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Modification of spontaneous emission rate of micrometer-sized light sources using hollow-core photonic crystal fibers*

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We investigate numerically and experimentally the modification of the spontaneous emission rate for micrometer-sized light sources embedded in a hollow-core photonic crystal fiber (HCPCF). The diameter of the light source is deliberately chosen such that they could be easily introduced into the central hole of the hollow-core photonic crystal fiber by capillary force. The photoluminescence from the microparticles is measured by using an inverted microscope in combination with a spectrometer. The modification of the spontaneous emission rate is observed in a wavelength region where there is no band gap. The experimental observations are consistent with the simulation results obtained by the plane wave expansion and finite-difference time-domain techniques.

Keywords: hollow-core photonic crystal fiber, spontaneous emission rate, local density of states
PACC: 4270Q, 4281D, 4280L, 4230L

1. Introduction

Early in 1946, it was indicated by Purcell that the spontaneous emission rate of an emitter (e.g. an atom or a molecule) depended not only on its energy structure but also on the optical properties of the surrounding environments.[1] In 1987, Yablonovitch suggested that the inhabitation of the spontaneous emission could be realized in the stop bands of photonic crystals (PCs) which periodically modulate the dielectric constant or the refractive index.[2] Since then, much effort has been made to control the spontaneous emission with PCs because of its potential applications in functional devices such as microcavity lasers, single photon sources, etc.[3−6] Basically, the emission properties of light sources, such as the emission rate, can be altered by changing the optical properties of the surrounding environments. It has been known that the spontaneous emission rate of an emitter is determined by the so-called local density of states (LDOS).[7,8] However, the LDOS characterizes only the total spontaneous emission rate. In order to describe the angular dependence of the emission behaviour, we have to use another physical quantity which is called fractional local density of states (FLDOS).[9]

In the previous studies, various PC structures have been employed to control or modify the emission properties of micrometer- or nanometer-sized light sources such as dye molecules and semiconductor quantum dots (QDs).[10,11] These PC structures include semiconductor microcavities with high quality factors, three-dimensional (3D) PCs composed of polymers, hollow-core PC fibers (HCPCFs), etc. Since the spontaneous emission rate of an emitter is determined by the LDOS,[7,8] it is necessary to accurately introduce the emitter into the specified position of a PC in order to realize the suppression or enhancement of the spontaneous emission rate. In the case of self-assembled QDs embedded in a PC cavity, a technique where the strain memory effect of vertically aligned QDs is used, has been developed to introduce a marker on the surface of the wafer for further processing the PC cavity.[12] Colloidal PCs with pseudogaps, which are self-assembled from the suspension of polystyrene (PS) spheres, have also been employed to investigate the spontaneous emission modi-

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fication of dye molecules.[9] In this case, dye molecules are introduced into the PCs by liquid infiltration and they are randomly distributed on the surfaces of PS spheres. As a result, the observed modification in frequency spectrum is an average value of all excited dye molecules located at different positions.

HCPCFs are a kind of PCFs where photonic bandgap effect is used to guide light in air (or in the central air hole).[13] This unique feature makes them quite attractive for both fundamental research and device applications. For example, enhanced stimulated Raman scattering has been observed in HCPCFs and various sensors have been built with HCPCFs.[14,15] Therefore, it is expected that they can also be conveniently employed to study the spontaneous emission modification, thereby some potential applications may be found in device fabrication. In this case, the key issue is how to selectively introduce micrometer- or nanometer-sized emitters into the central holes of HCPCFs. To do so, a technique where the surrounding air holes are selectively blocked, has been developed but an accurate control is necessary.[16]

In the present article, we demonstrate a simple and effective way to investigate the modification of the spontaneous emission rate by using a commercially available HCPCF (Air-6-800, Crystal Fibre) and luminescent microparticles (Eu-doped YVO_4). The diameter of the microparticles is deliberately chosen so that they can be easily introduced only into the central hole of the HCPCF. The modification of the spontaneous emission rate, which is symbolized as the change of the photoluminescence (PL) spectral shape, is observed in the emission band of the microparticles where no band gap exists in the HCPCF. The experimental observations are in good agreement with the simulation results based on the plane wave expansion (PWE) and finite-difference time-domain (FDTD) techniques.[17]

2. Numerical simulations based on the PWE and FDTD techniques

The HCPCF used in our study is designed for guiding light in a wavelength range of 760–800 nm in which the transmission loss is as low as 0.2 dB/km. Apparently, micrometer- or nanometer-sized light source emitting in this wavelength range is the best candidate for studying the modification of the spontaneous emission rate. However, only semiconductor core-shell QDs with a diameter of several nanometers are available in this spectral region. Most micrometer- and nanometer-sized light sources such as dye molecules and particles doped with rare earth ions emit in visible region. Therefore, we have carefully examined the band structure of the HCPCF and the spontaneous emission rate of a point source located in the central hole of the HCPCF by numerical simulations based on the PWE and FDTD techniques.

The cross section of the HCPCF observed by scanning electron microscope (SEM) is shown in Fig.1. It clearly shows the two-dimensional (2D) triangular lattice of air holes with a period of 1.6 μm running down the fiber. The diameter of the central air hole, the surrounding air holes, and the fiber are 6, 1.46, and 125 μm, respectively. According to the PWE method, we can easily derive the band structure of the 2D PC as shown in Fig.2. Figure 2(a) shows the band diagram in a wide wavelength range where no complete bandgap is observed. The enlarged band structure of the 2D PC in visible region is shown in Fig.2(b). As compared with the band structure in infrared region, the energy bands (or dispersion curves) in a visible region appear to be dense and flat. However, it is difficult to judge whether an enhancement (or suppression) occurs at a certain frequency.

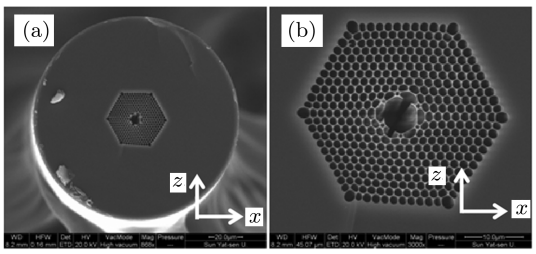

Fig.1. SEM images for the cross sections of the HCPCF (a) and the enlarged 2D PC structure (b).

Before moving to the experiments, we characterize the spontaneous emission rate of a point source located in the central hole of the HCPCF by using the FDTD simulation. Since the modification of the spontaneous emission originates mainly from the 2D PC in the transverse direction, 2D simulations are carried out for simplicity. The origin of the coordinate is set at the centre of the central air hole while the horizontal and the vertical directions are chosen to be x and z axes respectively. The grid sizes in both directions

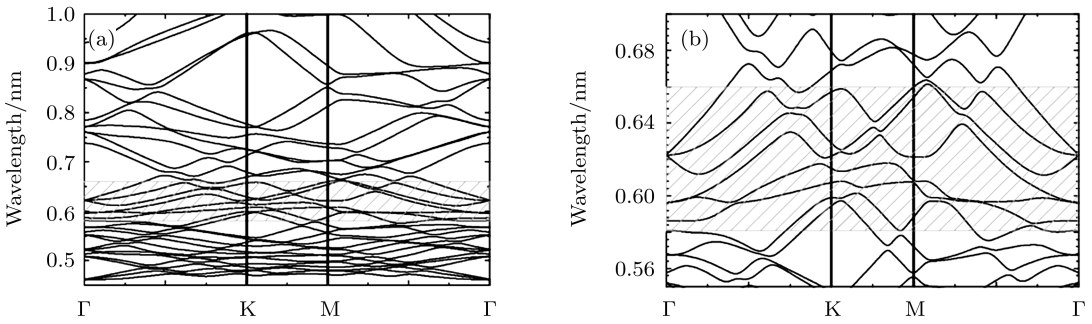

Fig.2. (a) Band structure of 2D triangular lattice of air holes in the HCPCF. (b) Enlarged band diagram in visible region. In both cases, the shadow area indicates the wavelength range corresponding to the emission band of the luminescent microparticles.

(x and z) are chosen to be 0.05 μm and a perfectly matched layer boundary condition is employed. In the simulation, a transverse-magnetic-polarized point source is located at different positions of the central hole and power detectors with different sizes of L = 6.3, 16, and 26.62 μm are aligned in the horizontal direction (x direction in Fig.1). The finite size of the power detector corresponds to the numerical aperture (NA) of the objective lens used in the experiment. The simulation results for point sources located at different positions are presented in Fig.3. In all cases, a reduction in detected power is observed at ∼620 nm while enhancements in detected power are found to be at ∼600 and ∼640 nm. Therefore, it is expected that the modification of the spontaneous emission rate can be observed for the luminescent microparticles embedded in the HCPCF.

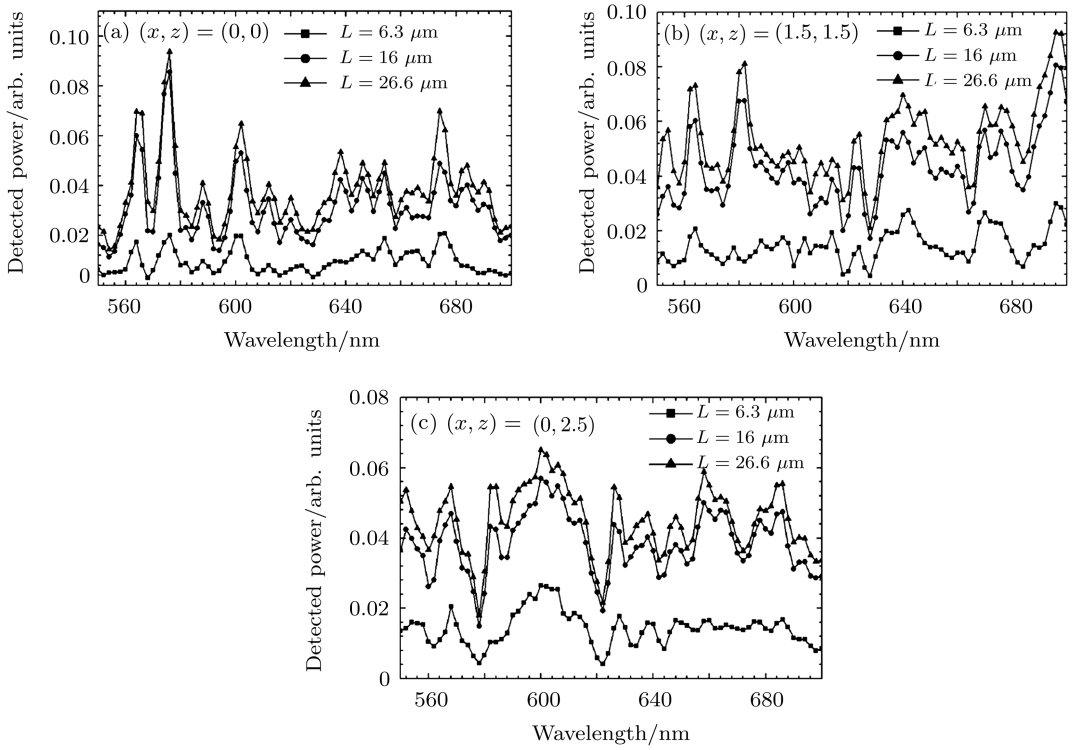

Fig.3. Wavelength dependence of the detected power for a point source located at different positions in the central hole of the HCPCF. (a) (x, z) = (0, 0); (b) (x, z) = (1.5, 1.5); (c) (x, z) = (0, 2.5). The size of the power detector L corresponds to the numerical aperture of the objective lens used in the experiments.

3. Experimental setup and results

In the experiment, we chose Eu-doped YVO_4 microparticles as emitters because of three reasons. First, the diameter of the microparticles ($\sim$5 μm) is smaller than that of the central air hole but larger than that of the surrounding air holes. Therefore, we can easily introduce them into the central hole by infiltrating the aqueous solution of the microparticles into the HCPCF. Second, the emission band of the microparticles is centred at $\sim$620 nm where a significant modification of the spontaneous emission rate is anticipated based on the simulation results. These two features make them quite suitable for the investigation of the spontaneous emission control by using the HCPCF. Finally, we point out that in our study the modification of the spontaneous emission rate in the high-energy bands of the HCPCF, where both enhancement and suppression may occur, is performed in the present study, which is different from that in the previous studies where the suppression of the spontaneous emission rate in the band gaps of PCs is investigated.

The excitation and the measurement of the PL of the microparticles included into the HCPCF are carried out by using a solid-state laser (532 nm) and an inverted microscope (Zeiss Observer A1) connected to a spectrometer. The experimental setup is schematically shown in Fig.4. In order to find out the angular dependence of the PL, objective lenses with different magnifications (5×, 10×, and 20×) or different NAs are used to collect the PL emerging from the HCPCF.

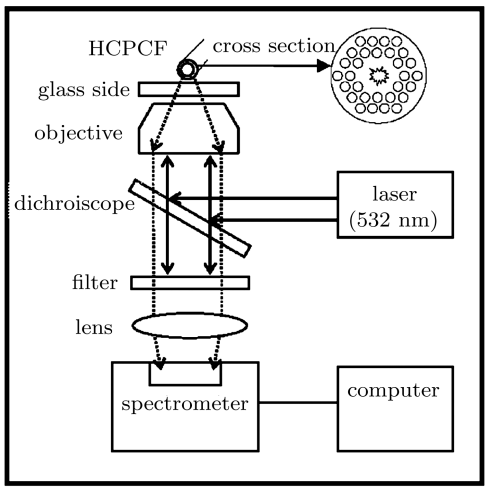

Fig.4. Schematic showing the experimental setup used to investigate the spontaneous emission modification of luminescent microparticles induced by an HCPCF.

In Fig.5, we show the photograph of the HCPCF with inserted luminescent microparticles recorded by using the charge coupled device (CCD) connected to the microscope. It can be seen that the microparticles located in the central hole of the HCPCF emit PL under the excitation of laser light. First, the PL of the microparticles on a glass slide was measured, and the spectra obtained by using objective lens with different NAs are compared in Fig.6. Then, we carried out PL measurements for the microparticles located in the HCPCF, and the results obtained by using objective lenses with different NAs are presented in Fig.7 (solid curves). In each case, the PL from the microparticles located on a glass substrate is also provided for comparison (dot curves). From the spectral shape, it follows obviously that the PL of the microparticles is significantly modified by the HCPCF. In addition, a weak angular dependence of PL is observed with the increase of collection solid angle. A detailed discussion will be given in the next section.

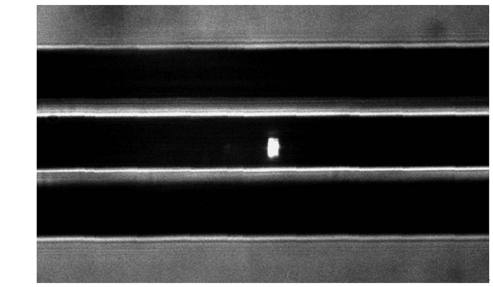

Fig.5. Photograph of the HCPCF with inserted luminescent microparticles.

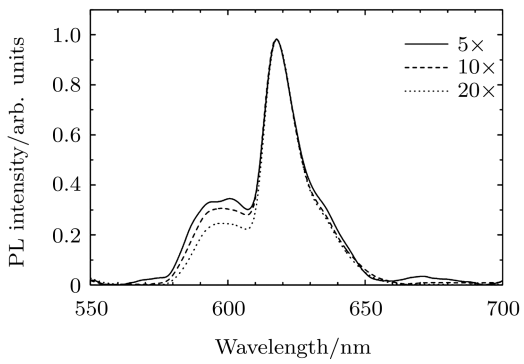

Fig.6. PL spectra for microparticles on a glass slide measured by objective lenses with different magnifications.

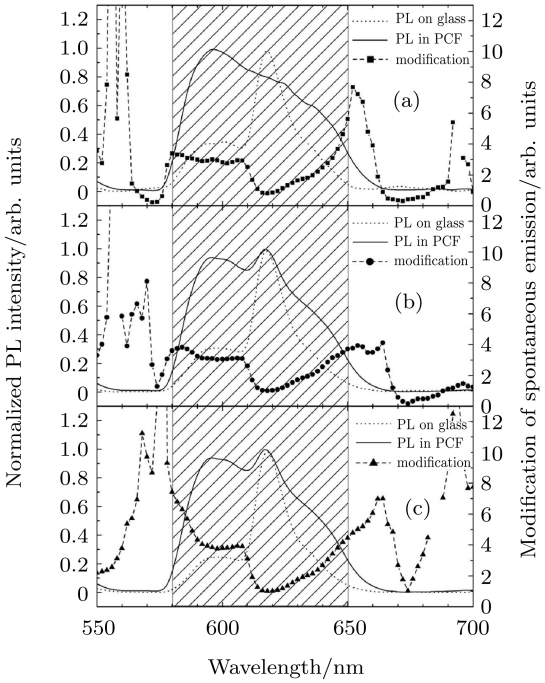

Fig.7. PL spectra for microparticles located in the HCPCF measured by objective lenses with different magnifications. (a) 5×; (b) 10×; (c) 20×. In each case, the PL spectrum for microparticles placed on a glass slide is also provided for reference. Both of them are normalized in order to compare the spectral shape.

4. Discussion

In order to clearly show the enhancement and the suppression of the spontaneous emission rate induced by the HCPCF, we have normalized the PL spectra obtained for microparticles located in the HCPCF and those located on a glass slide as shown in Fig.7. The modification of the spontaneous emission rate is symbolized as the change of the spectral shape. On the glass slide, the PL from the microparticles is dominated by a narrow peak centred at ∼620 nm. In addition, it exhibits a weak dependence on the collection solid angle when objective lenses with different NAs are used. A small change is observed at the left shoulder of the emission band (∼600 nm) as shown in Fig.6. When microparticles are included into the HCPCF, however, the spectral shape is modified dramatically. The PL intensities on both the short- and the long-wavelength sides of the emission band are enhanced while that around the peak is suppressed. Consequently, the spectral width is significantly broadened. In addition, the PL peak originally appearing at 620 nm disappears when an objective lens with a small NA (5×) is used. It becomes resolved again when objective lenses with large collection solid angles (10× or 20×) are used. Therefore, a suppression of the spontaneous emission rate is identified in a wavelength region of 610–630 nm which coincides with the attenuation of the spontaneous emission rate predicted by numerical simulations. Moreover, an enhancement of the spontaneous emission rate is observed at 610 and 640 nm which is also in agreement with the simulation results. Since the modification (enhancement or suppression) of the spontaneous emission rate is symbolized as the change of spectral shape, we can find out the relative enhancement (or suppression) over the entire spectral range by dividing the PL spectra obtained for microparticles located in the HCPCF by those obtained for microparticles placed on the glass slide. From the modification of the spontaneous emission rate shown in Fig.7, one can say that the spontaneous emission rates on both sides of the emission band are enhanced by a factor ranging from 3 to 8 or the spontaneous emission rate at the peak of the emission band is suppressed by a factor of 3–8.

In the experiment, we have carefully rotated the HCPCF such that the ΓM direction (vertical direction) is coincident with the axis of the objective lens. In this case, the objective lens with a small NA (5×) collects mainly the PL emitting in the ΓM direction. As a result, the suppression of the spontaneous emission observed in this direction is dominant and the original PL peak disappears completely. With the increase of the collection solid angle, i.e. by using the objective lenses with large NAs (10× or 20×), the angular dependence of the spontaneous emission rate becomes less obvious and the original PL peak becomes resolved again in the spectrum.

As mentioned at the beginning, the spontaneous emission rate depends not only on the DOS at the emission frequency but also on the location of the emitter. In addition, it usually exhibits anisotropy in space. In our case, emitters are distributed in a wide region although they are restricted in the central hole of the HCPCF. We have no information of the exact locations of the emitters in the central hole of the HCPCF. Therefore, we have simulated the spontaneous emission rates for single emitters located at some specific positions. The

PL we observed in the experiment is actually the average contribution of a large number of emitters located at different positions. In addition, the excitation conditions and the total number of emitters in the two cases (in the HCPCF and on the glass slide) are different, the modification of the spontaneous emission rate we derived is only a relative value. It means that the spontaneous emission rates on both sides of the emission band are enhanced with respect to that at the peak or the spontaneous emission rate at the peak is suppressed with respect to those on both sides of the emission band.

5. Conclusion

We investigated the modification of the spontaneous emission rate of luminescent microparticles by using commercially available hollow-core HCPCFs. The diameter of the luminescent microparticles is deliberately chosen such that their selective introduction into the central hole of the HCPCF becomes very simple. The modification of the spontaneous emission is clearly symbolized as the dramatic change of the PL spectral shape. The applications of this phenomenon may be found in the fabrication of functional devices.

References

[1] Purcell E M 1946 *Phys. Rev. Lett.* **69** 681

[2] Yablonovitch E 1987 *Phys. Rev. Lett.* **58** 2059

[3] Painter O, Lee R K, Scherer A, Yariv A, ÓBrien J D, Dapkus P D and Kim I 1999 *Science* **284** 1819

[4] Englund D, EdoWaks D F, Solomon G, Zhang B Y, Nakaoka T, Arakawa Y, Yamamoto Y and Vučković J 2005 *Phys. Rev. Lett.* **95** 013904

[5] Wang J, Yang D and Zhang H Z 2005 *Chin. Phys.* **14** 232

[6] Liu G Q, Wang J and Zhang H Z 2005 *Chin. Phys.* **14** 102

[7] Li Z Y, Lin L L and Zhang Z Q 2000 *Phys. Rev. Lett.* **84** 4341

[8] Wang X H, Wang R, Gu B Y and Yang G Z 2002 *Phys. Rev. Lett.* **88** 093902

[9] Barth M, Gruber A and Cichos F 2005 *Phys. Rev.* B **72** 085129

[10] Happ T D, Tartakovskii I I, Kulakovskii V D, Reithmaier J P, Kamp M and Forchel A 2002 *Phys. Rev.* B **66** 041303

[11] Lodahl, van Driel A F, Nikolaev I S, Irman A, Overgaag K, Vanmaekelbergh D and Vos W L 2004 *Nature* **430** 654

[12] Hennessy K, Badolato A, Petroff P M and Hu E 2004 *Photonics and Nanostructures* **2** 65

[13] Bjarklev A, Broeng J and Bjarklev A S 2003 *Photonic Crystal Fibres* (Kluwer Academic Publisher)

[14] Yan H, Gu C, Yang C, Liu J, Jin G, Zhang J, Hou L and Yao Y 2006 *Appl. Phys. Lett.* **89** 204101

[15] Jin W and Ho H L 2005 *Advanced Materials and Devices for Sensing and Imaging* II Wang A, Zhang Y and Ishii Y (eds.) in *Proc. SPIE* **5633** 157

[16] Myers S J, Fussell D P, Dawes J M, Mägi E, McPhefram R C, Eggleton B J and Martijin de Sterke C 2006 *Opt. Express* **14** 12439

[17] In this article, a commercially available software developed by Rsoft Design Group is used to perform PWE and FDTD simulations (http://www.rsoftdesign.com)