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Fabrication of column shape two dimensional photonic crystals: double developments in holographic lithography process

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Abstract: A simple and cost effective method to fabricate column shape two dimensional photonic crystals by holographic lithography technique using double development has been reported. In the process two dimensional big-dot-type periodic structures have been generated directly on positive photoresist, and then transferred to the substrate through the Si_3N_4 hard mask. By using double development, the photoresist exposed to high and medium intensity can be developed away effectively, while those exposed to low intensity can still be preserved. The period of the two dimensional structures can be easily controlled by adjusting the angle of two incident beams. The structures in a large period range over a relative large area have been generated with good uniformity and reproducibility. The processing parameters are discussed in detail.

Key words: holographic lithography; two dimensional photonic crystals; column shape; double developments

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全息光刻和二次显影法制备柱形二维光子晶体

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摘要: 采用全息光刻和二次显影的方法制备了柱形二维光子晶体.在此过程中,二维点状的周期结构首先在正性光刻胶上直接形成,然后经由Si_3N_4硬掩模转移到衬底材料上.利用二次显影的方法,曝光强度最强和曝光强度中等区域的光刻胶能够被同时充分显影,而曝光强度最弱区域的光刻胶则可以完全被保留下来.通过调节入射角,可以方便地调节二维结构的周期.利用此方法,在相对较大的面积上制备了不同周期的二维结构,二维结构具有很好的均匀性和重复性.文章对有关的工艺参数进行了详细讨论.

关 键 词: 全息光刻;二维光子晶体;柱形;二次显影

中图分类号: TN2 **文献标识码**: A

Introduction

Two dimensional photonic crystals (2D-PCs) have attracted great attentions due to their specific photonic bandgap. It is possible to design 2D-PCs to implement functions such as coupling surface of 2D-gratings, waveguides, resonators and so on[1-4]. 2D-PCs have been fabricated by compound semiconductors to improve the performance of active components operating in near-or mid-infrared region[5, 6]. Both electron beam lithography and holographic lithography are the common techniques to fabricate periodic structures as 1D

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gratings or 2D-PCs[7,8]. Electron beam lithography has the merit of high precision in controlling the feature size of periodic or nonperiodic structures, but with disadvantages of lower throughput and higher cost. As an alternative, holographic lithography can generate periodic structures on a relative large area in a short time with simple equipment. In this way, holographic lithography has become an attractive method. It has been used successfully to fabricate 1D surface grating for mid-infrared quantum cascade laser[9,10], but still unpopular for 2D structures. Generally, there are two different ways to generate two dimensional interference patterns by using holographic lithography: multiple exposure of interference patterns generated by two beams[11] or single exposure of an interference pattern generated by multiple beams[12,13]. The contrast of holographic patterns could not be very high regardless of single exposure or multiple exposure. But in relation to single exposure, multiple exposure has the advantages of simpler equipment and higher contrast in generating the interference patterns[11]. The principle of multiple exposure is based on the interference of two coherent lights to form a horizontal standing wave for grating patterns, which can be recorded on the photoresist. By using multiple exposure, different kinds of laser intensity are formed in the exposure area. With single development, photoresist exposed to high intensity can be fully developed away, while those exposed to low intensity may not be removed effectively. Therefore it is need to carefully control the exposure dose and developing time to generate dot-type 2D periodic structures by using positive photoresist and single development directly[14]. Also by using single development, the dots are often smaller than half of the period and the sizes of the dots are hard to control[15].

In order to solve these problems, we exploit a new developing method by changing the developing steps from one step to two steps. By using this unique developing method, photoresist exposed to low intensity can also be fully developed away, and big-dot-type periodic structures can be generated directly with easily controlled exposure dose and developing time. Based on this method, the column shape 2D-PC have been fabricated by using reactive ion etching (RIE) of Si_3N_4 hard mask and inductively coupled plasma (ICP) etching of the substrate.

1 Experimental details

The experiment setup of holographic lithography system is shown in Fig. 1. It consisted of a laser source, two reflective mirrors, a beam expander, a collimating lens and a sample stage. A He-Cd laser emitting at 441.6 nm was used as the exposure source, in which the laser light was expended and collimated into a TE-polarized beam of size 1.04 mm with output power of 147 mW, the coherence length of the laser was 10 cm. Two reflective mirrors were used to adjust optical path and arrange optical equipment conveniently on the available optical table. In order to transmit only the fundamental Gaussian output of the laser, a spatial filter consisting of objective lens and pinhole was used to filter and enlarge the beam. This spatial filter was built in adjustments in X, Y direction on the pinhole mount and in Z direction on the focusing objective. Through this Gaussian beam expanded, the laser beam had been changed in three ways. The diameter of the beam and radius of the phase front were both increased, whereas the intensity of the beam decreased. Because of the Gaussian intensity profile, a more uniform intensity distribution over the exposed area was created. In the sample stage, the reflective mirror and sample holder plate were assembled together with a fixed angle of 90°, so the angle of incidence of the reflected beam was always equal to the angle of the di-

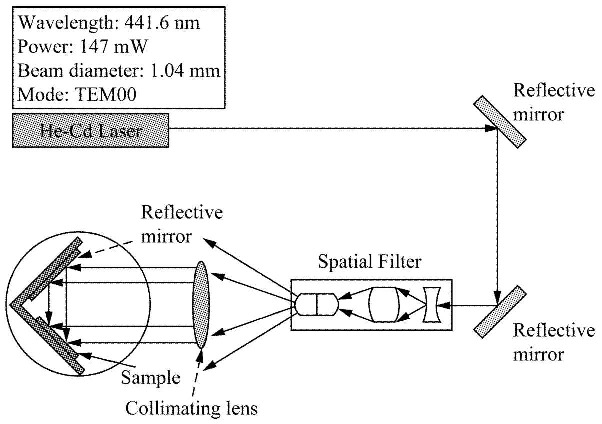

Fig. 1 Schematic diagram of the holography lithography system.
图 1 全息光刻系统结构示意图

rectly incident beam. By simply adjusting the stage orientation with respect to the direction of the laser beam, the period Λ of the patterns could be easily controlled based on the equation $\Lambda = \lambda/2\sin\theta$, where λ is the laser wavelength and θ is the half angle of the two incident beams. As an example, by increasing θ from 6.33° to 12.75°, the period Λ can be decreased from 2 μm to 1 μm according to the equation.

In the experiments, InP(100) substrate was used and cut into 2 × 2 cm^2 samples. After cleaning, 100 nm Si_3N_4 layer used as hard mask for transferring dot-type periodic structures to the substrate was deposited on the samples by plasma enhanced chemical vapor deposition (PECVD). Subsequently a diluted positive photoresist (SHIPLEY SPR6809) with thickness of 180 nm was spin coated on the Si_3N_4 layer. Thin photoresist layer was necessary due to the low beam intensity of this holographic lithography process. After spin coating the samples were soft baked, and then transferred to the holography lithography stage for exposing. The first exposure was carried out to create a 1D grating pattern, afterwards the samples were rotated 90° for the second exposure to create a square lattice pattern. The exposure time of the two exposure were both 40 s. After double exposure, a post exposure bake was performed to reduce the standing wave effects. The samples were developed in FHD 320 developer, which was 2:1 diluted with deionized water. Single development and double developments were carried out. For the process of double developments, the samples were developed for a short time at first, and then rinsed by deionized water and blown dry with nitrogen gas. Subsequently, the samples were developed once again without stir to reach the final pattern. The developer temperature was kept at room temperature around 20 ℃. After double developments, RIE of Si_3N_4 mask was performed using CF_4 with radio frequency (RF) power of 100 W. The etch time was 60 s at etching rate of 110 nm/min. Subsequently the photoresist on Si_3N_4 layer was washed away. Then the periodic structures were transferred into InP substrate by ICP etching using $CH_4/H_2/Cl_2$. The etch time was 50 s at etching rate of 900 nm/min. Finally, the Si_3N_4 hard mask was etched away using buffered HF and periodic structures were formed on the substrate.

2 Results and discussions

Because of the double exposure with a rotation of 90°, perfect periodic square structures can be obtained by simulating the theoretical calculation of intensity distribution of two-beam interference[14]. On the exposure areas three different kinds of laser intensity were formed as shown in Fig. 2(a). "0" was the region with the minimum laser intensity, "1" was the region with the medium laser intensity and "2" was the region with the maximum laser intensity. In order to generate dot-type periodic structures, the photoresist on the "1" and "2" region must be fully developed away simultaneously. Usually the samples were just developed for once, so photoresist on the "2" region with the high laser intensity could be easily developed away, while photoresist on the "1" region could not be developed away effectively due to the lower laser intensity. In order to develop away the resist on the "1" region fully, two ways of increasing the exposure dose or increasing the developing time had been attempted. However, if the exposure dose or developing time are increased, photoresist on the "0" region may also be developed away. Because of the relatively low contrast of the holographic interference pattern, this problem could not be solved merely by changing the exposure dose[15].

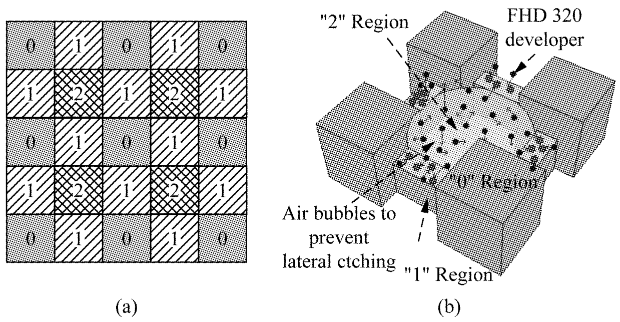

Fig. 2 (a) Schematic of three different kinds of laser intensity on the exposure areas; (b) Schematic diagram of double development
图 2 (a) 三种不同曝光强度的区域示意图;(b) 二次显影原理示意图

A double developing method has been demonstrated to solve these problems. The schematic diagram of double development is shown in Fig. 2(b). In this process the samples were developed for a short time at first. After the first development, photoresist on the "2" region could be fully developed away, while some photoresist on the "1" region were still left. In the second developing step, because of the small sizes of the holes, the developer outside the holes generated a pressure preventing the developer from flowing into the holes, leaving air bubbles inside region "2". The pressure was given by $p = s/r$, where s is the surface tension of developer and r is the radius of the hole. For a hole of 1 μm diameter the surface tension reaches ~0.7 atmosphere for water. In this case because the developer couldn't soak and flow into the holes, these could prevent the developer from etching laterally on the "2" region. As a result, only photoresist on the "1" region could be developed away further, while photoresist on the "0" region could still be preserved. Finally, big-dot- type periodic structures could be directly generated on the positive photoresist.

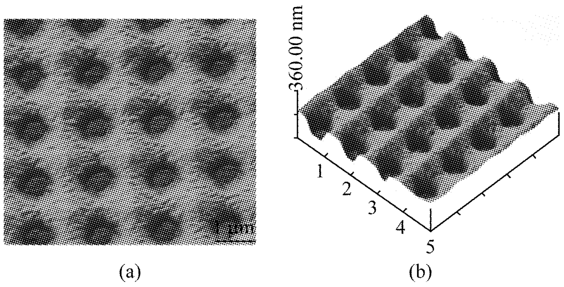

Fig. 3 AFM images of the photoresist after single development for 20 s
图 3 单次显影 20 s 后光刻胶的 AFM 图像

Figure 3(a) and(b) show the AFM images of the photoresist on the sample after single development for 20 s. In this case hole-type periodical structures were generated with the period of 1 μm. We also tried to increase the developing time to 30 s, as a result the holes expanded severely and some photoresist on the "0" region were fully developed away. Compared with single development, Fig. 4 show the AFM images of the photoresist on the sample after first and second development. Fig. 4(a) and(b) show the AFM images after the first development of 15 s. Hole-type periodical structures were generated with the period of 1 μm and the sizes of the holes were a little smaller than the holes as shown in Fig. 3. Fig. 4(c) and(d) show the AFM

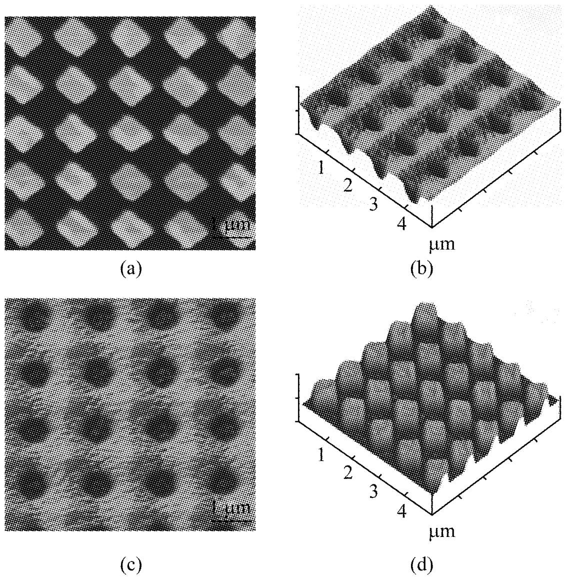

Fig. 4 AFM images of two development processes. (a) and(b): after first development; (c) and (d): after second development
图 4 二次显影过程的 AFM 图像(a)和(b): 第一次显影后;(c)和(d): 第二次显影后

images after second development of 15 s. From Fig. 4 (c) and(d), it could be seen that, big-dot-type periodical structures with the period of 1 μm had been generated effectively with good uniformity.

After double development, the dot-type periodic structures were transferred into InP substrate by using RIE of Si_3N_4 hard mask and ICP etching of the substrate. Fig. 5 shows the SEM images of the samples. In Fig. 5(a) the period of the structures is 1 μm with the sizes and heights of the column being 500 nm and 700 nm respectively. By changing the incident angle, column shape 2D-PC with period of 2 μm have also been fabricated as shown in Fig. 5(b). The sizes and heights of the column are about 1 μm and 703 nm respectively. A statistical study of the uniformity of the

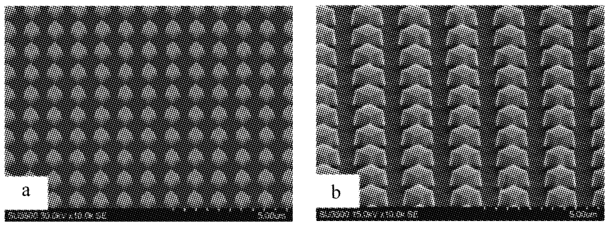

Fig. 5 SEM images of column shape 2D-PC on InP substrate with different period: (a) period of 1 μm; (b) period of 2 μm
图 5 InP 衬底上不同周期的柱形二维光子晶体的 SEM 图像: (a) 周期 1 μm;(b) 周期 2 μm

2D-PC with respect to the lattice constant and column diagonal in the area of 12 × 12 μm^2 has also been carried out. According to the statistical study, the nonuniform of the lattice constant and column diagonal with period of 1 μm is 2.9% and 3.6%, while the nonuniform of the lattice constant and column diagonal with period of 2 μm is 3.4% and 4.9%, respectively.

3 Conclusions

In conclusion, column shape 2D-PC on InP substrate have been fabricated by using holographic lithography technique. Adopting double development process, big-dot-type periodic structures have been generated on positive photoresist directly with easily controlled exposure dose and developing time. Si_3N_4 hard mask is chosen to transfer the dot-type periodic structures into the substrate due to its high selectivity in dry etching process. The period of the structures can be easily controlled by adjusting the angle of the two incident beams. 2D-PC samples in an area of larger than 2 × 2 cm^2 with good uniformity and reproducibilityhave been fabricated successfully, validated the feasibility of this unique process in the applications of various devices.

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