l_{\text{def}}$ vary over this area by about $\pm 4.5\%$. Moreover, the variation of the defect resonance frequency for the different positions is not statistically distributed. Instead, we observe a wafer concentric pattern. The spacing between the rings is on the order of half a millimeter. Independent of the specific etching parameters such as temperature and concentration of the hydrofluoric acid (HF) or the illumination, we observe this characteristic pattern. Such different optical properties are not due to our etching setup, because the streaming profile of the HF in our setup avoids gas bubbles or inhomogeneous HF concentration. In addition, we observed that the ring pattern is more pronounced for substrates with a higher doping level. A previous investigation suggested this implication as an electronic effect, induced by striations in the substrates used. Striations are an intrinsic property of silicon wafers and are due to dopants and contaminants (e.g., heavy metals) built in inhomogeneously during the crystal growth process.

By increasing the spatial resolution of our mapping technique, we were able to scan the sample with a spot size of $40 \times 40 \mu$m$^2$ and an increment of 25 $\mu$m. The experiment resolves the fine structure of the striations, indicating that the sample is indeed homogeneous in the areas between the striations. The 3D contour plot [Fig. 2(c)] visualizes the topology and indicates that the structure is homogeneous for typical device sizes of $100 \times 100 \mu$m$^2$. The estimated fluctuation is less than $\pm 0.5\%$ over this area. Overall, the characterized Fabry-Pérot structure consists of three components: the two 3D photonic crystals and the defect layer in between. This planar defect does not only act as an optical resonator, it also affects the pore growth below the defect layer. Thus we can expect that the homogeneity of an extended 3D photonic crystal without any defect layer should be even better than the reported values.

The macroporous silicon approach to 3D photonic crystals is based on a fast and versatile fabrication process acting on a large area. However, the observed spatial variations in the transmission spectra indicate the unavoidable influence of the substrate on the quality and homogeneity of the 3D structure. Since the initial material is converted into 3D simple cubic crystals, the inhomogeneity remains also in the final material. Generally, such a change in the resonance frequency stands for a geometrical modification of the assumed model system of intersecting air spheres. Therefore we consider in the next section the variations of the diameter of the spheres, the length of the modulation period, and the exact
shape of the voids to evaluate their influence on the size and central frequency of the complete photonic band gap.

III. NUMERICAL SIMULATION

Band structures of a simple cubic lattice of intersecting air spheres ($\varepsilon = 1$) in silicon ($\varepsilon = 11.6$) have been compiled using the well-established MIT package. In Fig. 3(a) the photonic bands are plotted along the $\mathbf{k}$ directions of the irreducible Brillouin zone. The maximum band gap appears for a sphere diameter $d_{\text{sphere}} = 1.20a$ around a central frequency of $f_0 = 0.482 \, [a/\lambda]$. The width of the band gap $\Delta f$ is 4.99% of the central gap frequency $f_0$. Figure 3(b) indicates the important geometrical parameters of the structure.

The most important parameter that has to be controlled is the porosity or the air sphere diameter. The dependency of the band gap on the diameter is shown in Fig. 4(a). As the fabrication process starts from a columnar structure that is later widened to the cubic arrangement, deviations from the ideal cubic symmetry can be expected. Figure 4(b) shows the dependency for ellipsoidal air volumes, whereas Fig. 4(c) concentrates on a deviating lattice constant for the pore growth direction, i.e., a tetragonal lattice distortion.

In Fig. 4(a) the diameter of the air spheres $d_{\text{sphere}}$ is altered around the optimum value of $d_{\text{sphere}} = 1.20a$. The complete photonic band gap between the fifth and sixth bands [cf. Fig. 3(a)] remains open for remarkable size variations of the spheres. In Fig. 4(b) we continue to assume a perfect cubic lattice geometry. The ellipsoidal air volumes arranged in the cell are characterized by their eccentricity $d_x/d_{\text{eff}} \in [0.90, 1.10]$. The effective air volumes correspond to sphere diameters $d_{\text{eff}} \in 1.15a, 1.21a, 1.30a$. The band structure calculations clearly show that perfect spheres with appropriate air filling ratio provide the maximum complete band gap. In Fig. 4(c) we study if combinations of ellipsoidal deformations of the air volume and tetragonal lattice distortions allow to increase the complete photonic band gap. In the parameter space analyzed, i.e., for tetragonal distortions $h \in [0.90a, 1.10a]$ and ellipsoidal deformations of the air spheres $d_x/d_{\text{eff}} = 0.975, 1.0, 1.025$ ($d_{\text{eff}} = 1.20a$), the answer is negative. There are $h-d_x/d_{\text{eff}}$ combinations that reveal a larger gap than the sphere for the same tetragonal distortion $h$. But the absolute maximum of the gap size remains for a sphere in a simple cubic lattice.

Gap maps show the physical behavior of air bands and dielectric bands, bordering a band gap, in a comprehensive way. The central frequency of photonic band gaps shifts with the mean refractive index of the photonic crystal. In Fig. 5(a) we display the gap map calculated for increasing diameter of the air spheres. From Fig. 4 we know that the interval of frequencies inside the gap reaches 5%, but the frequency slope in the gap map is high.

Let us assume an optical device, which works with a fixed frequency near the center of the gap map, utilizing the effect of inhibited photon transfer. The computations, provid-

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**FIG. 3.** (a) Band diagram for the simple cubic structure of intersecting air spheres. The inset represents the Brillouin zone for all computations. (b) Cross section of the structure model with the parameters varied to simulate fabrication tolerances.

**FIG. 4.** Size variations of the complete photonic band gaps of Si-based photonic crystals vs sphere diameter (a), ellipsoidal deformations (b), and tetragonal distortions (c). (a) Variation of $d_{\text{sphere}} \in [1.1a, 1.3a]$. (b) Ellipsoidal air volumes of effective diameters $d_{\text{eff}} \in 1.15a, 1.21a, 1.30a$ with an eccentricity of $d_x/d_{\text{eff}} \in [0.90, 1.10]$. (c) Tetragonal lattice distortions $h \in [0.90a, 1.10a]$ with ellipsoidal deformations of the air spheres $d_x/d_{\text{eff}} = 0.975, 1.0, 1.025$ ($d_{\text{eff}} = 1.20a$).
ing Fig. 5(a), show that a variation of the air sphere diameter of 0.032a, which is 2.65% relative to the total sphere diameter, is sufficient to push the working frequency outside the gap. Expressed in terms of the air filling fraction, the value of 80.5% is optimum. A change of the air volume of ±4%, which could be caused by striations in the basic material, will prevent an overall gap for one fixed frequency. Nevertheless, there are possible applications that might profit from a very narrow, precise complete band gap. There is, for example, plenty of volume inside the photonic crystals of simple cubic type to fill the pores for tuning the dielectric contrast, or to apply nonlinear optical effects to shift or switch the gap.

If it can be managed to keep the air filling fraction of the photonic crystals constant on a large scale, solely the shape effects of the air volumes can influence the photonic properties. Figures 5(b) and 5(c) show specific gap maps, where the air filling is assumed to be constant. The graph of Fig. 5(b) shows that pure elliptical deformations are uncritical if the porosity is near optimum. The figure has little slope, which makes the photonic band gap relatively robust with respect to ellipsoidal deformations of up to ±10%. This message holds correspondingly for the tetragonal lattice distortions assumed in Fig. 5(c), in particular, for a shortening of the h axis. To keep the air volume fraction constant, the air spheres shrink proportional to the shortening of the h axis of the lattice, and vice versa. The presented curves of Figs. 5(b) and 5(c) are not perfectly smooth because of the nonequidistant steps in the d/a, dvy scale and the numerical grid being applied to nonspherical and noncubic models.

Each structural parameter can be varied by about ±10% before the band gap closes if all the other parameters are fixed to its optimum value, allowing for 4.99% gap size. This estimation holds if a local shift of the central gap frequency can be accepted or is desired (cf. Fig. 4) for the application intended. However, if the potential optical device is driven with one fixed frequency [see Fig. 5(a)], there are very hard demands on the wafer homogeneity being restricted on the contiguous wafer areas experimentally identified.

IV. CONCLUSION

Photoelectrochemical etching of photonic crystals is attractive due to its potential of large-scale integration. The sample area is solely limited by the wafer dimensions, presently to 150 mm, and the depth of the pores is restricted by the wafer thickness. The quality of the silicon substrate used, especially the occurrence of striations, constrains the homogeneity of the macropores. The presented physical method of characterizing homogeneity clearly quantifies the limitations of a uniform pore growth. In particular, for 3D photonic crystals some global disorder is present, which has been optically measured by large-scale mapping of planar defect states. Scanning with higher spatial resolution yielded a fluctuation of ±0.5% on an area of 100 × 100 μm². This value is significantly lower than the calculated tolerance interval of ±10% and the resulting macroporous silicon samples could be considered as homogeneous for this typical device sizes. As long as the doping level of the silicon wafer varies, there will be striations, because the fabrication of macroporous silicon suffers from doping fluctuations regardless of the actual 3D shaping. In particular, for the strongly modulated samples with the high voltage period, these striations will be critical. In conclusion, the experimentally gained facts on the homogeneity of the 3D photonic crystals provide clear evidence that the theoretical requirements of a complete photonic band gap can be fulfilled in all wafer areas between the striations, i.e., on a device-size scale. Applying rather low potentials during the pore growth can minimize the influence of the striations, which will as far as possible disappear when neutron beam doped substrates will be utilized.

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