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Achieving Wide Band Gaps and a Band Edge Laser Using Face-Centered Cubic Lattice by Holography

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1. Introduction

Complete band gaps (CBGs) is an important research topic in the study of photonic crystals (PCs). In 1990, K.M.Ho *et al.* demonstrated theoretically that a diamond structure possesses CBGs [1], since then, great interests were attracted in fabricating three-dimensional (3D) PCs in order to obtain CBGs. Several methods were reported, and CBGs were achieved in the range of microwave or sub-microwave. Theoretical analysis shows that although CBGs can be obtained by diamond structure, a strict condition has to be satisfied, i.e., the modulation of refractive index of the material used should be larger than 2.0 [2, 3]. So, many scientists paid their attention to find the materials with high the refractive index. Some of them tried to fill the templates with high refractive index materials to increase the modulation of the refractive index [4-6], and CBGs were obtained. However, the achieved CBGs in 3D PCs were mostly in microwave and infrared regions [5, 7-9].

Holography is a cheap, rapid, convenient, and effective technique for fabricating 3D structures. In 1997, holographic technique was introduced to fabricate the face centered cubic (fcc) structure [10]. Campbell *et al.* fabricated fcc structure in terms of holographic lithography [11]. Several authors reported their works in this topic [2, 3, 7, 12]. Toader and John also showed theoretically a five-beam “umbrella” configuration for synthesis of a diamond photonic crystal [13]. Because both the value and modulation of the refractive index of the holographic recording materials are commonly low so that the PCs made directly by holography would not have CBGs. For example, the epoxy photoresist is generally used in holographic lithography [11, 14-16], but, its refractive index is $n=1.6$ which is a little bit too low. They may, however, be used as templates for the production of inverse replica structures by, for example, filling the void with high refractive index and burning out or dissolving the photoresist [11], and a good work was done by D.C.Meisel [17] etc. But, to find the materials with large refractive index is not easy, and the special techniques needed are very complicated and expensive. It limits the applications of PCs, especially for industrial productions.

Although band gaps can be broadened by means of multi-structures [18], CBGs in visible range had not yet been achieved, especially by using the materials with low refractive index. Therefore, it is a big challenge to fabricate 3D PCs possessing CBGs in visible range by using

the materials with low refractive index. Although some efforts had been made [18], CBGs in visible range had not yet been achieved by using the materials with low refractive index. Therefore, it is a big challenge to fabricate 3D PCs possessing CBGs in visible range by using materials with low refractive index, though it is greatly beneficial for future PC industry. As the first step for achieving this, it is important to obtain very wide band gaps. In the previous investigations [19, 20], it was evidently shown that the anisotropy of a photonic band gap in a two-dimensional photonic crystal is dependent on the symmetry of the structure, and as the order of the symmetry increases, it becomes easier to obtain a complete band gap. One would naturally ask whether or not such a method is applicable to 3D PCs. In view of this question, in this paper we proposed a holographic method for fabricating triple-diamond structure by using materials with low refractive index, and the features of the band gap in such a complex diamond structure were then studied experimentally.

On the other hand, as a new kind of laser source band edge laser has the significance in the applications of PCs, and is helpful for further understanding of the characteristics of PCs. Therefore, it attracts great attentions. In this paper, the emission properties of the band edge laser in a fcc PC fabricated by holography was studied. The different emission properties of the band edge laser pumped by picosecond and nanosecond pulses were investigated experimentally. Some intriguing phenomena were revealed.

2. Complex diamond structure

2.1 Experimental method and the samples

It is known that a cell of diamond structure consists of two cells of fcc structures, and the two cells have a distance of one quarter of the diagonal length of the cell along the diagonal line. Therefore, the diamond structure can be implemented by holography through two exposures: an fcc structure is recorded in the first exposure, then, after the recording material is translated one quarter of the diagonal length along the diagonal line of the fcc structure a second exposure is made to record another fcc structure. In this way, a PC with diamond structure can be obtained.

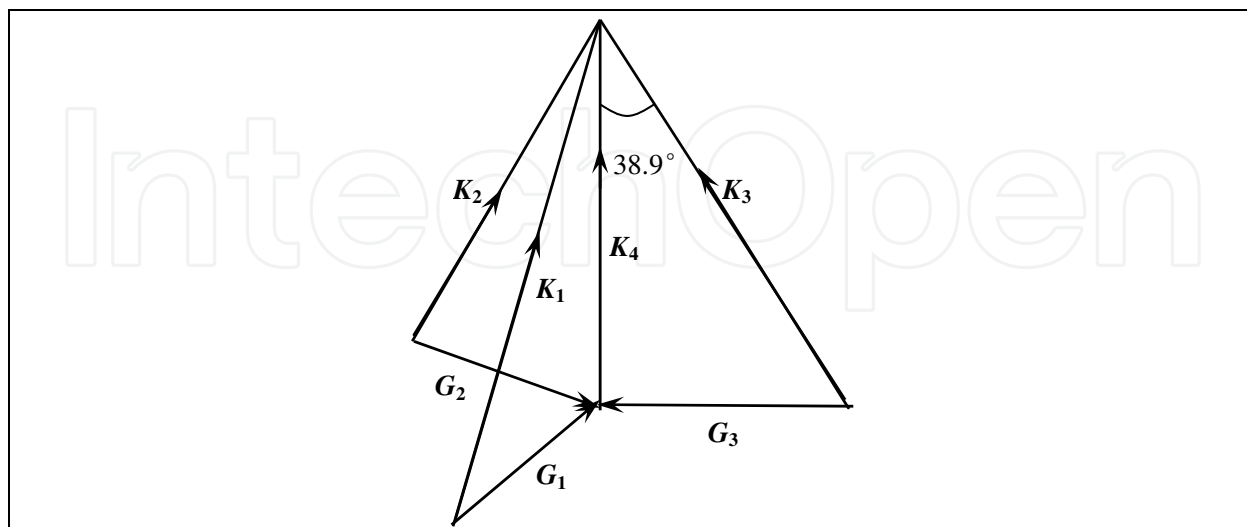


Fig. 1. Schematic optical layout for recording fcc structure.

Fig. 1 shows schematically the optical layout in our experiments. Four beams splitted from a laser beam were converged to a small area. The central beam was set along the normal

direction of the surface of the plate, while the other three outer beams were set around the central beam symmetrically with a angle of 38.9° with respect to the central one. The laser used was a diode pumped laser working at 457.9 nm with line width of 200 kHz (Melles Griot model 85-BLT -605). The polarization state of each beam was controlled to achieve the best interference result [21, 22]. The recording material was mounted on an one-dimensional (1D) translation stage driven by a stepping motor with the precision of $0.05\mu m$ per step. Also, the translation stage was mounted on a rotary stage. The holographic recording material used was dichromated gelatin (DCG) with refractive index $n = 1.52$. The maximum value of its refractive index modulation Δn can reach more than 0.2, but generally is around 0.1. It is a very small value for obtaining wide band gaps. The thickness T of the material was $36\mu m$. The DCG emulsion was coated on an optical plate glass with flatness of $\lambda/10$ and without any doping.

It is well known that there are several directions with high symmetry in the first Brillouin zone of an fcc structure [22]. During our experiments, two exposures as mentioned above (in paragraph 4) were made firstly in $\Gamma-L$ ([111]) direction to get a diamond structure. And then, a second, even a third diamond structure was implemented by changing the orientation of the recording material through rotating the rotary stage to other symmetric directions i.e. the direction of $\Gamma-X$ ([100]) and/or the direction of $\Gamma-K$ ([110]) in the first Brillouin zone of the first fcc structure. In this way, photonic crystals with one, two even three diamond structures were fabricated. It should be pointed out that when the second or the third diamond structure was recorded, the angle between any two beams should be changed to guarantee all the beams inside the medium satisfy the relation shown in Fig. 1 so that the standard diamond structures can be obtained.

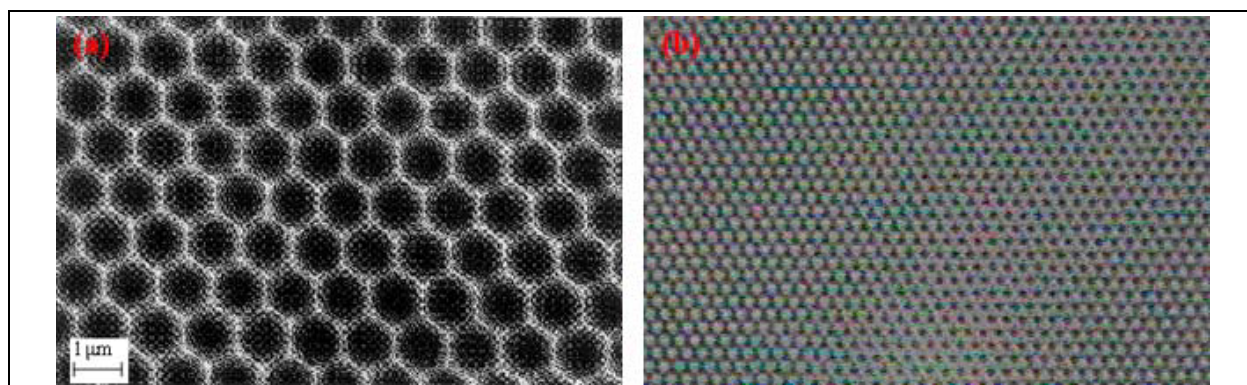


Fig. 2. (a) SEM image of an fcc structure made with photoresist of $1\mu m$ thickness. (b) Microscopic image of an fcc structure made with photoresist of $1\mu m$ thickness.

Since the hologram made with DCG is a phase hologram, there is only the distribution of the refractive index and no plastic effect inside the hologram, so, SEM image can not be obtained. To verify the structure in the hologram, a same fcc structure was implemented using same optical layout but with the photoresist of $1\mu m$ thickness. Fig. 2 shows the structure at [111] plane, (a) shows the SEM image and (b) shows the optical microscopic image taken by a CCD camera mounted on an $1280\times$ microscope, the pixel size of the CCD is $10\mu m$. The theoretical calculated diamond lattice formed by 8 beams as mentioned above is shown in Fig.3. It should be pointed out that the shape of the maximum of interference appears as an American football. In this figure the vertical bar shows a gradient of bottom to

top corresponding to the outer to inner region of a cross-sectional cut of a football. The gradient of the outer surface is related to the value of the vertical bar. Whether a diamond lattice can be obtained depend on how extend the two footballs. If the major parts of the two footballs are overlap the structure can not considered as a diamond lattice. If the two footballs can be recognized independently, it means that a diamond lattice is formed. In our experiments, to get optimal interference result the absorption coefficient α of the material was chosen as $\alpha = 1/T$ [23]. According to previous works [23], it is well known that for holographic recording materials the relation between the optical density and the exposure is nonlinear. The holographic exposure can be controlled in the region of stronger nonlinear dependence between optical density versus exposure as discussed previously [24]. In this way, two fcc lattice can be recognized so that the structure can be considered as a diamond lattice.

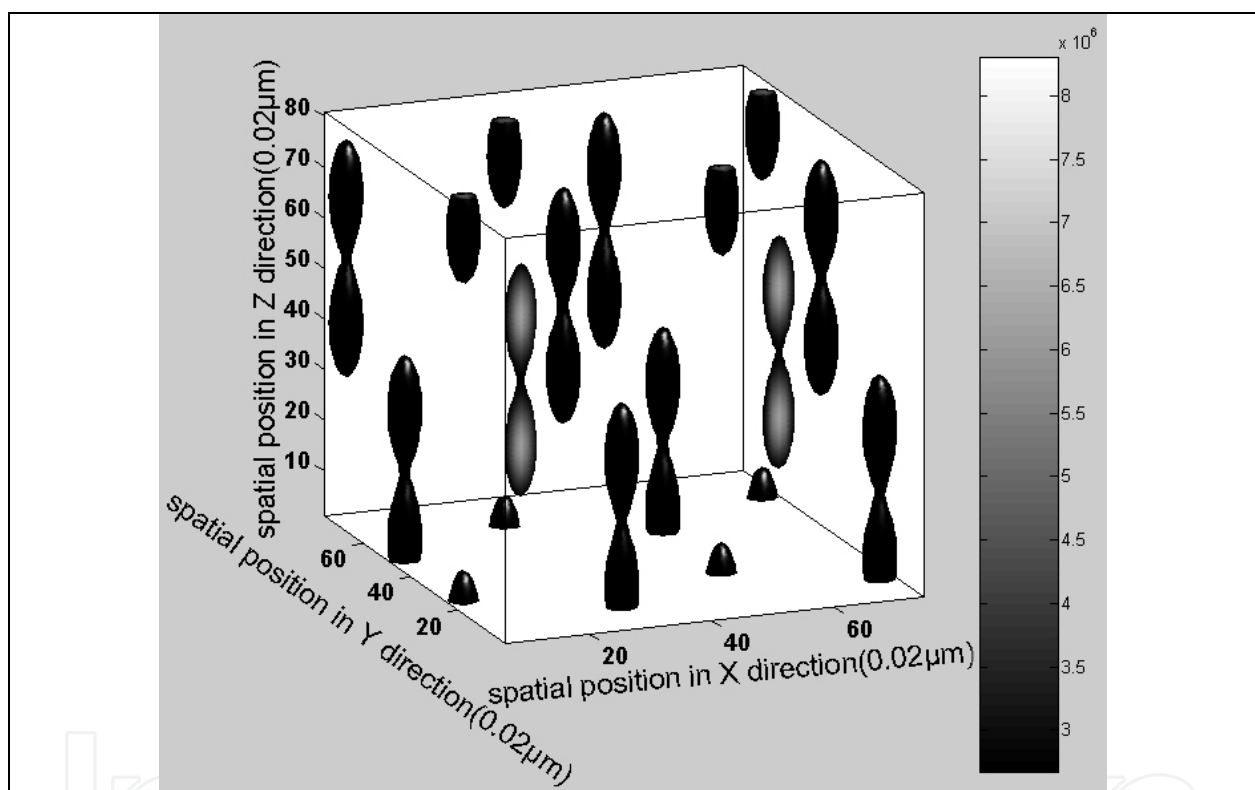


Fig. 3. Diamond lattice formed by two fcc lattice in nonlinear exposures. The vertical bar shows a gradient of bottom to top corresponding to the outer to inner region of a cross-sectional cut of a football. The gradient of the outer surface is related to the value of the vertical bar.

The transmission spectra of PCs with diamond structure made with DCG were measured. In the measurements, a J-Y 1500 monochrometer was employed. To minimize the energy loss from reflection at large incident angle, a right triangular prism was used. The measuring set-up geometry is shown in Fig. 4. The measured range of the wavelength was 390 nm~800 nm. The [1 1 1] plane of the PC measured was set firstly at an arbitrary orientation. The incident angle of the collimated white light beam was changed from $0^\circ \sim \pm 90^\circ$ (the incident angle is in the plane parallel to the paper surface), Then, the PC was rotated to other orientations (the orientation angel is in the plane perpendicular to the paper surface), and the incident

angle of the collimated white light beam was still changed from $0^{\circ} \sim \pm 90^{\circ}$. The orientation angel and the incident angle are the angles φ and θ in a spherical system indeed.

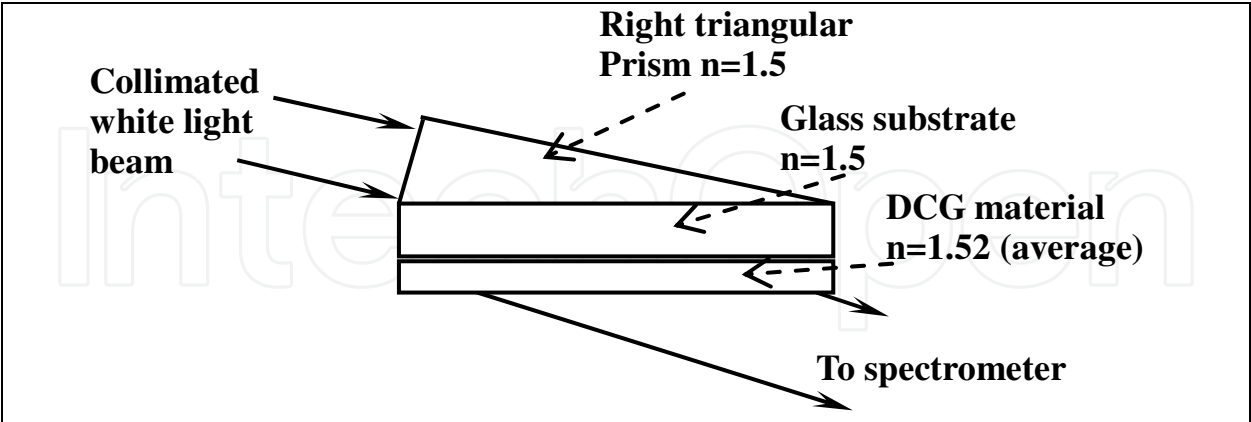


Fig. 4. Set-up geometry for measuring transmission spectra of photonic crystals.

2.2 Measured spectra and discussions

The measured transmission spectra of a fabricated triple diamond PC are shown in Fig. 5(a), (b) and (c). (a), (b) and (c) represent measured results at different orientations of the sample. And each curve in the figures gives the spectrum measured at a certain incident angel and a certain orientation. Different curves give results at different incident angels and orientations. Thus, whether or not the common gap exists can be determined by the overlap of all the curves. It can be seen that, for the PC with three diamond structures (6 fcc structures) the width of the band gaps reached 260 nm, the ratio between the width and the central wavelength of the gaps reached 50 %, and there is a common band gap with a width about 20 nm at 450~470 nm in a range of 150° of the incident angle. The common gap achieved using DCG with very low refractive index exists in a wide range which reaches 83 % of the 4π solid angle. Although a complete band gap for all directions is not obtained in our experiments, it is significant to achieve such a wide angle band gap by using a material (DCG) with very low refractive index, because this angular tuning range can satisfy most applications in practice.

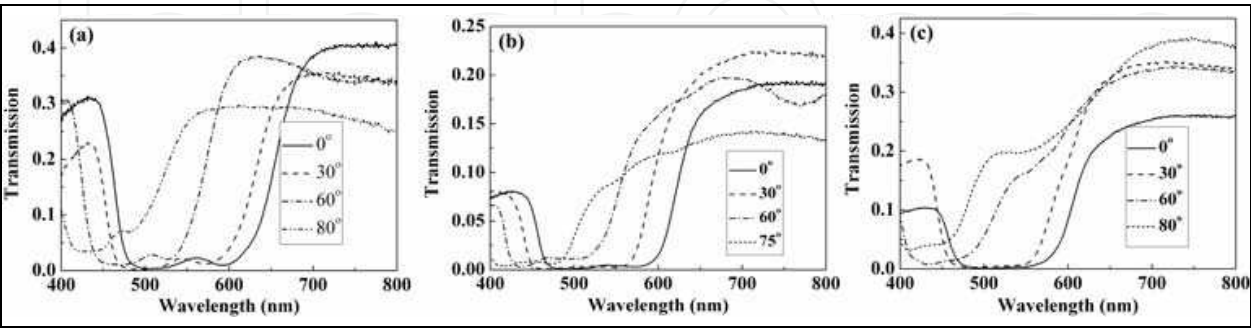


Fig. 5. Measured transmission spectra of PC including 3 diamond lattices recorded by holography in $\Gamma - L$, $\Gamma - X$ and $\Gamma - K$ directions of an fcc lattice respectively. (a), (b) and (c) correspond to the measured results. The angles appeared in (a), (b) and (c) are the incident angle which is in a plane parallel to the paper surface. (a) Orientation angle is 0° . (b) Orientation angle is 30° . (c) Orientation angle is 90° .

This interesting result comes from complex diamond structure. The photonic crystal with triple diamond structures is an actual multi-structure not a stack of several same structures. The $[111]$ direction of the first diamond structure is actually the $\Gamma-X$ direction of the second diamond structure and the $\Gamma-K$ direction of the third diamond structure respectively. When a beam incidents normally on the PC, the beam is in the $[111]$ direction of the first diamond structure. When the incident angle changes, the beam deviates from the $[111]$ direction of the first diamond structure but tends to approach the $[111]$ direction of the second or the third diamond structure. Therefore, though the incident angle changes, the light beam keeps always around the $[111]$ direction of the other diamond structures. The narrow region in the $K-\omega$ dispersion relation of a diamond lattice may be expanded by other diamond lattices.

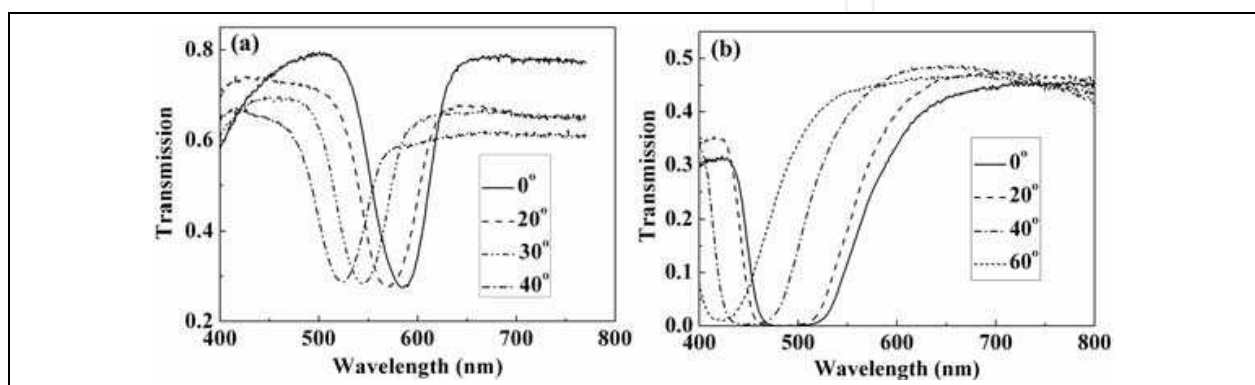


Fig. 6. Measured transmission spectra of a PC at different orientations and incident angles recorded with one and two diamond lattices. (a) Measured transmission spectra of a PC at different orientations and incident angles recorded with single diamond lattice recorded by holography in $\Gamma-L$ direction of an fcc lattice. (b) Measured transmission spectra of a PC at different orientations and incident angles recorded with two diamond lattices recorded by holography in $\Gamma-L$ and $\Gamma-X$ directions of an fcc lattice respectively.

The advantage of a multi diamond structure can be seen more clearly from the comparison to those with one and two diamond structures. Fig. 6 gives the measured transmission spectra of PCs with one and two diamond lattices at a certain orientation respectively. It can be found that with the increase of the number of diamond lattice the width of band gaps of a PC can be broadened effectively. For two diamond structures, the space angle range of the common gap is 80° , and for the single diamond structure the common gap becomes 40° . This means that the common gap can be enlarged or created by means of multi-structures. The physical origin of such a phenomenon can be understood from the change of structure symmetry. With the increase of the number of diamond lattice, the symmetries around the center of the structure become higher. This made it easier to obtain a broader response in many directions (common band gap in a wide range of angles), which is similar to the case of two-dimensional PCs (See Refs. [19, 20]).

2.3 Conclusion

In summary, the width of the band gaps in a PC made by the material with low refractive index is broadened greatly by multi diamond structures. A common band gap in a range of 150° of the incident angle is obtained. This technique will be greatly beneficial to achieve complete band gaps by the materials with low refractive index.

3. Band edge laser

Theoretical analysis has shown that a PC doped with gain materials can achieve lasing based on the characteristics of the high state density at the band edge of a PC [25-28]. Experimentally, the band edge laser in 1D [29], 2D (to dimension change to 2D) [30, 31] and 3D [32, 33] PCs have been reported. Holography has shown good potential in fabricating PCs [10, 11, 13, 34, 35]. Recent research shows that the 1D PC and quasicrystals PC, with gradual distribution of the refractive index materials made by holography using DCG, can also achieve band edge lasing [36, 37]. Using DCG, the present authors fabricated a 3D lattice structure which achieved wide band gaps [38], and sometimes achieved complete band gaps [39, 40]. Based on these studies, a band edge laser in an fcc PC was achieved in our laboratory. Although many studies on band edge lasers have been reported [25, 41-43], the different output properties of band edge lasers pumped by picosecond (ps) and nanosecond (ns) pulses have not yet been investigated. However, because the pump duration is related directly to the behavior of band edge laser emission, this kind of research is significant for understanding further the mechanism of the band edge laser. Recently, we found that the band edge laser showed obvious different emission properties depending on whether it was pumped by a ps laser or by a ns laser. Therefore, the investigation of such a problem becomes important. The following outlines our work.

3.1 Experimental method and the samples

The set-up geometry for fabricating the fcc structure is the same as that in Ref. [38]. In our experiments, the laser was a diode pumped laser working at 457.9 nm with line width of 200 kHz (Melles Griot model 85-BLT-605). The holographic recording material was DCG. It was coated on an optical glass plate with a flatness of $\lambda/10$ without any doping. DCG is a phase type holographic recording material with a refractive index of 1.52 and a modulation of refractive index less than 0.1. The thickness of the DCG is 36 μm . After exposure, the DCG plate was developed in running water at 20°C for two hours, and then soaked in a Rhodamine 6G solution bath with a concentration of 0.125 mg per milliliter water at the same temperature for 60 min, enabling the dye molecules to diffuse deeply into the emulsion of the gelatin. The DCG plate was dehydrated in turn by soaking it in 50 %, 75 % or 100 % isopropyl alcohol with the same concentration of Rhodamine 6G at 40°C for 15 min each. After dehydration, the DCG plate was baked at 100°C for 60 min in an oven.

3.2 Measured spectra and discussions

Figure 7(a) shows the measured transmission spectrum (the black line) of the fcc structure without dye doping and the absorption spectrum of Rhodamine 6G (the red line) respectively using a J-Y 1500 monochromator. It can be seen that the fcc structure has band gaps with a width (FWHM-full width at half maximum) of 80 nm. The center of the absorption spectrum of Rhodamine 6G is located just at the shorter wavelength edge of the band of the fcc structure. Fig. 7(b) shows the photoluminescence of Rhodamine 6G excited by a 532 nm laser beam. It can be seen that the whole photoluminescence also locates at the shorter wavelength edge of the band of the fcc structure.

Figure 8 shows the set-up geometry for measuring the emission of the band edge laser. In our experiments, the measured structure was pumped by a ns pulsed Nd:YAG laser with a pulse duration of 8 ns and a ps pulsed Nd:YAG laser with a pulse duration of 30 ps. The pulse laser with a repetition rate of 10 Hz is the most common commercial equipment in

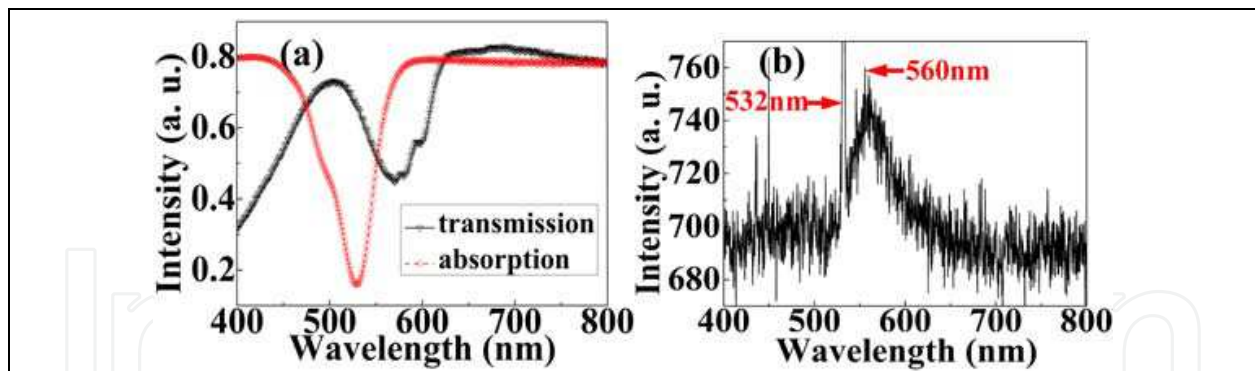


Fig. 7. (a) Transmission spectrum of the fcc structure and absorption spectrum of Rhodamine 6G. (b) Photoluminescence of Rhodamine 6G excited by a 532 nm laser beam.

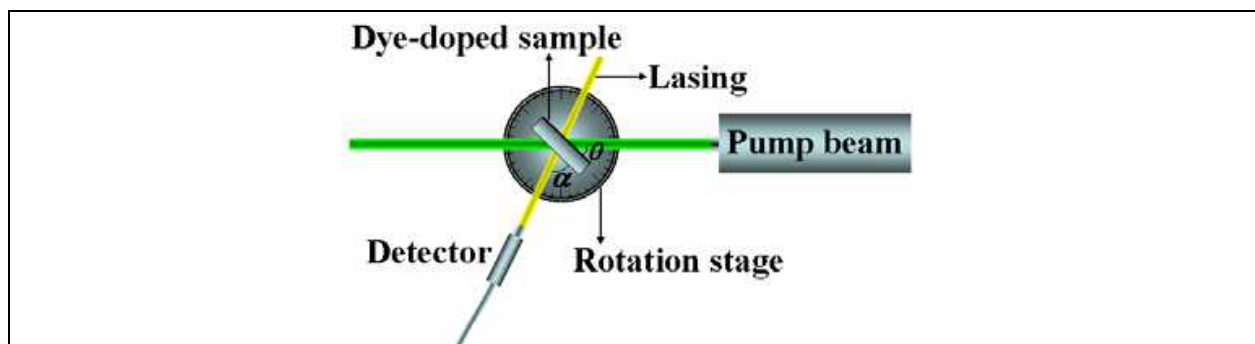


Fig. 8. Set-up geometry for measuring the emission of the band edge laser. θ is the angle of the pump beam with respect to the surface of the sample. α is the angle of the emission beam with respect to the surface of the sample.

wide use, for example, in the work of [36] and [37]. Therefore, such lasers running at 532 nm and repetition rates of 10 Hz were used. During measurements, the laser emission was collected by a lens, which was focused into a fiber and fed to a spectrometer. It should be clarified that the aim of the present work is to find the different emission characteristics of a band edge laser pumped by a ns laser and a ps laser. It is not to investigate the relationship between the band gap and the incident angle. In our experiments, lasing can be found in certain ranges of the incident and the output angles (corresponding to different directional band gaps). The following experimental results are the best for the corresponding conditions.

Figure 9 shows the emission spectra of the band edge laser pumped by a ps laser. For the case pumped by a ps laser, the size of the pump beam is 3 mm and the maximum pulse energy is 40 mJ. It can be seen that there are several modes; each of them with a width less than 0.4 nm. The threshold value is about 1.65 GW/mm². The lasing modes are not stable and no one is dominant among the several modes. The results shown in Fig. 9 are only two of the measured spectra at different pump power intensities. Furthermore, the measured spectra recorded at different times under the same pump power intensity are quite different, and the heights and the positions of the peaks in each spectrum are different (See Fig. 10).

Figure 11(a) shows the emission spectra of the band edge laser pumped by a ns laser. For the case pumped by the ns laser, the size of the pump beam is 13 mm and the maximum pulse energy is 1.5 J. It can be seen that the lasing threshold is around 6 MW/cm². This is only an estimated value since the lasing emission around the threshold is unstable. The threshold is

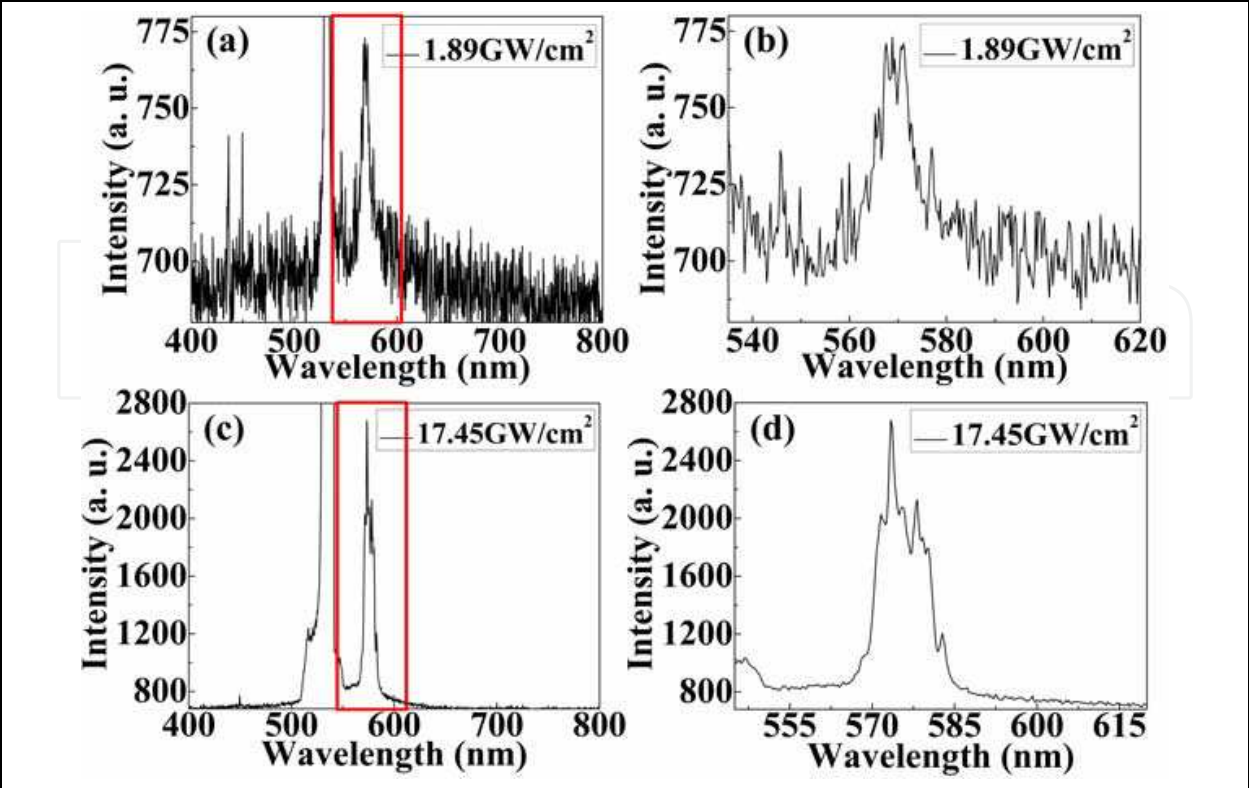


Fig. 9. Measured emission spectra of the band edge laser at different pump power intensity pumped by a ps laser. $\theta \approx 65^\circ$ and $\alpha \approx 50^\circ$. (a) and (c) are the spectra measured at different pump power intensities, (b) and (d) are the enlargement of the inset (red box) in (a) and (c).

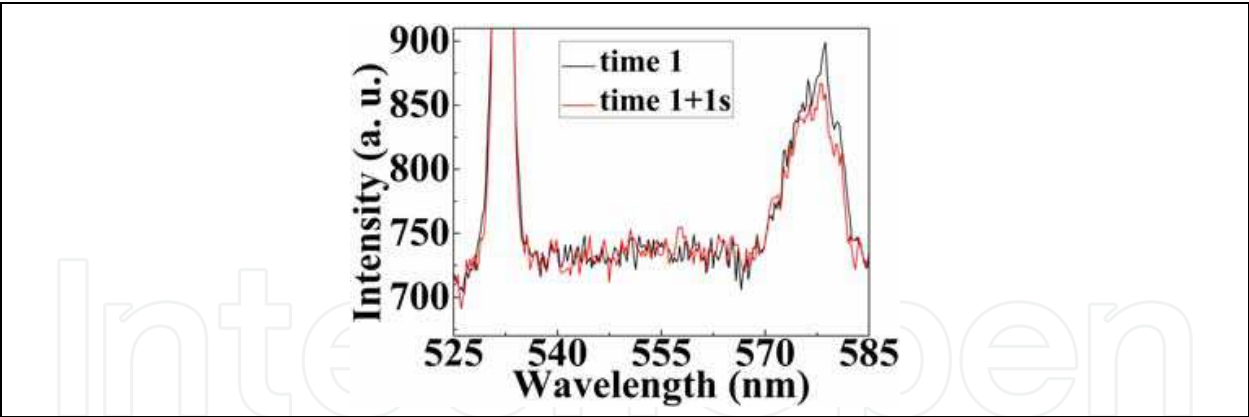


Fig. 10. The measured spectra recorded at different times under same pump power intensity.

much lower than that pumped by a ps laser. The right inset in Fig. 11(a) shows the intensity (green line) and the line width (blue line) vs. pump power intensity. From Fig. 11(b) it can be seen that when the pump power intensity is in a certain range (higher than the lasing threshold), there are two emission modes in competition. The two peaks in Fig. 11(b) are both located in the range of the gain profile. If there is no competition, the two peaks will grow with the increase of the pump power intensity. However, in our observations, only one peak grows when the pump power intensity increases, and the height of the other is always kept very low. Obviously, it is the phenomenon of competition. In addition, it is

found that one emission mode is higher when the pump power intensity is higher. When the pump power intensity is high enough there is only one mode in the emission that becomes dominant. This mode has a width of several nanometers, and the intensity of the emission is very high corresponding to the photoluminescence. Our experimental observations show that the dominant emission mode is quite stable, i.e., for every pump pulse, the frequency position and the output intensity always remain unchanged.

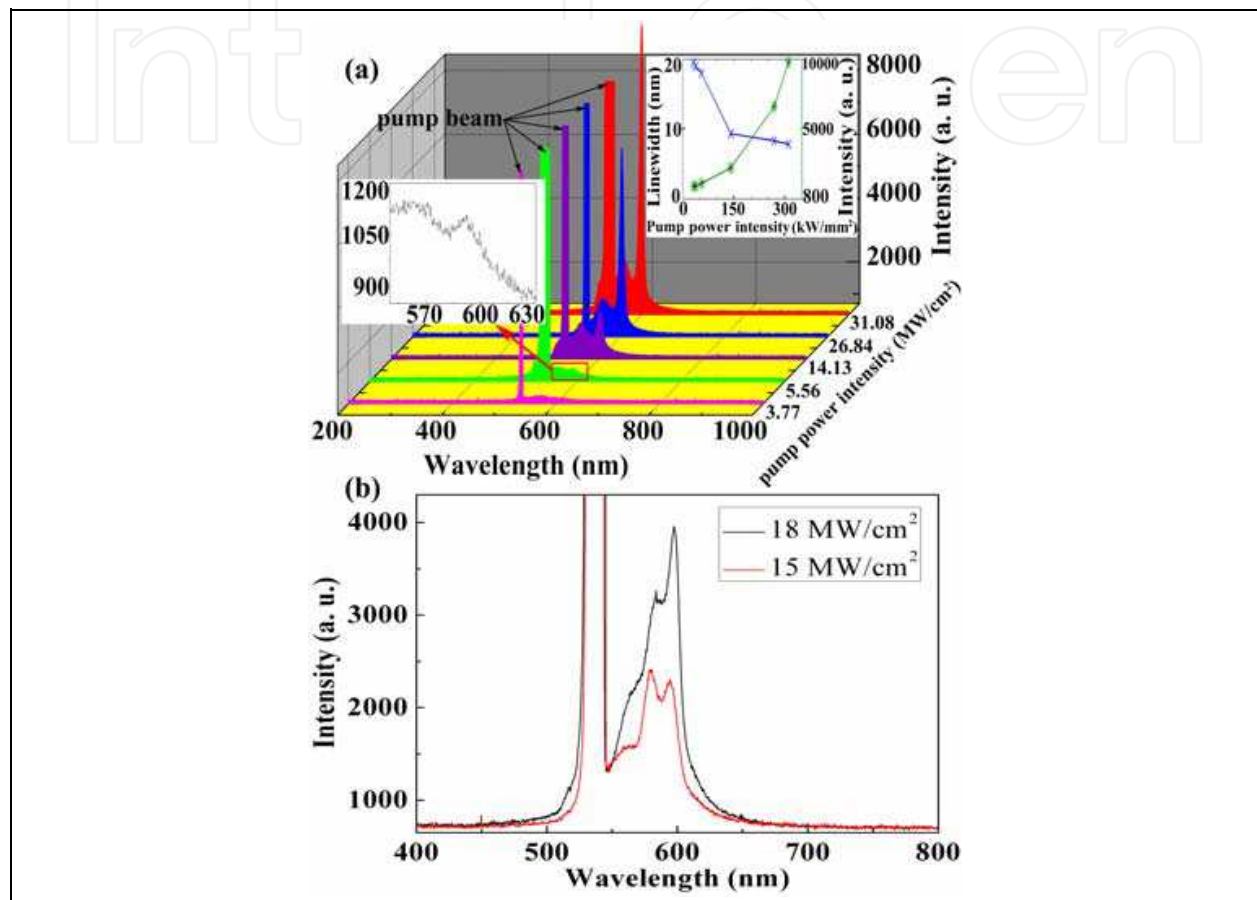


Fig. 11.(a) The emission spectra of dye-doped fcc PC as a function of pump power intensity pumped by a ns laser. In the right inset, the green line corresponds to the emission intensity of lasing, and the blue line corresponds to the line width of the lasing emission. It should be noted that the pump beam is saturated on the CCD used. (b) Measured spectra when the pump power intensity is in a certain range (higher than the lasing threshold).

There are many studies on the theoretical investigation of band edge lasers [25, 41-43]. These theoretical works are strictly quantitative analyses, and are suitable for most band edge lasers. Therefore, we will not repeat them here, but will provide a brief analysis on the physical mechanism to discuss the difference between the cases pumped by a ps laser and by a ns laser.

It is known that one of the important features of a PC is the high density of the mode at the band edge. If gain material exists in a PC, and the range of the gain is at the band edge, it will provide a lasing condition. The modes just inside the gain range may be lasing. As observed in Figs. 9 and 11, there are multi modes for the case pumped by a ps laser, but there is only one dominant mode for the case pumped by a ns laser. The reason is that in the

case pumped by a ns laser, the pulse width is about 10 ns, i.e., the interaction between the pump beam and the material will last for about 10 ns, which is a relatively long period, and the "mode competition" will have finished. As a result, one mode becomes dominant in the emission and is very stable. However, in the case pumped by a ps laser, the interaction between the pump beam and the material will only last for about 30 ps, and the "mode competition" cannot finish in the period of one pulse. Therefore, multi modes will appear in the emission. It is also found that the lasing threshold pumped by a ns laser is about three orders of magnitude less than that pumped by a ps laser. The reason is that lasing needs a build up time. For the case pumped by a ps laser if the pump power intensity is lower, and the pump pulse is over, the lasing has not built up. To build up the lasing needs higher pump power intensity. However, for the case pumped by a ns laser, the process of building up has a long enough duration. Thus, much lower pumped power intensity can build up the lasing. It should be pointed out that, the life time of the used energy level of Rhodamine 6G is around the magnitude of 10^{-7} s [44, 45], which is only one order of magnitude of the pulse width when a ns laser is used (10 ns), but is four orders of magnitude longer than the pulse width when a ps laser is used (30 ps).

3.3 Conclusion

In summary, when the band edge laser is pumped by a ps laser or by a ns laser it shows different emission characteristics. The number of emission mode, the line width, the lasing threshold and the stability of the lasing are obviously different.

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