

Fabrication of high-quality colloidal photonic crystals with sharp band edges for ultrafast all-optical switching*

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Application of the pressure controlled isothermal heating vertical deposition method to the fabrication of colloidal photonic crystals is systematically investigated in this paper. The fabricated samples are characterized by scanning electron microscope and transmission spectrum. High-quality samples with large transmissions in the pass bands and the sharp band edges are obtained and the optimum growth condition is determined. For the best sample, the transmission in the pass bands approaches 0.9 while that in the band gap reaches 0.1. More importantly, the maximum differential transmission as high as 0.1/nm is achieved. In addition, it is found that the number of stacking layers does not increase linearly with concentration of PS spheres in a solution, and a gradual saturation occurs when the concentration of PS spheres exceeds 1.5 wt.%. The uniformity of the fabricated samples is examined by transmission measurements on areas with different sizes. Finally, the tolerance of the fabricated samples to baking was studied.

Keywords: characterization, photonic crystal, dielectric materials, optical switch

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1. Introduction

During the last decade, photonic crystals (PCs) formed by periodic modulation of dielectric constant or refractive index have been explored for the construction of both passive and active functional devices.^[1-4] Among them, the all-optical switch is one of the key components for the ultrafast signal processing and telecommunication in the future. Thus, how to realize all-optical switches with ultrafast response time, low switching energy, and small size has always been a great challenge for physicists and engineers. It seems that PCs with Kerr nonlinearity may satisfy the requirements for speed, energy, and size and appear as a promising candidate to this goal. Accordingly, different mechanisms have been proposed to realize all-optical switches based on nonlinear PCs.^[5-8] Actually, the first idea utilizing the band edges of nonlinear PCs was proposed a decade ago.^[5] Basically, the signal light that is intentionally located near the band edges can be switched between an on state and an off state by applying a control light that shifts the

band edges through nonlinearity-induced refractive index change.^[5] Since the nonlinear response time based on the Kerr effect is extremely fast, the main issue to be solved is how to reduce the switching energy to a low level so that the integration of a large number of switches on a single chip will become feasible. Apparently, the sharpness of the band edge as well as the transmission change across the band edge affects significantly the performance (e.g., the switching power) of the constructed all-optical switches. In principle, the sharpness of the band edges is closely related to the density of imperfections existing in PCs. On the other hand, it is well known that significant enhancement of nonlinearity is anticipated in PCs by utilizing the slow light effect.^[9,10] In addition, it has been shown that the nonlinearity enhancement is inversely proportional to the square of group velocity.^[11] Thus, the switching energy can be dramatically reduced if the nonlinearity of PCs can be significantly enhanced for the control light. However, both theoretical analysis and experimental observation indicate that the reduction of group velocity in PCs is severely restricted

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by the imperfections present in PCs.^[12–14] Therefore, how to improve the quality of PCs has become a crucial issue that determines the realization of all-optical switches with low power consumption. In experiment, much effort has been devoted to the fabrication and the characterization of nonlinear PCs that can be employed to realize all-optical switches.^[15–19]

It has been demonstrated that colloidal PCs synthesized from solutions containing polystyrene (PS) spheres suspended in a fluid (e.g., water or ethanol) are quite suitable for this purpose.^[18] Ultrafast all-optical switching was realized by utilizing the shift of band edges. However, the switching operation was observed only by pumping the PC with an amplifier because the switching energy, which was restricted by the sharpness of the band edges, was quite large.^[18] As mentioned above, the sharpness of band edges is governed by the density of imperfections that are inevitably introduced in the fabrication process. Therefore, how to reduce the density of imperfections has become a key issue to be solved. Until very recently, however, high-quality colloidal PCs that are suitable for making all-optical switches were successfully fabricated by utilizing a technique named pressure controlled isothermal heating vertical deposition (PCIHVD).^[20] The achievement of high-quality colloidal PCs was demonstrated through an accurate control of the evaporation rate of water vapour. The good quality of the fabricated PCs was manifested in the sharp band edges with a slope as large as 0.07/nm, the high transmission in the pass bands (> 0.8), and the low transmission in the band gaps (< 0.02). In this article, we present a systematically investigation on the fabrication and characterization of colloidal PCs by using this technique, with emphasis on how to optimize the vapour pressure and the concentration of PS spheres. The fabricated samples are characterized by scanning electron microscope (SEM) and transmission spectrum. The effects of measurement setup and baking on the transmission properties of the fabricated samples are also discussed.

2. Optimization of the growth condition

A detailed description of the PCIHVD method was presented by Zheng *et al.*^[20] and we will not repeat it here. Based on the experimental observations, the authors found that for their apparatus the op-

timum growth temperature and vapour pressure were 35°C and 39 mmHg (1mmHg = 1.3332×10^2 Pa), corresponding to an evaporation rate of about 0.71 g/cm²h. Physically, the evaporation rate can be calculated from Stefan's law

$$\dot{m}_l = (DP_t/lR_lT) \ln[(P_t - P_{ls})/(P_t - P_{lw})], \quad (1)$$

where D is the diffusion coefficient of water vapour in air, l is the distance from the liquid surface to the open end of the vial, R_l is the gas constant of water vapour, T is the temperature at the liquid surface, and P_t , P_{ls} , and P_{lw} are the total pressure in the growth chamber, the pressure at the open end of the vial, and the pressure at the liquid surface, respectively. In particular, it is noticed that the evaporation rate depends not only on the vapour pressure and growth temperature but also on the configuration of the vial.

We establish a fabrication system which is similar to that in Ref.[20]. A schematic drawing for our fabrication system is shown in Fig.1. In order to achieve a better control of the vapour pressure and to protect the pump, we have added a bottle containing silica gel between the vial and the pump. To do so, the water vapour driven out of the vial will be absorbed by silica gel and it does not enter into the pump or return to the vial. Hence, the vapour pressure is maintained to be stable during the fabrication process. The PS spheres used in our study are purchased from Duke Corporation. The diameter of the PS spheres is 260 nm with a dispersion of less than 3%. In order to find out the optimum growth condition for our apparatus, we fix the growth temperature at 35°C and adjust the vapour pressure to around 39 mm Hg. The samples fabricated under different vapour pressures are characterized by both SEM and transmission spectrum. According to

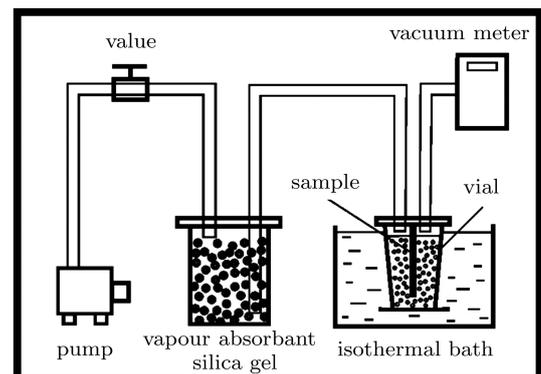


Fig.1. Experimental setup for the fabrication of colloidal PCs by using the PCIHVD method.

the SEM images, we can roughly judge the quality of the samples by inspecting the ordering of PS spheres and the densities of defects and dislocations. On the other hand, the sample quality should be clearly manifested in the transmission spectra. Thus, we can determine the sample quality by examining the transmission in the pass bands and the sharpness of the band edges.

In experiments, we first try to synthesize colloidal

PCs from an aqueous solution in which the concentration of PS spheres is $c = 0.5$ wt.%. The vapour pressure P_t varies from 30 to 50 mmHg. Under this growth condition, the fabrication process generally lasts 6–8 h. The fabricated samples are characterized by SEM and transmission spectrum. The SEM images for the samples grown at 30, 39, 45, and 50 mm Hg are shown in Figs.2(a), 2(b), 2(c), and 2(d) respectively while the corresponding transmission spectra are presented in Fig.3.

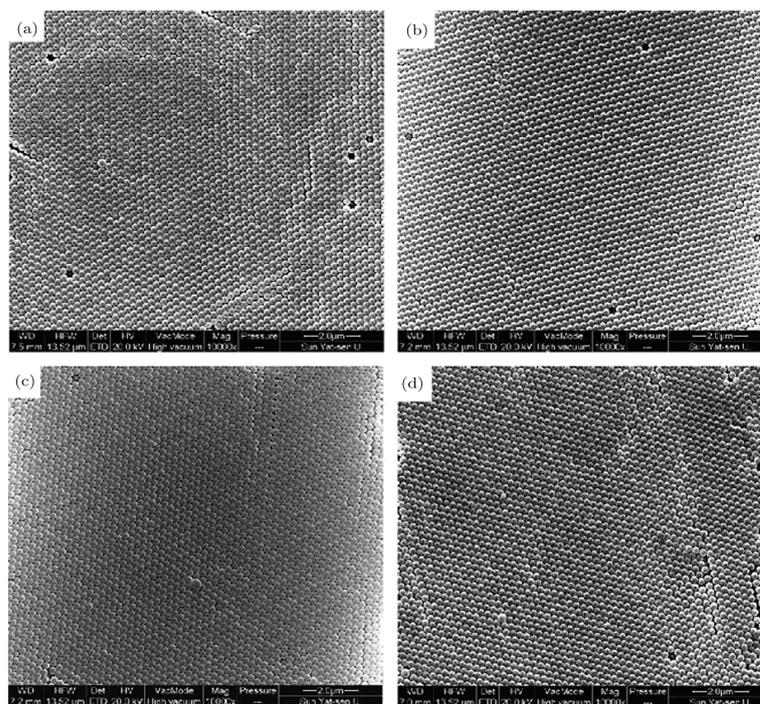


Fig.2. SEM images (top view) for the colloidal PCs synthesized under different values of vapour pressure (P_t): 30 (a), 39 (b), 45 (c) and 50 mm Hg (d), with the diameter and the concentration of PS spheres being 260 nm and 0.5 wt.% respectively.

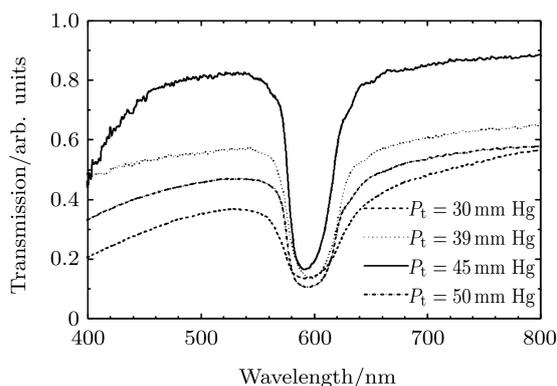


Fig.3. Transmission spectra for the colloidal PCs synthesized under different vapour pressures, with the diameter and the concentration of PS spheres being 260 nm and 0.5 wt.% respectively.

From the SEM images, we can easily find that the morphologies for the samples obtained at $P_t = 39$ and 45 mmHg are better than those for the samples synthesized at $P_t = 30$ and 50 mmHg. This is reflected in the fact that the densities of defects and dislocations found in the former are less than those in the later. However, a more accurate judgment of sample quality can be given by transmission spectrum measurements which are sensitive to the imperfections existing inside the sample.

The transmission spectra measured for the four samples described above are shown in Fig.3. In all samples, a photonic band gap centred at about 590 nm is clearly seen. The transmission in the band gap, which is generally determined by the total number

of stacking layers, is found to be about 0.15. It is apparent that the best sample is achieved at $P_t = 45$ mmHg. The good quality of this sample is manifested in two aspects. One is the large transmission in the pass bands which is above 0.8 in most of the frequency range. This value is much higher than those observed in other samples, indicating that the density of imperfections is greatly reduced. As a result, a transmission change as large as 65% can be achieved across the band edges. The other is the sharpness of the band edges that are closely related to the tail states originating from the structural imperfections. In Fig.3, sharper band edges and narrower band width are clearly observed in this sample. The large transmission change in combination with the sharp band edges is crucial for the realization of all-optical switches with good performance and low power consumption.

3. Dependence of the thickness on the concentration of PS spheres

As compared with the other fabrication methods, the PCIHVD technique has a major advantage in achieving well-ordered structures with very few imperfections. As mentioned above, this feature is clearly reflected in the sharp band edges as well as the large transmissions in the pass bands. In practical applications, it is generally required to fabricate colloidal PCs with a sufficiently large number of stacking layers in order to realize some functionalities. Therefore, the issue how to obtain high-quality colloidal PCs with many stacking layers arises and needs studying.

Although a large transmission change across the band edges has been achieved in the sample fabricated at $P_t = 45$ mmHg, this value does not fulfil the requirement for practical switching devices. Intuitively, a further improvement in the transmission change can be achieved by either reducing the density of imperfections or increasing the number of stacking layers. The former may result in an enhancement of the transmission in the pass bands while the latter may cause a further deepening of the band gap. Therefore, we try to vary the concentration of PS spheres in the solution used for the synthesis of colloidal PCs and study its influence on the sample thickness and quality.

Still, we use the procedure described above to

find out the optimum vapour pressure for the solution with PS spheres of 1.0 wt.%. In Fig.4, the transmission spectra measured for four samples fabricated under different vapour pressures are shown. Compared with the samples shown in Fig.3, the samples in Fig.4 are improved in two aspects that are clearly observed. One is the sample quality, which exhibits a much weaker dependence on the vapour pressure. It implies that the strict requirement for obtaining good samples is relaxed to some extent. Since the control of vapour pressure is crucial to the fabrication process, this feature makes it easier to obtain samples with good quality. The other is steeper band edges and lower transmission in the band gaps, which are exhibited in the best sample obtained at $P_t = 45$ mmHg. The steepness of the band edges will be analysed later.

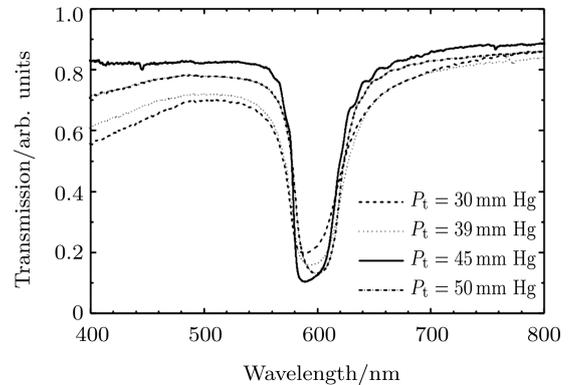


Fig.4. Transmission spectra for the colloidal PCs synthesized under different vapour pressures, with the diameter and the concentration of PS spheres being 260 nm and 1.0 wt.% respectively.

The numbers of stacking layers in the samples fabricated from solutions with different concentrations of PS spheres are examined by inspecting the sample cross sections obtained from the SEM measurements. The results are shown in Fig.5. It can be seen that the PS spheres are well stacked layer by layer with a few stacking faults. The number of stacking layers is counted and its relationship with the concentration of PS spheres in the solutions is shown in Fig.6. When the concentration of PS spheres increases from 0.5 to 1.0 wt.%, we observe a linear increase of the stacking layer from 14 to 42. After that, the increase in number of stacking layers becomes slower. Finally, it seems that the number of stacking layers does not change too much with the increase of concentration of PS spheres. The maximum number of stacking layers is around 55. This behaviour is different from those ob-

served by using other fabrication methods.^[21–23] Even for the PCIHVD method, our observation is slightly different from that found in other reference,^[20] where

a linear increase of the stacking layer was found for concentrations up to 2.0 wt.%.

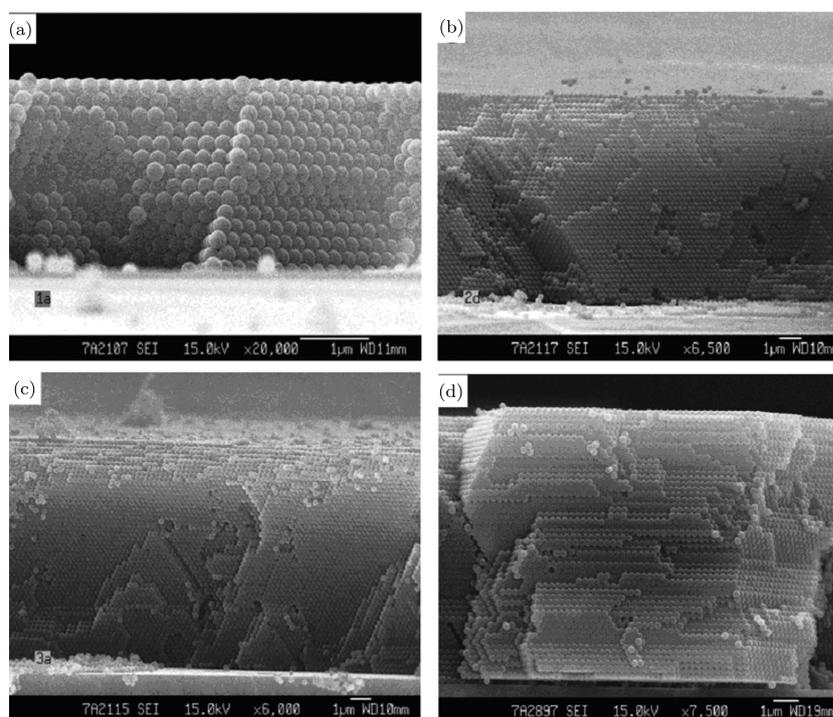


Fig.5. SEM images (cross section) for the colloidal PCs synthesized under optimum condition from solutions, with concentrations of PS spheres being 0.5 (a), 1.0 (b) 1.5 (c), and 2.0 wt.% (d).

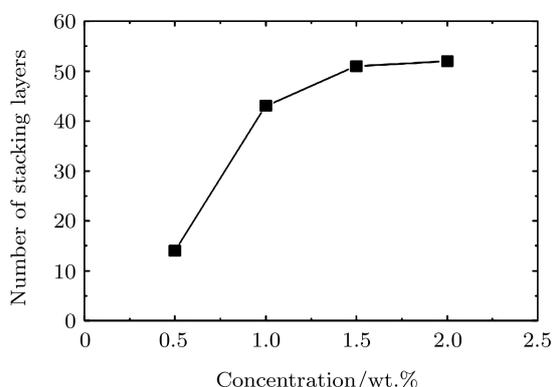


Fig.6. Dependence of the total number of stacking layers in the fabricated colloidal PCs on the concentration of PS spheres.

4. Sharp band edges of the fabricated samples

As mentioned at the beginning, the switching energy of the PC-based switches is mainly determined by sharpness of the band edges. For practical applications, it is highly desirable to obtain colloidal PCs with

extremely sharp band edges that can be employed to realize switching operations with low power consumption. Therefore, it is necessary to characterize the sharpness of the band edges of the fabricated samples. In Fig.7, we compare the transmission spectra of the best samples obtained from solutions with different concentrations of PS spheres. Although the number of stacking layers increases with the increase of the concentration of PS spheres, we do not see the further narrowing of the bandgap width and the further deepening of the band gap. This observation accords with that found in other reference^[24] where the decrease in bandgap width with the increase of stacking layers was found to be saturated when the number of stacking layers exceeded 10. By examining the transmission in the pass bands as well as in the band gap, it is found that the best sample is achieved at a concentration of 1.0 wt.%. For this sample, the transmission in the pass bands approaches 0.9 while that in the band gap reaches about 0.1. For the sample synthesized at a concentration of 0.5 wt.%, it has nearly the

same transmission in the pass bands. However, its transmission in the band gap is not sufficiently low. For the remaining two samples, their transmissions in the band gap are similar to that of the best sample. Unfortunately, their transmissions in the pass bands are lower than that in the best sample.

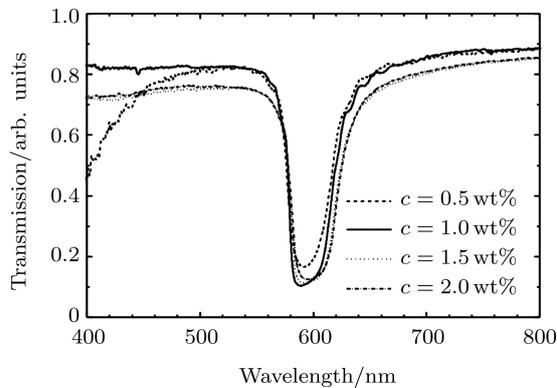


Fig.7. Transmission spectra for the colloidal PCs synthesized under optimum condition from solutions with different concentrations of PS spheres.

We have also examined the band edges of the four samples and the results are presented in Fig.8 where the differential transmissions (i.e. $dT/d\lambda$) for all the samples have been calculated. For clarity, their differential transmission spectra are shifted vertically. It is found that the edge of the air band, which is located

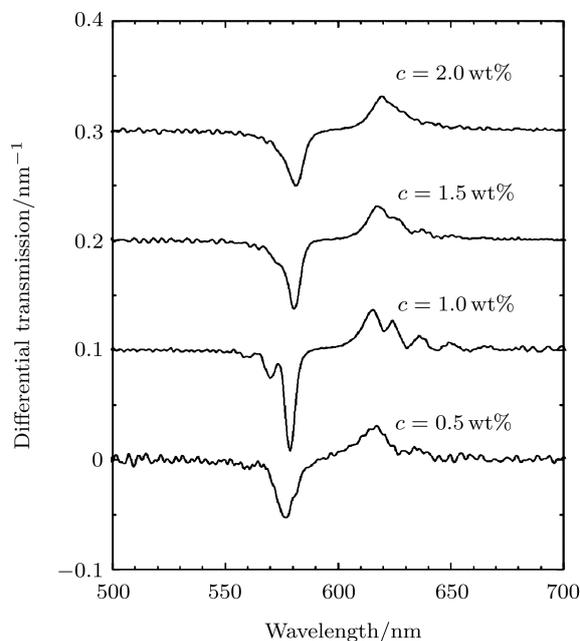


Fig.8. Differential transmission spectra showing the sharpness of the band edges of the colloidal PCs fabricated from solutions with different concentrations of PS spheres.

on the short-wavelength side, is always sharper than that of the dielectric band. We believe that the dispersion of the PS spheres, which results in tail states near the edge of the dielectric band, is responsible for this behaviour. It is remarkable that for all the samples the maximum differential transmissions exceed $0.05/\text{nm}$. Particularly, it is noticed that a very large differential transmission has been achieved to be nearly $0.1/\text{nm}$ in the best sample. This value is greater than that reported in Ref.[20], indicating that our fabrication system is improved and the growth condition is optimized.

5. Uniformity of the fabricated samples

The uniformity of the fabricated samples is crucial to fabricating devices, and it is examined by measuring the transmission spectra at the same position but with different sizes. Two experimental configurations are employed in the measurements and they are shown in Figs.9(a) and 9(b). In the first scheme, an objective lens and a diaphragm are located between the light source and the monochromator. The monotonic light beam emerging from the slit of the monochromator has a strip shape. The width of the strip is 1 mm while its height H is varied by adjusting the width of the diaphragm. The transmission spectra obtained in areas with different sizes are compared in Fig.9(c). It is observed that the transmission in the pass bands exhibits its dependence on the size of the measurement area while that in the band gap does not. With measurement area decreasing, the transmission in the pass band is found to first increase and then decrease. Since this transmission is closely related to the density of defects in the measurement area, the influence of the defects on the transmission is larger when the measurement area is too large or too small. For the best situation, we can observe the Fabry-Perot fringes but they are not obvious. In the second measurement scheme, two objective lenses are used in front of and in rear of the monochromator separately. The measurement size is changed by moving the sample around the focus point of the second objective lens. The beam diameter Φ varies in a range of 1–2.5 nm, i.e. $\Phi = 1\text{--}2.5 \text{ nm}$. The transmission spectra measured in different areas are presented in Fig.9(d) for comparison. In this case, the two features described

above are clearly seen. In the case where the density of defects is small, the transmission in the pass bands is found to be slightly higher. Moreover, we can clearly observe Fabry-Perot fringes in the pass bands.

These behaviours are probably due to the relatively small measurement areas in this measurement configuration.

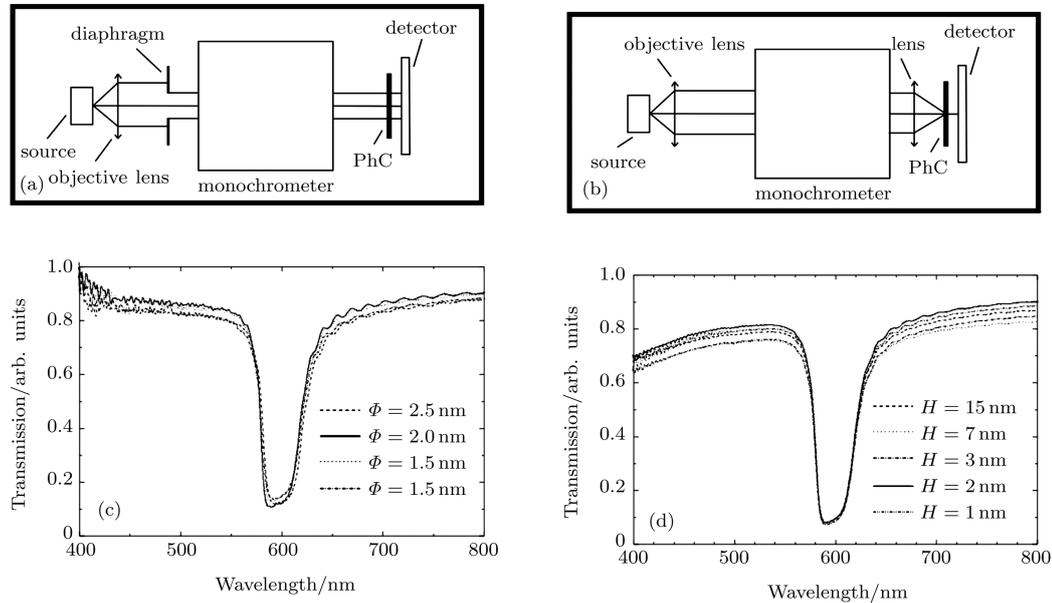


Fig.9. Experimental configurations for measuring the transmission spectra by using a diaphragm (a) and a focused lens (b), and their corresponding transmission spectra obtained in areas with different sizes (c) and (d).

6. Effect of baking on the sample quality

In order to fabricate devices using colloidal PCs, it is necessary to increase the stability as well as the mechanic strength of the fabricated samples. Therefore, a baking process is usually employed at the end of the fabrication. Thus, we need to find out the tolerance of the samples to the baking temperature and time because a high-temperature baking may damage or even destroy the sample while a low-temperature

one will have little effect on the sample strength. We have tried to bake a fabricated sample at 110°C for different times and their corresponding transmission spectra after baking are shown in Fig.10(a). It can be seen that no obvious change is found in the transmission spectrum after a one-minute baking. However, the band gap in the transmission spectrum almost disappears when we increase the baking time to 5 min, implying the collapse of the PC structure. This experiment indicates that the baking temperature of 110°C is too high. At this temperature, the sample quality degrades quickly and care must be taken when the

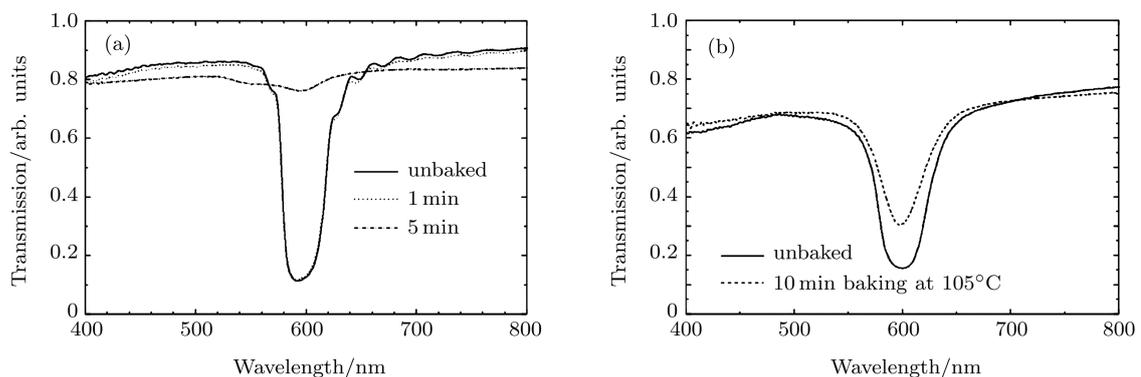


Fig.10. Transmission spectra for a fabricated sample baked at 110°C for different times (a), and the change in transmission spectrum for a fabricated sample baked at 105°C for 10 min.

baking time exceeds 1 min. Then, we have tried a lower temperature of 105°C for other sample, and the obtained result is shown in Fig.10(b). In this case, with the baking time being 10 min for the sample, it is found that the transmission in the band gap increases from 0.15 to 0.30 while that in the pass bands does not change. It means that there appears a degradation in sample quality but it is not significant. Therefore, we conclude that the suitable temperature for baking is around 105°C.

7. Conclusions

We have systematically investigated the fabrication of colloidal PCs by using the PCIHVD method,

aiming at the achievement of colloidal PCs with sharp band edges that can be employed to construct all-optical switches with lower power consumption. The fabrication system is improved and the growth condition is optimized. The fabricated samples are characterized by SEM and transmission spectrum. High-quality samples with large transmissions in the pass bands and the sharp band edges are obtained. More importantly, a differential transmission as large as 0.1/nm is demonstrated. The dependence of stacking layer on concentration of PS spheres is derived, and the uniformity of the fabricated samples is confirmed. Finally, we find a suitable temperature for baking the samples. We believe that our experimental results are helpful for the development of switching devices based on colloidal PCs.

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