Fabrication of Nanoimprint stamps for photonic crystals

Kouba J., Kubenz M., Mai A., Ropers G., Eberhardt W., Loechel B.

aBESSY GmbH, Anwenderzentrum für Mikrotechnik; Albert-Einstein-Str. 15, 12489 Berlin, Germany
b micro resist technology GmbH, Köpenicker Str. 325, 12555 Berlin Germany
E-Mail: josef.kouba@bessy.de

Abstract: We report on fabrication of nanoimprint stamps for fabrication of two dimensional photonic crystals in visible range of spectra. Nanoimprint stamps made of silicon and/or nickel were successfully fabricated using electron beam lithography and advanced dry etching techniques. The quality of the stamps was evaluated using scanning electron microscopy. The fabricated stamps were also evaluated by imprinting them into suitable polymer materials.

1. Introduction

Photonic crystals are electromagnetic materials with spatially periodical dielectric properties. First proposed by Yablonovitch [1] and John [2], the spatial periodicity of photonic crystals leads to Bloch-Floquet theorem and to the existence of photonic bands, analogous to electron bands in crystalline solids. Further, for a certain spatial dielectric functions, the dispersion relation reveals band gaps, regions of frequencies in which photons can not propagate through the material.

Photonic crystals enable strong confinement and localization of light on sub wavelength scale and thus enable to reach high field intensities and intensive interaction between the light and material. The resulting effects are believed to play a key role of future core components of novel electro optical applications. The potential applications reach from simple waveguides or splitters over multiple wavelength demultiplexers and wavelength filters to advanced applications such as single photon sources or laser resonators. Apart from telecommunication sector, future sensor and data processing industry may profit out of photonic crystals. Concepts already exist for pure optical switching elements, marking the first step to build an optical transistor.

One of the major drawbacks of broader use of photonic crystals is their cost intensive fabrication. Sub 100 nm dimensions require typically the use of high resolution electron beam lithography (EBL). An alternative process is the use of nanoimprint lithography (NIL). NIL seems to be a promising low-cost high-throughput fabrication method of sub-100 nm scale features and has been already successfully applied to fabricate various devices with dimensions in low sub 100 nm region [3], [4]. Nanoimprinting has also proven to be suitable method for fabrication of photonic crystals in IR range [5].

In this work we present fabrication of nanoimprint stamps for photonic crystals in visible and near IR range of spectra. The stamps were fabricated using EBL followed by anisotropic etching of silicon using an advanced silicon etch process. In order to fabricate nickel stamps, the silicon structure was
used as a template for electroplating of nickel. Nanoimprint stamps were evaluated using electron scanning microscopy. Using the stamps, several imprints into suitable resist were performed and imprinted resist structure analyzed. For comparison we present results of fabrication of photonic crystals for visible range of spectra using standard method including EBL and dry anisotropic etching.

2. Design and fabrication techniques of photonic crystals

Due to their simpler fabrication as well as for the purposes of their easier integration, investigations at BESSY limit to 2-dimensional photonic crystals in form of so called photonic crystal slabs [6]. These are waveguides with incorporated photonic crystal pattern. In the waveguide in-plane direction, band gaps arise from the photonic crystal structure, in the third direction, the wave guiding principle limits the out-of-plane propagation. In order to create photonic crystal slabs showing band gaps, the waveguide needs to be patterned with periodic array of holes. Triangular lattice of holes results in largest band gaps [7].

Materials used for photonic crystals need to be transparent in the interested frequency region and possess high refractive index (>2) in order to reveal photonic band gaps after patterning. Especially for visible and near IR region, this presents a significant problem, since only few materials with high refractive index are also transparent in this wavelength spectra. At BESSY silicon nitride (SiN) is being used for photonic crystals in visible range and near-IR range. Its high and over the deposition parameters adjustable refractive index, transparency in visible region and its standard use in microelectronic industry make it an excellent candidate for photonic crystal devices. Triangular hole array with corresponding band gap for silicon nitride is shown in Fig. 1.

![Fig. 1 – Triangular hole array with characteristic dimensions: a - crystal period, r - radius of holes; corresponding photonic band structure of silicon nitride photonic crystal with a=280 nm, r/a=0.4 revealing band gap (highlighted) in visible range of spectra](image)

For photonic crystals in visible range of spectra, structural dimensions in sub 100 nm scales have to be realized as can be seen in Fig. 1, illustrating some of the challenges during the fabrication of photonic crystals for near IR and visible wavelengths.

3. Experimental

PECVD Silicon nitride with a 0.30 µm core thickness and 1.6 µm thick silicon dioxide cladding was used to fabricate photonic crystals for visible and near IR range of spectra. OXFORD Plasmalab 80 Plus with 400 °C Al electrode was used to grow the SiN layer. SiN was deposited using mixture of SiH diluted in N₂ (1:19) and NH₃ at 350 °C, 650 mTorr and power of 20 W. By carefully adjusting the gas ratio, refractive index can be tailored between 1.8 and 2.2.
EBL was realized in LEO1560 equipped with Nanomaker® pattern generator from Interface Company (Moscow) at 30 keV energy. Single layer of 2.2M poly-methyl-methacrylate (PMMA) was used. 10s development was carried out in AR 600-50 (Allresist, Berlin), followed by IPA rinse.

For etching of nanoimprint stamps in silicon, nickel mask obtained by lift-off, evaporating 10 nm thin layer of nickel and dissolving the resist using N,N-dimethylformamid. After RIE nickel was removed using 45% solution of HCl.

For RIE of silicon nitride, alternatively PMMA resist was used directly as a mask. After RIE, remaining resist was dissolved using N,N-dimethylformamid followed by short O2 plasma etching step to remove the organic remains.

RIE was performed using OXFORD Plasmalab 80 Plus. Silicon stamps were etched with SF6 at gas flow rate of 20 sccm, pressure of 15 mTorr, RF power of 20 W and ICP power of 220. For passivation of sidewalls C4F8 with gas flow rate of 15 sccm was used. In order to achieve good thermal contact with ground electrode, substrates were glued to a 4 inch wafer. Etching recipe was optimized in order to achieve vertical and smooth sidewalls and high selectivity.

Two different etching recipes were used to etch silicon nitride. Using nickel mask, modified silicon etch recipe was used by increasing the gas flow rate of C4F8 to 18 sccm. For resist mask, modified silicon dioxide etching recipe was found more appropriate. Etching was performed using a mixture of 25 sccm of Ar and 20 sccm of CHF3 at power of 40 W RF and pressure of 45 mTorr. Both etching recipes were optimized in order to achieve vertical and smooth sidewalls and high selectivity.

After RIE, protecting film of CxFy was removed using O2 etch step.

In case of Ni stamps, silicon stamps were used as electroplating templates. After careful cleaning using O2 plasma and diluted HF (10 %), the silicon stamps were treated in weak Ar plasma for 15 min. prior to be covered with evaporated nickel seed layer of 10 nm thickness. After the nickel deposition, the stamps were moved to the plating solution immediately. Electroplating was performed in nickel sulphamate solution (pH=3.8) at temperature of 50 °C with current density of 1 A/dm² resulting in a growth rate of approximately 12 µm/hour. After the electroplating, silicon template was dissolved using 40 % KOH.

NIL was performed in the labs of microresit technology GmbH [8], using Obducat’s 2,5 “Nanoimprint machine” [9]. To improve the anti-adhesion properties, the stamp was treated using vapor phase deposition of Tridecafluoro- (1,1,2,2)- tetrahydrooctyl-trichlorosilane (F13-TCS) according to method proposed in [10]. NIL was performed using single layer of thermoplastic resist with thickness of 300 nm. Stamping was performed in two steps. In the first step pressure of 50 bar was applied for 10 min. at the temperature of 100°C. In the second step pressure was reduced for 30 min to 15 bar at the temperature of 140°C.

4. Results

Several samples of photonic crystals were first fabricated using standard EBL followed by nickel lift-off and reactive ion etching of silicon nitride. Crystals with hole diameter between 180 and 350 nm and different filling ratio corresponding to band gap frequency between 600 and 1200 nm were fabricated. Fig. 2 shows some examples of fabricated structures. Due to high pattern density and small feature size, advanced algorithm of proximity correction was needed in order to achieve homogenous hole diameter distribution over the entire sample area. Due to careful optimization of the silicon nitride etching process, minimal feature size of less than 50 nm and aspect ratios of over 5 could be successfully realized in our photonic structures.
Nanoimprint stamps were fabricated using EBL and advanced silicon etching process. The silicon etching was optimized with respect to the sidewall verticality and smoothness. Due to the small thickness of nickel mask of only 10 nm, high selectivity had to be reached. Both requirements could be met by properly adjusting the ratio between the etching and passivating gas as well as by finding appropriate plasma power and gas pressure. Fig. 3 illustrates the quality of the optimized silicon etching process. Structures with extremely smooth and vertical sidewalls even on sub 100 nm scale could be fabricated. Features as small as 40 nm with aspect ratios of more than 10 were successfully realized.

Using EBL followed by nickel lift-off and the advanced silicon etch process, several nanoimprint stamps were fabricated. Templates for stamping of photonic crystal structures as well as large area fields of lines and spaces with different line width were incorporated into the design of the Nanoimprint stamp. Fig. 4 and Fig. 5 show some examples of templates on the fabricated stamps. Minimal successfully patterned feature size was 50 nm resulting by overall structure height of 220 nm in an aspect ratio of more than 4. Overall area of one field was approximately 1 x 1 mm. Large field and dense structures needed again very careful proximity correction and dose optimization in order to be able to perform lift-off step and subsequent etching successfully. At this phase, the use of extreme high molecular weight PMMA resist was found very helpful.
In order to convert the silicon stamp into more mechanically stable and robust nickel stamp, electroplating step was used. After cleaning the surface of silicon stamp, the seeding layer was evaporated and nickel electrodeposited. Due to smooth silicon templates, the quality of nickel stamps was very good showing sharp edges and smooth sidewalls. The structure of silicon was fully transformed into nickel. By lines under 100 nm, defects in form of non-sufficient filling of nickel structures were observed. These defects can be probably derived from too high aspect ratio of silicon templates and are subject of further investigations. Fig. 6 shows some examples of Ni stamp structures.
Using fabricated stamps, test imprints were conducted in the labs of *microresist technology GmbH*, Berlin. Imprints using silicon stamps into slightly thicker polymer layer were successful. Lines with width of 50 nm and aspect ratio of 4 as well as the photonic crystal structures could be successfully transferred into the resist. Very few defects were observed throughout the whole field. Apart from features with aspect ratio exceeding 4 – 5, all structures were successfully transferred. Fig. 7 shows examples of imprinted structures.

Fig. 8 shows a detail view on 50 nm lines and photonic structures demonstrating the achieved quality of the imprint. Vertical and sooth sidewalls can be observed. These two parameters are essential for subsequent transfer of the resist pattern into underlying silicon nitride in order to form the photonic crystal.
5. Conclusion
We present a reliable fabrication method of silicon and nickel stamps for NIL of photonic crystals for visible and near IR range of spectra thus allowing the use of this promising mass fabrication technology for fabrication of novel photonic devices. In this work, e-beam lithography, lift-off and RIE process were simultaneously optimized in order to obtain suitable silicon etch process to fabricate silicon stamps with vertical and smooth sidewalls and feature sizes in sub 100 nm range. We successfully fabricated silicon stamps with smallest feature sizes below 50 nm and aspect ratios of more than 5. We also present a simple method of converting the silicon stamp into a more mechanically stable nickel stamp by means of electroplating. Direct comparison of EBL-written structures with the imprinted ones confirms very good quality of fabricated stamps.

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