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### Cover Page Footnote

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## Simulation and Experimental Study of a 2D Photonic Crystal Structure that Reflects a Quantum Dots Emission in the Normal Direction

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### Abstract

Two-dimensional photonic crystal structures not only confine light and guide waves laterally but also reflect light in the normal direction due to a slow Bloch mode effect. However, evidence of the utilization of this structure as a mirror is required. Therefore, in this work, a simulation was made and experimental results were obtained to prove that there was an increase in the intensity of reflected CdSe colloidal quantum dots emission in the normal direction when a 2D photonic crystal structure was used. A thin TiO<sub>2</sub> film was shaped into a two-dimensional photonic crystal structure using a simple sol-gel and polystyrene-mask-etching procedure. This structure was then placed on top of the thin CdSe quantum dots film layer. The emission of quantum dots onto the two-dimensional photonic crystal structure was compared to quantum dots emission onto a flat, thin TiO<sub>2</sub> film. An increase in the quantum dots emission of up to 105% was in the presence of the two-dimensional photonic crystal structure. This finding is very useful for photonic device applications, such as light-emitting diodes, laser systems and bio-tagging detection systems.

### Abstrak

**Studi Simulasi dan Eksperimen Struktur Kristal Fotonik Dua Dimensi sebagai Pemantul Emisi Kuantum Dot Masalah Arah Normal.** Kristal fotonika dua dimensi tidak hanya dapat mengukung cahaya dan memandu gelombang ke arah sisi horizontal, tapi juga mampu memantulkan cahaya ke arah normal dikarenakan efek moda slow Bloch. Namun bukti tentang penggunaan struktur ini masih dibutuhkan. Untuk itu dalam penelitian ini hasil simulasi dan percobaan telah didapatkan untuk membuktikan bahwa terjadi peningkatan pantulan emisi CdSe kuantum dot colloid pada arah normal saat kristal fotonika dua dimensi digunakan. Sebuah lapisan tipis TiO<sub>2</sub> dibentuk menjadi kristal fotonika dua dimensi dengan menggunakan teknik sol-gel yang sederhana dan etching dengan menggunakan polystyrene. Struktur ini diletakkan di atas lapisan kuantum dot CdSe. Emisi dari kuantum dot di atas kristal fotonika dua dimensi dibandingkan dengan emisi kuantum dot di atas lapisan tipis dan datar TiO<sub>2</sub>. Peningkatan emisi kuantum dot hingga mencapai 105% berkat kehadiran kristal fotonika dua dimensi. Hasil ini sangat berguna untuk aplikasi divais fotonika seperti LED, sistem laser dan sisten deteksi bio-tagging.

*Keywords: photoluminescence, photonic crystal, quantum dots, slow Bloch mode*

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### Introduction

Photonic crystal (PC) structures are promising submicron-scale photonic devices due to their unique optical property, the so-called photonic band gap. It is well known that PC can be a good reflector of light at wavelengths corresponding to the photonic band gap (PBG) region [1]. In the case of a three-dimensional PC structure, the reflection mechanism works in three directions. For two-dimensional (2D) PC structures, this reflection

obviously occurs in the lateral direction only, in the same direction as in an array structure. However, for 2D PC, there is another mechanism of reflection in the non-lateral (normal) direction. This mechanism comes into play inside the PC structure with the so-called slow Bloch modes. The slow Bloch modes mostly occur at the  $\Gamma$ -point, which is the normal direction of the PC structure [2,3]. We noticed that the implementation of a slow Bloch mode in optical devices is very rare, due perhaps to limited theoretical and experimental evidence.

Further simulation and experimental evidence for this mechanism is required. Moreover, a 2D PC structure is designed to be applied in micron-size photonic devices, where a nano-emitter such as a colloidal quantum dots (QDs) is used.

Meanwhile, colloidal QDs have been conjugated with several optical structures to create advanced optical devices such as bio-sensors, bio-imagers, light-emitting diodes, solar cells, photovoltaic devices, etc. The conjugation of QDs with PC structures has led to more advanced applications, such as microcavity nano-lasers, single quantum sources for quantum computing, light-enhancing devices, etc. Research on the conjugation of QDs emission with opal PC structures was published several years ago; e.g., one research group claimed the first observation of QDs emission enhancement using a PC structure, which was due to light-matter interaction at the edge of the PBG [4]. Furthermore, it has been reported that the PBG of the PC influences the QDs emission, resulting in emission enhancement, peak shifting, peak splitting, peak narrowing and a longer lifetime of QDs emission [4-7]. The effect of PC on QDs emission has become an interesting research subject and some research has been conducted to reveal the reasons behind the modification of the QDs emission. Recently, some researchers have shown that the stop band in the photonic band gap can modify the QDs emission in a certain direction [4,5]. The increase in QDs emission is mainly due to the reflection properties of the PC structure.

A light reflector is very useful in a photonic device system. It is well-known that light reflectors made of a thin metallic layer or of multiple layers of dielectric materials reflect incident light. However, this kind of device only has one optical function, i.e., light reflection in the normal direction. In a nano-photonic device it is necessary to have a multi-functional device. A 2D PC is a promising multi-functional photonic structure, as it can guide waves and confine light laterally and reflect light in the normal direction. Therefore, in this work, we theoretically and experimentally prove that a 2D PC structure can reflect incoming light in the normal direction. Here, cadmium selenide (CdSe) colloidal QDs were used as the light source and its emission increased in the normal direction. The simulation was conducted first to confirm the reflectance spectra and to find the best parameters for the PC structure. A PC structure was then fabricated and tested. The novelty of this work is that the simulated 2D PC structure is especially designed as a reflector of a visible wavelength and the fabrication process used here is simple and fast for large scale fabrication.

As mentioned earlier, a 2D PC structure is usually used to reflect, confine and guide light in both lateral (x and y axes) directions. In this work, we used a 2D PC

structure as a reflector in the normal (z axis) direction with a slow Bloch mode. A brief explanation is given below. The slow Bloch modes mostly occur at the  $\Gamma$ -point, which is the normal direction [2,3]. At these points, the curvature of the photonic band diagram is zero or close to zero. We see several benefits in these points. First, these highly symmetrical points generate a slow group velocity. Another benefit is that these points couple the normal direction of light to the lateral direction of the light [8,9]. This is the key point of a slow Bloch mode. The coupled light temporarily moves in the lateral direction before it escapes in the normal direction. These slow Bloch modes temporarily trap normal incident light and some light with a small angle of incidence. The ability of slow Bloch mode to couple a small angle of incident light is due to the degree of the curvature of the photonic band. Once the coupled light moves laterally, it starts to escape in the normal direction. In this case, the coupling time and escape time are important parameters.

A basic depiction of the reflectance process is given in Figure 1. When the normal incident light enters the 2D PC, there are two paths of light. First, the light passes through the PC structure similarly to the way light penetrates a transparent, thin film layer. Second, the light is coupled and temporally confined in the PC structure before escaping from it (noted as  $\Delta\phi$  in Figure 1). Thus, the reflectance of the light depends on the phase difference between the first path ( $\phi_1$ ) and the second path of the light ( $\phi_1 + \Delta\phi$ ). When the two paths destructively interfere with each other, the light is normally reflected. On the other hand, when the two paths constructively interfere with each other, the light is transmitted out of the other side of the 2D PC structure.

## Methods

This research consists of two parts, i.e., simulation work to estimate the structural parameters of the PC structure before its fabrication and experimental work to fabricate and to measure the influence of the 2D PC structure on QDs emission. Based on the simulation result, a 2D PC with structural parameters similar to those of the simulation results was fabricated. The simulation work was done using the FDTD (finite difference time domain) method, graphical user interface software and a hexagonal array structure to ease the simulation and fabrication processes. The schematic input of the simulation is shown in Figure 2a. An array of pillars forming a 2D hexagon (shown in Figure 2b) was placed in the middle of the simulation region. This 2D array represented a slab (hanging) PC structure. A plane wave from an incident electromagnetic source was directed upward from underneath the array. This wave was an emission of QDs. The incident light source contained

wavelengths of 400 to 700 nm. In order to simulate the reflected spectrum, a reflectance mode monitor plane was placed beneath the incident source. This reflectance spectrum was then confirmed with a transmittance mode monitor plane placed above the PC structure.

In this simulation, the distances between the incident source and the PC structure, between the reflectance monitor and the 2D PC structure and between the transmittance monitor and 2D PC structure are ignored. The 2D PC structure in this simulation is a 2D slab structure. Experimentally, this slab (hanging) structure is impossible, since the PC must be grown or deposited on a substrate. However, since the height of the PC pillars is very small compared to that of the experimental glass substrate, the slab structure was chosen to simplify the simulation. Several simulated parameters of the pillars, such as pillar diameter, pillar height and the refractive index of the pillar material, were obtained. The distance between two adjacent pillar centers was 300 nm.

The 2D PC structure was fabricated using a simple sub-micron sphere-masking process. The complete schematic procedure for fabricating the 2D PC structure is shown in Figure 3a. First, a thin titanium dioxide ( $\text{TiO}_2$ ) layer was deposited on the glass substrate using the sol-gel method. The fabrication of a thin  $\text{TiO}_2$  layer was

described in previous work [10]. Second, one layer of a closely packed structure of 300 nm diameter polystyrene spheres was deposited on the  $\text{TiO}_2$  layer using a convective method [11]. In order to reduce the pillar diameter of the PC structure, the polystyrene spheres were etched slightly, using reactive ion etching (RIE) with  $\text{O}_2$  gas. The etched polystyrene spheres were used again as etching masks to etch the  $\text{TiO}_2$  layer in order to obtain the pillar structure of  $\text{TiO}_2$  as the 2D PC structure.  $\text{O}_2$  and  $\text{Cl}_2$  gases were used in this second application of the RIE process. Finally, the complete  $\text{TiO}_2$  2D PC structure was obtained by removing the remaining polystyrene spheres using ultrasonication. A scanning electron microscope was used after each step of the fabrication to monitor the process. In addition, a flat  $\text{TiO}_2$  layer with the same thickness as the 2D PC structure was fabricated on another glass substrate as a reference or control layer for QDs emission.

A source emitted CdSe colloidal QDs with an emission peak wavelength at 605 nm to test the reflectance effect of the 2D PC structure. The QDs were mixed with ultraviolet UV-cured resin to make a layer of QDs on a glass substrate approximately 500  $\mu\text{m}$  thick. Emission of QDs was detected using a photoluminescence setup, as shown in Figure 3b. Photoluminescence measurement was carried out using an inverted microscope and a 50X

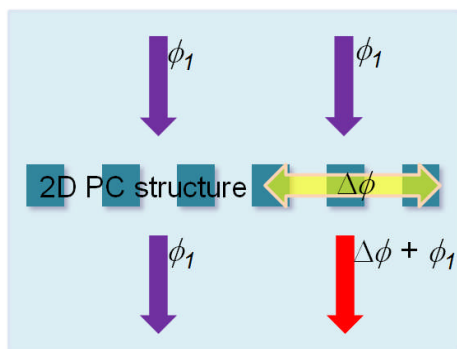


Figure 1. Schematic Process of Reflection by a 2D PC Structure using a Slow Bloch Mode

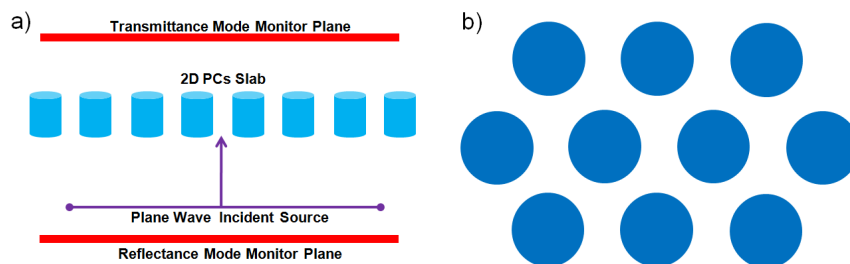
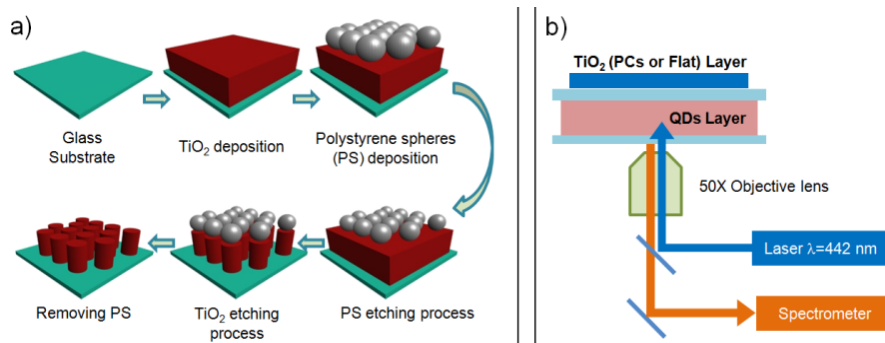


Figure 2. Schematic Input of the Simulation to obtain the Structural Parameters of the 2D PC Structure (a) Top View of the Hexagonal Pillar Array of the 2D PC (b)



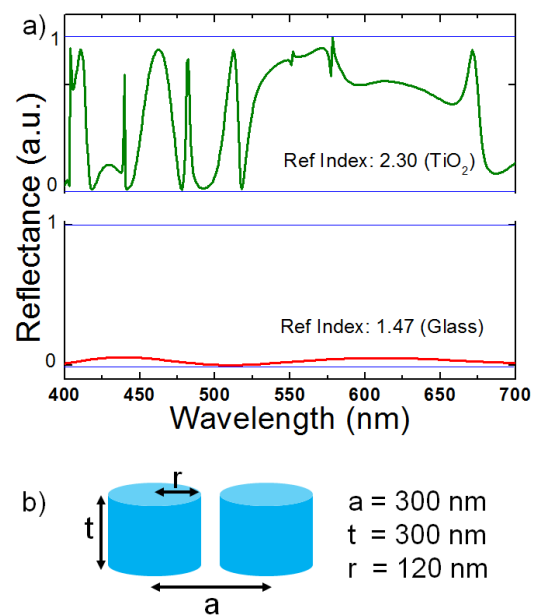
**Figure 3. Schema of the 2D PC Structure Fabrication (a) and the Photoluminescence set-up to Measure QDs Emission (b)**

objective lens. The QDs layer was placed on the sample stage of the microscope and the fabricated 2D PC structure was placed on top of the QDs layer. A He-Cd laser produced a continuous wave with wavelengths of 442 nm at power of 10 mW that was used to excite the QDs layer. The direct QDs emission and the reflected QDs emission were detected by a USB type spectrometer MAYA<sup>®</sup> from Ocean Optics. The laser was defocused onto the QDs samples to avoid bleaching the QDs.

## Results and Discussion

A slow Bloch mode effect was achieved by simulating a pillar type 2D PC structure. First, the simulation was done to determine proper material parameters for a 2D PC structure. Two transparent materials for the visible spectrum region were selected, i.e., glass (refractive index 1.46) and the TiO<sub>2</sub> layer (refractive index 2.3–2.8). These two materials are transparent materials that are easy to obtain and shape into a PC structure. The simulation results from normal direction reflectance of a 2D PC using these two materials are shown in Fig 4a. The simulated 2D PC structure was a pillar type structure with a radius of 120 nm, a height of 300 nm and a distance between pillars of 300 nm (as shown in Figure 4b.). The simulation revealed that a material with a high refractive index gives a higher reflectance than does a material with a low refractive index. The glass PC structure did not produce a large enough reflectance effect in the visible spectrum region. The maximum reflectance was only 0.75, so not good enough for practical application.

On the other hand, the TiO<sub>2</sub> PC structure yielded a very high reflectance in the normal direction. The maximum reflectance is broad: wavelengths from 515 to 660 nm. This result is promising for applied photonic devices. A material with a high refractive index is better for a PC structure. Basically, in a PC structure, light propagation is restricted by the array of pillars. The light actually has to travel between two materials alternately, i.e., air and solid pillar material. The difference between the refractive indices of the air and the pillar materials generates the



**Figure 4. Reflectance Spectra of 2D PC Structures made from Material with two Different Refractive Indices (a) based on Specific Pillar Parameters (b)**

phase shift of the reflected light. A greater difference in the refractive indices between the air and the pillar materials creates a greater phase shift of the reflected light. The degree of phase shift influences the coupling and escape time of the light in the PC structure. Therefore, the higher refractive index of pillar material yielded better reflectance in the normal direction, as shown in this simulation.

Next, the simulation was repeated with changes to determine the best reflectance spectrum of the 2D PC structure using TiO<sub>2</sub> (refractive index 2.3) material. The pillar height and pillar radius were varied to obtain the proper and best reflectance spectrum required for QDs application. The simulation results are given in Figure 5. Note that the choice of a 300 nm center-to-center distance yields good reflectance in the visible region.

The results for simulation in the normal direction of a 2D PC structure with different pillar heights ( $t = 50$  nm, 150 nm, 250 nm, 300 nm, and 500 nm) are shown in Figure 5a. There is no significant relationship between PC height and the reflectance spectrum in these simulation results. For real application, a high and broad reflectance spectrum was required. On the one hand, in the blue color region (wavelengths from 400-480 nm), a PC structure 50 nm in height gives good reflectance. On the other hand, in the green to red color region (wavelengths from 510-660 nm), a PC structure 300 nm in height yields a good, broad reflectance spectrum. Although the other PC structures give high reflectance, they have a narrow reflectance spectrum that is not good enough for photonic device applications. Further simulation was done using PC structures 300 nm in height, since their high, broad reflectance offers potential for photonic devices. The simulation was then carried out with different radii of pillars to obtain the proper and best parameters.

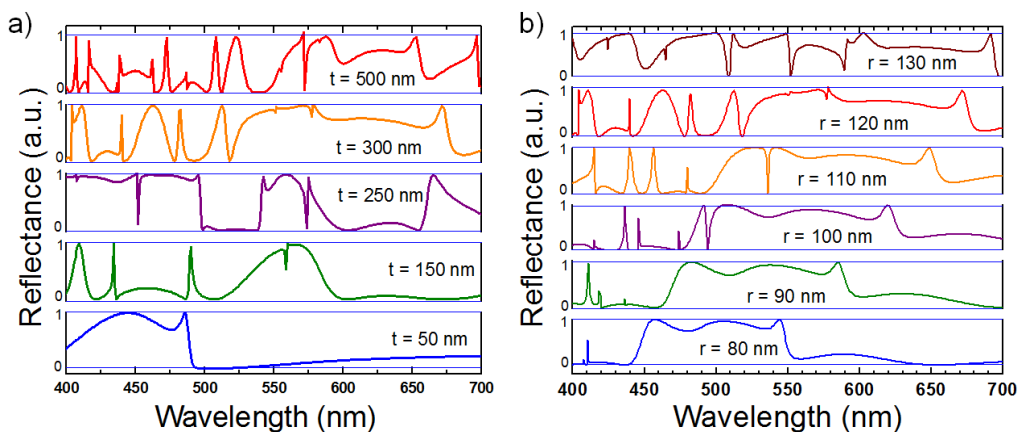
The simulation results from PC with a pillar height of 300 nm and with different radii are given in Figure 5b. The pillar radius varied from 80 nm to 130 nm with intervals of 10 nm. There is a clear relationship between pillar radius and reflectance spectrum. High, broad reflectance is obtained in every simulated reflectance spectrum, except at a radius of 130 nm, when there is no clear reflectance spectrum. Broad reflectance shifts to longer wavelengths as the pillar radius increases. This simulation gives a great potential PC structure for applications. Based on the simulation result, it was decided to fabricate a PC structure with  $\text{TiO}_2$  material, in a hexagonal array of pillars, with a center-to-center pillar distance of 300 nm, a pillar height of 300 nm and a pillar radius of 120 nm.

The fabrication of PC structures was shown in detail in Figure 3a. This step-by-step process was monitored using a scanning electron microscope (SEM). The SEM

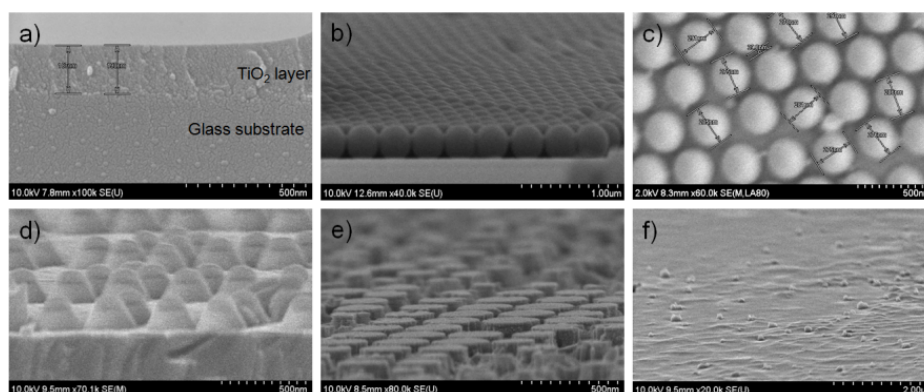
image at every step is given in Figure 6. The  $\text{TiO}_2$  layer was successfully deposited on the glass substrate. After one deposition process, a thin layer of  $\text{TiO}_2$  from 160 to 190 nm thick was achieved, as shown in Figure 6a. The boundary between the glass substrate and the  $\text{TiO}_2$  layer was clearly observed. The fabricated layer was solid, hard and thin. In order to obtain the 300 nm thickness of the  $\text{TiO}_2$  layer, the deposition was done twice. After  $\text{TiO}_2$  deposition, a monolayer of polystyrene spheres 300 nm in diameter was deposited. A bird's-eye view of polystyrene deposition is shown in Figure 6b. The polystyrene monolayer had a hexagonal closely packed structure. The diameter of the polystyrene spheres was then reduced using the RIE etching process. The reduced polystyrene spheres still maintained a hexagonal closely packed structure after this, as shown in Figure 6c. These spheres were then used as a mask for the  $\text{TiO}_2$  layer, using the RIE etching process. During this etching process, the open part of the  $\text{TiO}_2$  layer was etched and the  $\text{TiO}_2$  layer under the polystyrene sphere part survived, as shown in Figure 6d.

Unfortunately, during the etching process, the spheres were etched at an etching rate slower than that of the  $\text{TiO}_2$  layer.

Therefore, after removing the polystyrene spheres, we obtained the truncated pillar shapes shown in Fig 6.e. The top diameter of the pillar is a little bit smaller than the bottom diameter of the pillar. This truncated pillar shape is difficult to avoid when polystyrene spheres are used as a mask layer. The advantage of this technique is that it is a fast and cheap method to obtain a large area of a patterned sample. Apart from that, the pillar array nonetheless maintains a hexagonal closely packed structure. Although a  $\text{TiO}_2$  2D PC structure was indeed formed, imperfections occurred in some places. Nevertheless, the  $\text{TiO}_2$  2D PC structure was successfully and completely fabricated. For comparison, a flat  $\text{TiO}_2$  layer with a similar thickness to that of the PC, shown in



**Figure 5. Simulated Reflectance Spectra for 2D PC Structures with Different Pillar Heights (a) and Different Pillar Radii (b)**

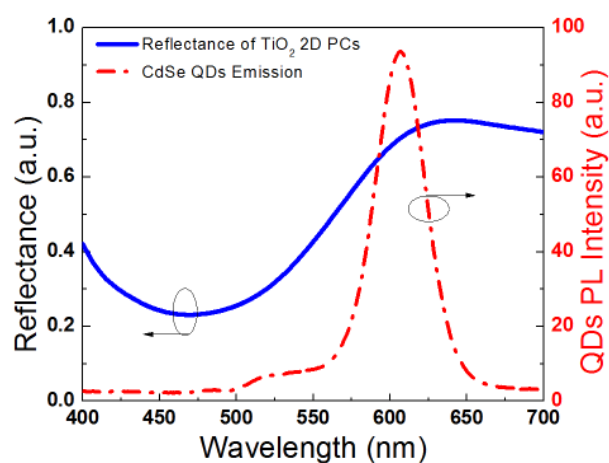


**Figure 6.** The TiO<sub>2</sub> Layer Fabricated using the Sol-gel Method (a); Closely Packed Polystyrene Spheres (b); Etched Polystyrene Spheres using RIE (c); the Etched TiO<sub>2</sub> Structure using RIE (d); the Final Product, a 2D PC Structure (e) and a Flat TiO<sub>2</sub> Layer as a Reference Layer (f)

Figure 6f, was fabricated. This flat TiO<sub>2</sub> layer was used as a reference layer for TiO<sub>2</sub> in order to measure the reflectance of the PC structure and the total reflected emission of the CdSe QDs layer.

Optical measurements were carried out to obtain the reflectance spectrum of the fabricated 2D PC structure, as shown by the solid blue curve in Figure 7. The reflectance spectrum of the TiO<sub>2</sub> 2D PC structure was obtained using the flat TiO<sub>2</sub> layer to provide a reference reflectance spectrum. The reflectance of the 2D PC is low, with wavelengths from 400 to 520 nm. The reflectance increases at wavelengths beyond 550 nm. We concluded that our fabricated 2D PC was a good reflector in the normal direction at wavelengths ranging from 550 to 700 nm. The measured reflectance spectrum had the same trend as the simulated reflectance. High reflectance occurred in the green to red color region. Due to the imperfections in the fabricated 2D PC, its measured reflectance spectrum was not exactly the same as that of the simulated one. The imperfections in the PC structure included the truncated height of the pillars and imperfections in the regularity of the hexagonal array. Since high reflectance occurs at wavelengths of 550 to 700 nm, the red CdSe QDs, which emit light with a wavelength of 605 nm, were used to show that the fabricated 2D PC was a good reflector in the normal direction.

The emission of the CdSe QDs was measured using the set-up shown in Figure 3b. First, the CdSe QDs emission with the flat TiO<sub>2</sub> layer was measured. This was the reference QDs emission. The emission is shown as a solid thick red dotted line in Figure 8a. The emission peak wavelength was around 607 nm. After that, the flat TiO<sub>2</sub> layer was replaced by the TiO<sub>2</sub> 2D PC structure and the QDs emission was measured again. The QDs emission onto the TiO<sub>2</sub> 2D PC structure increased significantly, as shown by the solid thin blue line in Figure 8a. This increase clearly shows that the 2D PC structure acted

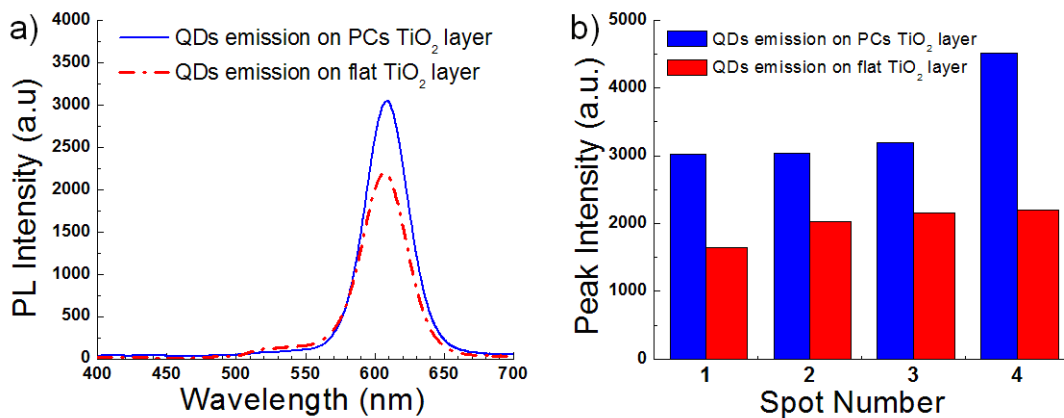


**Figure 7.** Reflectance Spectrum of the 2D PC Structure (Solid Blue Curve) and the QDs Emission Spectrum (Dotted Red Curve)

as a good reflector in the normal direction. The emission peak wavelength was around 609 nm, which is shifted by 2 nm from the reference emission. This shift is clearly due to the reflectance spectrum of the 2D PC structure. In the region of the QDs emission, the reflectance of the 2D PC structure at a longer wavelength is slightly higher than that at a shorter wavelength. This pushes the reflected QD emission spectrum shifted slightly to a longer wavelength. This finding is similar to our previous finding on the effect of a PC structure on QDs emission [12]. Here, we obtained better reflectance than in previous work [1,2]. In addition, our fabrication technique was able to produce a large area of a 2D PC structure using a simpler fabrication method.

In addition, QDs emissions onto the flat TiO<sub>2</sub> layer and onto the TiO<sub>2</sub> 2D PC structure were measured at four different positions. The comparison of QDs emission peak intensities is shown in Figure 8b. The QDs emission





**Figure 8. The QDs Emission Spectrum with a 2D PC Structure and a Flat TiO<sub>2</sub> Layer (a) and Increase in the QDs Emission for several QDs Spots (b)**

is always higher onto the 2D PC structure. The increase in the measured QDs emission varies from 48 to 105%. The degree of perfection of the 2D PC structure influences the ability of the 2D PC structure to function as a reflector. The ability of the 2D PC structure to reflect light in the normal direction could lead to great photonic devices. However, further development is still required to fabricate perfect 2D PC structures that cover a large area.

## Conclusions

The simulation and experimental study of 2D PC structures as reflectors in the normal direction have been covered in this research. The simulation was used to determine proper parameters for the 2D PC structure. It was found that the 2D PC structure could be applied as a reflector when the 2D PC was made of TiO<sub>2</sub> material in a hexagonal array of pillars, with a pillar-to-pillar distance of 300 nm, a pillar height of 300 nm and a pillar radius of 120 nm. The fabrication of 2D PC using simple sol-gel and polystyrene-sphere-masking techniques produced a functional 2D PC structure. Although some imperfections mark the 2D PC structure due to the fabrication process, the TiO<sub>2</sub> 2D PC structure reflects the QDs emission in the normal direction, as expected. An increase in QDs emission of up to 105% was achieved due to the high reflectance of the 2D PC structure. Therefore, a simulation and experimental evidence have revealed that a 2D PC structure acts as a reflector for normal incident light. Our finding opens potential applications of 2D PC structures to improve photonic devices, i.e. increasing light extraction efficiency in light emitting diodes, increasing sensitivity of bio-sensors, etc.

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