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Abstract: We reported in this paper the use of annular beam illumination to control the transverse and vertical sizes of two-dimensional dot array structures fabricated with two photon photopolymerization. Dot arrays with an element diameter of less than 300 nm, a lattice spacing less than 1 mm has been produced. At the same time, the structure height exhibits a 1.94 times increase averagely and the height/width aspect ratio is improved from 1.21, for the circular beam illumination case, to 2.76 after using annular beam illumination.

Key words: Aspect ratio, Femtosecond laser, Obstruction, Photopolymerization.

Two-dimensional (2D) sub-micron dielectric arrays have been intensively investigated recently for their potential usage in photonic crystal devices, which are promising candidates for achieving integrated optical circuits because of their bandgap properties [1–4]. Compared with three-dimensional (3D) photonic crystals (PCs), the simple structures of 2D PCs are more suitable for integrated circuit applications such as photonic waveguides and microcavity lasers [5, 6]. 2D PCs are most commonly fabricated with semiconductor materials by using the electron beam lithography technique. However recently, photosensitive resin becomes another alternative material for fabricating PC structures due to the fact that polymer resin is easy to handle and can be doped with other specially featured materials such as quantum dots, nano-particles or photosensitive materials that have a non-linear response to light. One of the powerful techniques used for fabricating PC structures is to utilize multi-photon induced photopolymerization, which can be achieved under the tight focus of an ultrashort-pulsed laser beam by a high numerical-aperture objective lens. Fabrications on 3D PCs with this method have been reported intensively [7–11]. However, due to the difficulties caused by spherical aberration and multiple scattering, fabrication of 3D PCs is limited to small scales, which hampers the potential of achieving complex photonic crystal based devices. To overcome this problem, 2D PCs are good candidates because they may be fabricated with much reduced complexities [12–14].

It is well known that the lattice spacing of a PC structure determines the possible wavelength range of the band gap: the larger the lattice spacing, the longer the wavelength region of the bandgap. Therefore in order to generate a band gap at near infrared or visible region, it is necessary to reduce the transverse size of rods and the lattice spacing to sub-micron range, which is smaller than the diffraction limit. This can be partially achieved by controlling the fabrication laser power near the polymerization threshold [7, 14, 15]. To further reduce the size of the rods and the lattice spacing, point spread function engineering techniques for manipulating laser focal spots are required. Here we introduce a simple point spread function engineering technique, in which case annular illumination is used instead of circular illumination. The introduction of annular illumination not only reduces the lateral sizes of 2D dot array structures, but also simultaneously increase the vertical size of the rods. This results in a significant improvement in the height/width aspect ratio of the fabricated rods, which provides better light confinement in the vertical dimension.

In this paper, we demonstrate the using of annular beam illumination in the fabrication of two-dimensional dot arrays with the two-photon photopolymerization technique. To introduce annular beam illumination, we co-axially insert different sized opaque disks into the collimated light beam path in our experiment. The focal spot under annular beam illumination is
elongated in the axial direction and at the same time is narrowed in the lateral direction, which results in a large height/width aspect ratio. By varying the size of the obstruction disk, the aspect ratio (height and width) of the fabricated rods can be manipulated.

2. Fabrication of 2D dot array structures under circular and annular beam illumination

A schematic diagram of the experimental setup is shown in Fig. 1a. A pulsed laser (SpectroPhysics Mai Tai) with a wavelength of 760 nm and a repetition rate of 80 MHz is used as the light source. The beam expansion system leads to a collimated illumination beam over the back aperture of an oil immersion objective (Olympus: 60°/C 2 NA = 1.25). A diaphragm matches the beam diameter with the back aperture of the objective. The reflected light from the sample is split by a dichroic mirror into a confocal detection arm, which allows for the accurate detection of the interface between the cover slip and the resin. The sample is made of a thin film of Norland optical adhesive, spin coated on a cover slip before exposure by the pulsed laser. The Norland resin has only one absorption band between 350 and 380 nm in the UV region, which allows for two-photon excitation at a wavelength of 760 nm. The shutter, the scanning stage and the PMT are all controlled by a computer and the fabrication process is monitored in situ by a CCD camera. After the fabrication process, the unpolymerized resin is removed and only the fabricated structures remain on the cover slip.

The fabricated structures were characterized with both a scanning electron microscope (SEM) and an atomic force microscope (AFM, NT-MDT, solver LS). Although an SEM is a commonly used instrument to measure the dimensions of nano-structures and has the advantage of showing the morphology of the structures, the SEM in our experiment cannot provide the accurate height information even by a side view. As a result of that, AFM is applied in our experiment. Compared with SEM, AFM is more functional in our characterization because it can provide both vertical height and lateral dot diameters (represented by the full width at half maximum) simultaneously.

Fig. 1b shows an SEM image of the 2D dot array structure fabricated with an exposure power of 10 mW and an exposure time of 100 ms. The top view SEM image reveals that the diameter of each dot is less than 300 nm and the lattice spacing is approximately 900 nm. By carefully controlling the laser power and the exposure time, the dot size can be optimized. Fig. 2(a–d) present images of the structures fabricated at the same laser power of 40 mW but different exposure times of 25, 100, 300, 700 ms, respectively. The structures are scanned by an SEM at a tilting angle of 50 degrees with respect to the normal direction to the cover slip. It is noted that when the exposure time increases from 300 ms to 700 ms, the height of the structures increases from 0.9 μm to 1.02 μm and the corresponding width changes from 0.71 μm to 0.82 μm.

Fig. 2. SEM images of dot arrays fabricated with the same power (40 mW), but different exposure time: a) 25 ms; b) 100 ms; c) 300 ms and d) 700 ms. Scale bar 2 μm.
Fig. 3 summarizes the dimensions of the fabricated structures as a function of the exposure time for three power levels. As is shown in fig. 3a, the response of the resin to the laser shot can be divided into three zones, the threshold zone, the saturation zone and the damage zone. Two-photon induced excitation can only trigger photopolymerization in the region where exposure energy is higher than the threshold. It is noted that the dot size increases rapidly with the increasing of exposure time until it reaches the saturation zone, where the dot size does not change appreciably when the exposure time prolongs. If the exposure time is too long or the intensity is too high, the high temperature and pressure in the focal region lead to bulk solidification, which makes the fabrication results uncontrollable. We call this region the damage zone. Similarly, three regions can also be derived in the dependence of the dot height on the exposure time (fig. 3b). The dependence of dot dimensions on the laser intensity and exposure time allows the generated structures to be controlled accurately. Fig. 3c depicts a plot of the height/width aspect ratio with respect to exposure time. It can be seen that the aspect ratio almost keeps constant as the exposure time and the illumination intensity increase except for the region close to the threshold. This phenomenon can be well explained by the fact that the focal spot under the full illumination of a given objective lens is fixed [7]. The only exception region occurs close to the threshold is due to the material response. The aspect ratio is approximately 1.21 for all the three exposure power levels in the saturation zone, which is smaller than the aspect ratio of 1.7 for a multiphoton excitation point spread function for NA = 1.25 and λ = 760 nm. As a result of the constant aspect ratio of the multiphoton excitation point spread function, it is impossible to manipulate the dot height and width independently using circular beam illumination.

Such a difficulty can be overcome by utilizing annular beam illumination. It is well known that an annular lens has a larger focal depth and a slightly smaller transverse focal spot size than a circular lens [16], which offers the possibility to provide a considerable larger aspect ratio when the size of the obstruction disk is large enough. Fig. 4 shows the comparison of the intensity distribution in the focal region between a circular lens and an annular lens of ε = 0.9 (ε is the ratio between the size of the obstruction disk and the size of the back aperture of an objective). It is clearly seen from the plot that the focal length of the annular lens is 6.5 times of that under circular beam illumination; however, the transverse dimension is only 73 percent of the corresponding value for a circular lens, which leads to a more than 8 time increase in the axial/transverse aspect ratio.

Fig. 3. Dependence of a) the dot width; b) dot height, and c) aspect ratio on exposure time for a circular lens (ε = 0).

Fig. 4. Light distribution a) in the focal region of a circular lens and b) an annular lens (ε = 0.9) for NA = 1.25, where u and v are the normalized axial and transverse optical coordinates [16], respectively.
We experimentally demonstrate these phenomena by introducing a circular opaque disk obstruction, coaxially into the light path, as showed in fig. 1, which induces annular beam illumination over the back aperture of the lens. Fig. 5 presents two AFM images of two lines of dots fabricated with obstruction sizes of $\varepsilon = 0.34$ and $\varepsilon = 0.72$, respectively. It can be clearly seen from fig. 5b that the rods show obvious decrease in the transverse diameter and increase in the axial height.

The power coupled into the objective during the fabrication is kept constant at 30 mW for all three obstruction cases shown in fig. 6. It is noticed from fig. 6a that a noticeable decrease in dot width is observed. For example, the dot width is reduced averagely in the saturation zone from 0.82 $\mu$m to 0.69 $\mu$m when the obstruction disk ($\varepsilon = 0.72$) is inserted. It is also seen from fig. 6b that a more significant improvement in the dot height can be achieved under annular illumination. For example, the dot height increased averagely in the saturation zone from 1 $\mu$m to 1.94 $\mu$m when the obstruction disk ($\varepsilon = 0.72$) is inserted. Consequently, the improvement in the aspect ratio is more significant after introducing annular beam illumination (fig. 6c), i.e. the aspect ratio is improved from 1.21 for circular illumination to 2.76 for annular illumination ($\varepsilon = 0.72$).

Fig. 5. AFM images of dot lines: a) $\varepsilon = 0.34$; b) $\varepsilon = 0.72$.

Fig. 6. Dependence of a) the dot width; b) dot height and c) aspect ratio on exposure time for annular lenses with different epsilons. d) Dependence of the focal volume on epsilon (dashed line); and theoretical (solid line) and experimental (dots) aspect ratio as a function of the obstruction size.
Theoretically, there is no limitation for increasing the aspect ratio, as shown in fig. 6d (solid line). However in practice, it will be limited by other contributing factors such as the laser power, the quality of the beam profiles, the objective lens and the material response. Nevertheless a more significant improvement can be expected if a larger obstruction disk of $\epsilon > 0.9$ is used. The experimental dependence of the aspect ratio on $\epsilon$ is also shown in fig. 6d (dots). Although the absolute values from experiment are overall smaller than theoretical values, the trends of the experimental and the theoretical curves are in a good agreement, which confirms the feasibility of manipulating beam profiles to improve the aspect ratio. It is also noticed that for a given illumination intensity, the exposure time that is required to reach the saturation zone for a larger obstruction case is longer than that when a smaller obstruction disk is inserted. This is due to the increase of the focal volume under annular illumination caused by the elongation of the focal spot. The dependence of the focal volume on the obstruction disk size is also shown in fig. 6d (dashed line). It is demonstrated that higher laser power is required for a larger obstruction disk sizes due to the increase of the focal volume. This is an understandable feature since the polymerization volume at each shot also increases proportionally.

3. Conclusion

Using point-by-point two-photon photopolymerization we have fabricated height and width controllable two-dimensional dot array structures with a commercially available optical resin. The lateral dot size can be controlled by changing the exposure power or time, while the vertical dimension can be fully manipulated by using annular beam illumination. Dot arrays with an element diameter of less than 300 nm and the lattice spacing of less than 900 nm have been successfully produced. The dependence of the dot dimensions on the obstruction size is investigated and it has been found that after inserting the largest sized obstruction in our experiment, the aspect ratio can be increased averagely by 2.28 times from 1.21 to 2.76. Combining the two methods allows for the full control of the parameters in three dimensions in nano-fabrication.

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References