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### Citation
Optics Express, 21(9):10651-10658

### Issue date
2013

### Resource Type
Journal Article / 学術雑誌論文

### Resource Version
publisher

### DOI
10.1364/OE.21.010651

### URL
http://www.lib.kobe-u.ac.jp/handle_kernel/90002583
Numerical analysis on the feasibility of a multi-layered dielectric sphere as a three-dimensional photonic crystal

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Abstract: The radiative decay rate of a dipole emitter inside the core of a multi-layered dielectric sphere is theoretically investigated. It is shown that, when the thickness of each layer coincides with a quarter wavelength, the multi-layered sphere has a great potential to work as a three-dimensional photonic crystal with a high quality factor and a small mode volume. From the analysis of the dipole position dependence of a radiative decay rate, we show that a smaller core radius, a quarter wavelength at the smallest, is more suitable for real applications. The investigation on the tolerance for thickness nonuniformity reveals that the thickness variation of 10% is tolerable.

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OCIS codes: (160.5293) Photonic bandgap materials; (160.5298) Photonic crystals; (160.1245) Artificially engineered materials.

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1. Introduction

A three-dimensional photonic crystal is an artificially engineered periodic dielectric material that exhibits bands and gaps in the photonic density of states [1–4]. The unique property of the photonic bandgap, i.e., the prohibition on the propagation of electromagnetic waves for all wavevectors, enables to control light with amazing facility and produce effects that are impossible with conventional optics. For example, by introducing artificial defect and/or light-emitters inside the photonic bandgap material, various scientific and engineering applications, such as control of spontaneous emission [1, 2], thresholdless lasers [3], and single photon source [4] can be realized.

A multi-layered sphere, consisting of a core sphere covered by concentric spherical dielectric layers (spherical stack), is a class of a three-dimensional photonic crystal. When the optical thickness of each layer coincides with a quarter wavelength, the stack works as a Bragg mirror and can confine photons three-dimensionally inside the core sphere. The multi-layered sphere potentially has much higher quality factors (Q) compared to a bare microsphere because the stack permits the modes with stronger radial oscillation [5]. Due to the complete point symmetry about the center of the sphere, the photonic density of states inside the core can be designed with higher flexibility than in other conventional photonic crystals. In fact, a complete photonic band gap in the visible region can be realized in a multi-layered sphere with refractive index contrast in the stack as low as 0.15 [6], which is one order of magnitude smaller than that required for the conventional photonic crystals with a diamond structure [7].

Experimentally, due to a rapid progress in the field of nano technology, there have been numerous reports on the coating of dielectric spherical layers on the surface of nanoparticles by chemical solution processes [8–13]. However, so far, the thickness of each layer has not
been carefully controlled so that the modification of the photonic mode density of states could not be achieved. Theoretically, in 1993, Brady et al. [14] studied electromagnetic modes inside a multi-layered sphere without a radiating dipole. In 1994, Sullivan et al. [15] calculated the radiative decay rate of a radiating dipole placed at the center of a multi-layered sphere. In 2005, Moroz [16] presented a general solution for the radiative decay rate of a radiating dipole placed in an arbitrary position in a multi-layered sphere. The algorithm has no limitation on the dipole position, the number of layers, the layer thickness and the layer medium. By employing the algorithm, he calculated the radiative decay rate of a dipole placed in a three- or four-layered sphere consisting of metal and dielectric layers [16, 17].

In this work, we apply the algorithm to a multi-layered sphere, consisting of quarter-wave dielectric concentric layers. We show that it has a great potential to work as a three-dimensional photonic crystal with a high $Q$ and a small mode volume ($V$). In addition, we study the tolerance for the position of the dipoles and the variation of the layer thickness to achieve the high performance. We demonstrate that the tolerance is large enough to be realized by using the state of the art chemical synthesis technology.

2. Calculation procedure

A normalized radiative decay rate, which is defined as a radiative decay rate of a dipole emitter inside the multi-layered sphere with respect to that in the free-space medium, can be calculated by a procedure described in Ref. [16]. First, a dipole is placed in the core of multi-layered sphere and the electric and magnetic fields were calculated by a recursive transfer method. Then the time-averaged total radiated power (radiative loss) was calculated by integrating the time-averaged Poynting vector over the surface of a sphere with an infinite radius. The ratio of the calculated radiative loss to that of a dipole placed in a free space results in the normalized radiative decay rate (Eq. 126 in Ref. [16]). In the case that the dipole is placed off the center of the sphere, the normalized radiative decay rate depends on the orientation of the dipole. The orientation average of the normalized radiative decay rate ($\Gamma/\Gamma_0$) was calculated by $\Gamma/\Gamma_0 = 1/3\Gamma_\perp/\Gamma_0 + 2/3\Gamma_\parallel/\Gamma_0$, where $\Gamma_\perp/\Gamma_0$ and $\Gamma_\parallel/\Gamma_0$ are the normalized radiative decay rate of the radially- and tangentially-oriented dipoles. Note that $\Gamma/\Gamma_0$ corresponds to the well-known...
Purcell factor.

3. Results and discussion

Figure 1 shows a schematic of a multi-layered sphere. It consists of a core covered by concentric spherical dielectric layers. In this work, the refractive index of the core ($n_c$) is fixed to 1.91, which corresponds to that of yttrium oxide (Y$_2$O$_3$) [18]. The spherical dielectric layers consist of alternate high- and low-index layers (HLs and LLs). The first layer on the surface of the core is a LL. The refractive index of LLs ($n_L$) is fixed to 1.45, which is equal to that of silicon dioxide (SiO$_2$) [19]. The refractive indices of HLs ($n_H$) are 1.61, 1.91 or 2.50, which corresponds to those of aluminum oxide (Al$_2$O$_3$), Y$_2$O$_3$, and titanium dioxide (TiO$_2$), respectively [18, 20, 21]. The thickness of each layer is a quarter optical wavelength ($\lambda_0/4n_H$ or $\lambda_0/4n_L$), where $\lambda_0$ is designed to be 500 nm. The refractive index of surrounding matrix ($n_M$) is 1.0, assuming that the sphere is in vacuum. Note that all these materials can be synthesized by chemical solution processes [8–12].

Figure 2(a) shows the normalized radiative decay rate of a dipole inside a bare sphere. The sphere radius ($R_s$) is varied from $\lambda_0/4n_c$ to $7\lambda_0/4n_c$. We can see that the normalized radiative decay rate have some peaks, arising from resonances to so called whispering gallery modes. The number of the peaks increases with increasing $R_s$, while the peak intensity is independent of $R_s$. Figure 2(b) shows the normalized radiative decay rate of a dipole placed at the center of
Fig. 3. (a-c) Dependence of $\Gamma/\Gamma_0$ spectra on the number of layers. $n_H$ are chosen to be (a) 1.61, (b) 1.91, and (c) 2.50, respectively. (d) Peak value of $\Gamma/\Gamma_0$, (e) $Q$, and (f) $V$, as a function of the number of layers.

the core coated by 4 layers (2 HLs and 2 LLs). $n_H$ is 1.91. The core radius ($R_c$) is the same as $R_s$ in Fig. 2(a). We can see that when $R_c$ equals to $(2n + 1)\lambda_0/4n_c$ ($n$: integer), the normalized radiative decay rate is the largest at $\lambda_0$ (2.48 eV in energy), while it is suppressed around $\lambda_0$ when $R_c$ coincides with $2n\lambda_0/4n_c$. This result is qualitatively similar to the case of an one-dimensional Bragg resonator [22]. In the case of $R_c$ of $(2n + 1)\lambda_0/4n_c$, the peak value of the normalized radiative decay rate of a dipole placed in a 4-layered sphere is twice larger than that of a dipole placed in a bare sphere. The coating of only 4 layers has a significant effect on the normalized radiative decay rate.

Figure 3(a) shows the dependence of the normalized radiative decay rate on the number of layers. $n_H$ and $R_c$ are 1.61 and $\lambda_0/4n_c$, respectively. We can see that the peak value of the normalized radiative decay rate at $\lambda_0$ increases with increasing the number of layers. When the number of layers is 20, it reaches about 10. In Figs. 3(b) and 3(c), $n_H$ are changed to 1.91 and 2.50, respectively. The normalized radiative decay rate is larger for larger $n_H$. In Fig. 3(c), the normalized radiative decay rate becomes as high as 40000 at the resonant peak, when the number of layers is 20. In addition, beside the resonant peak, we can see a region where the normalized radiative decay rate is suppressed to as low as 0.0001. The suppression region corresponds to a so called photonic band gap. The size of the band gap, which is defined as the energy difference between the upper and lower band, is as large as 0.9 eV with the center of 2.48 eV. In Fig. 3(d), the peak value of the normalized radiative decay rate is plotted as a function of the number of layers. It grows exponentially with increasing the number of layers and/or $n_H$. The $Q$ and $V$ of the multi-layered sphere are plotted in Figs. 3(e) and 3(f). $Q$ is
obtained by $\omega_0/\Delta \omega$ where $\omega_0$ and $\Delta \omega$ are the resonant frequency and the full width at half maximum of the normalized radiative decay rate spectrum. $V$ is obtained from the following relation [23]

$$\frac{\Gamma}{\Gamma_0} = \frac{3Q(\lambda/4n_c)^3}{4\pi^2 V}$$

where $\lambda$ is the mode’s wavelength. Similar to the normalized radiative decay rate, $Q$ shows exponential dependence on the number of layers. On the other hand, $V$ tends to saturate when the number of layers increases. The higher $n_H$ causes the saturation at smaller number of layers. At the saturation region, $V$ is smaller for higher $n_H$ because higher index contrast can confine electromagnetic fields more strongly. It reaches $2.1(\lambda/2n_c)^3$ for the $n_H$ of 2.5. These results suggest that the multi-layered sphere has a great potential working as a three-dimensional photonic crystal with a high $Q$ and a small $V$.

Figure 4(a) shows $\Gamma_{\perp}/\Gamma_0$ and $\Gamma_{\parallel}/\Gamma_0$ spectra as a function of the position of the dipole ($r$) with respect to $R_c$. $n_H$ and $R_c$ are 1.91 and $5\lambda/4n_c$ respectively. The number of layers is 15. When $r/R_c = 0$, $\Gamma_{\perp}/\Gamma_0$ and $\Gamma_{\parallel}/\Gamma_0$ spectra are identical. With increasing $r/R_c$, the shape of the $\Gamma_{\perp}/\Gamma_0$ and $\Gamma_{\parallel}/\Gamma_0$ spectra becomes different, due to the coupling with different modes inside the multi-layered sphere. Figure 4(b) demonstrates $\Gamma/\Gamma_0$ as a function of $r/R_c$. When the dipole is located at the surface of the core sphere ($r/R_c = 1$), the peak value of the $\Gamma/\Gamma_0$ is one order of magnitude smaller than that of the case of the dipole located at the center ($r/R_c = 0$). This implies that, when $R_c$ is $5\lambda/4n_c$, the control of the emitter position in the core sphere is indispensable for the real application. With decreasing $R_c$, the position dependence of the normalized radiative decay rate becomes smaller.

Figure 4(c) shows the results for the $R_c$
Fig. 5. $\Gamma/\Gamma_0$ spectra as a function of $\sigma$ of layer thickness distributions. $\sigma$ is (a)5%, (b)10%, and (c)20%, with respect to a quarter optical wavelength. (d)Peak value of $\Gamma/\Gamma_0$, (e)$Q$, and (f)$V$ as a function of $\sigma$.

We can see that $\Gamma_\perp/\Gamma_0$ and $\Gamma_\parallel/\Gamma_0$ spectra have the same peak energy and no other peaks appear for any $r$ from 0 to $R_c$. This small dipole position dependence is due to the absence of other electromagnetic modes which can be coupled with the dipole because of the small core radius. In Fig. 4(d), $\Gamma/\Gamma_0$ is plotted as a function of $r$. The peak value of the $\Gamma/\Gamma_0$ decreases only 50% with increasing $r$. This decrease is much smaller than the case of $R_c$ of $5\lambda/4n_c$. In addition, when $R_c$ is $\lambda/4n_c$, there are no peaks other than the main peak in the photonic band gap. These results suggest that fine control of the emitter position is not necessarily required when $R_c$ is $\lambda/4n_c$. It should be noted here that $\lambda/4n_c$ is of the order of several tens nanometers when $\lambda$ is in the visible region. The fabrication techniques of luminescent particles in this size range have been well established in the field of chemical synthesis of nanoparticles in a solution phase [24,25].
Figure 5(a-c) shows the normalized radiative decay rate as a function of the standard deviation of the layer thickness ($\sigma$). $n_H$, $R_c$ and the number of layers are 1.91, $\lambda/4n_c$, and 15, respectively. In Fig. 5(a), solid spectra are the calculation results of $\Gamma/\Gamma_0$ for 10 patterns of randomly-created thickness distributions with $\sigma$ of 5% with respect to a quarter optical wavelength. The dotted spectrum is a result for $\sigma$ of 0%. We can see no significant deterioration of the peak value of $\Gamma/\Gamma_0$ in the case of $\sigma$ of 5%, although the peak positions fluctuate. Figures 5(b) and 5(c) are results for $\sigma$ of 10% and 20%. With increasing $\sigma$, the normalized radiative decay rate spectra broaden and the peak value decreases. In Figs. 5(d), 5(e), and 5(f), the peak value of $\Gamma/\Gamma_0$, $Q$, and $V$, respectively, averaged over 100 patterns of thickness distributions are plotted as a function of $\sigma$. With increasing $\sigma$, the peak value of $\Gamma/\Gamma_0$ and $Q$ decreases significantly, while $V$ increases slightly. In order to keep the deterioration of $\Gamma/\Gamma_0$ smaller than 30% compared to the original value, $\sigma$ should be less than 10% of a quarter wavelength. This requires thickness control with the accuracy of the order of nanometer. This seems feasible to achieve experimentally by the state of the art chemical synthesis technology [26].

4. Conclusion

In conclusion, the radiative decay rate of a dipole emitter inside the core of a multi-layered dielectric sphere has been theoretically investigated. It is shown that, when the thickness of each layer coincides with a quarter wavelength, the multi-layered sphere works as a photonic crystal with a high $Q$ and a small $V$. The dipole position dependence of the radiative decay rate is found to be smaller for smaller core radius, suggesting that the core radius of a quarter optical wavelength, corresponding to several tens nanometers for visible wavelengths, is suitable for applications. It is also found that the thickness variation should be less than 10% to keep the deterioration of the radiative decay rate less than 30%. These results suggest that a chemical synthesis in a solution phase is a feasible fabrication method of the multi-layered sphere as a three-dimensional photonic crystal.