High-Purcell-factor dipolelike modes at visible wavelengths in H1 photonic crystal cavity

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The optimization of H1 photonic crystal cavities for applications in the visible spectral range is reported, with the goal to obtain a versatile photonic platform to explore strongly and weakly coupled systems. The resonators have been realized in silicon nitride and weakly coupled to both organic (fluorophores) and inorganic (colloidal nanocrystals) nanoparticles emitting in the visible spectral range. The theoretical Purcell factor of the two dipolelike modes in the defect has been increased up to ~ 90 , and the experimental quality factor was measured to be ~ 750 . © 2010 Optical Society of America
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Light-matter interaction in nanometer-sized and fully confined systems is a hot research topic at infrared wavelengths and in the visible spectral range [1–4]. When a quantum light emitter, such as a quantum dot, is coupled to an optical resonant mode, the interaction between the two oscillators can lead to two different coupling regimes, known as strong and weak coupling. In weak coupling conditions, the resonant mode modifies the dynamic of the emitter, allowing the engineering of its spontaneous emission. Indeed, the spontaneous emission rate of an emitter weakly coupled to a photonic eigenstate is modified by a factor $F = 3/4 \pi^{-2} Q/V(\lambda/n)^3$, where Q and V are the quality factor and the modal volume of the photonic mode, respectively. If the system is instead in the strong coupling regime, the confined excitons and photons coherently exchange energy with a coupling strength inversely proportional to V , i.e., $g \propto 1/\sqrt{V}$. Therefore, in both regimes the dynamic of the coupled system is strongly dependent on the properties of the photonic mode and, notably, on the electromagnetic field confinement in time and spatial domains.

In order to observe these phenomena at visible wavelengths, different optical resonators have been proposed [2,4], but two-dimensional (2D) photonic crystal (PhC) cavities represent the most promising structures, since they give the best control on the optical properties of the confined system. At present, PhC resonators for applications in the visible spectral range are based on various geometries [5–7] and on higher-order modes of the widely studied single-point defect [8] [also known as an H1 cavity, sketched in Fig. 1(a), inset]. The H1 cavity consists of a missed hole in a triangular PhC lattice, which allows two orthogonally polarized resonant modes in the photonic band gap (hereafter referred to as x - and z -pole modes). The H1 resonator presents several advantages

with respect to other PhC configurations for applications in quantum optics. For instance, the cross polarization of x - and z -pole modes and the absence of higher-order modes can be exploited to satisfy the requirements of quantum information algorithms [9]. As compared with other PhC point defects, the H1 cavity presents the lowest V , thus enhancing QED phenomena in both strongly and weakly coupled systems. The confinement in extremely small modal volumes is more challenging for visible light, since the constraint to use low-absorption materials in this spectral range does not allow recourse to high-refractive-index semiconductor membranes, thus reducing the effectiveness in localizing optical modes. Nevertheless, the aforementioned advantages, together with the increasing interest toward the realization of efficient emitting devices in the visible spectral range, foster theoretical and experimental studies to find alternative routes to improve light confinement in low-index H1 systems.

In this Letter, we theoretically and experimentally investigate a new design strategy to realize H1 PhC defect nanocavities in low-index materials. The confinement in the slab has been significantly improved, while the orthogonally polarized dipolelike behavior of the two photonic states allowed in the H1 defect has been preserved and no other resonant modes introduced.

An H1 cavity has been obtained in a triangular lattice of air holes with period a and radius r in a silicon nitride (Si_3N_4) slab having refractive index $n = 1.93$ and thickness t . The electromagnetic behavior of such a structure was investigated by using plane-wave expansion and 3D finite difference time domain algorithms.

Our approach to realize ultrasmall-volume PhC cavities while keeping high Q factors in the visible range and preserving the dipolelike shape of the

Fig. 1. (Color online) (a) Dependence of modal volume and Q factor on the thickness of the slab t . Inset, photonic crystal H1 cavity. (b), (c) Photonic band structure of the structure for $t=0.7a$ and $t=1.55a$, respectively. (d) Dependence of the Purcell factor on t (for $S=0$). (e) Modification of the resonant frequencies and of the Q factor of the degenerated modes when two cavity neighboring holes are moved, as shown in the inset. The holes are moved closer to (farther from) the center for $S<0$ ($S>0$). (f) E_z for the x -pole mode in an unmodified H1 cavity. (g) E_x for the z -pole mode in an unmodified H1 cavity. (h) E_z for the x -pole mode in an H1 cavity with $S=0.2a$. (i) E_x for the z -pole mode in an H1 cavity with $S=0.2a$.

modes first involves the optimization of the thickness of the PhC slab. As shown in Fig. 1(a) for the x -pole mode, the Q factor is quite constant for $t<1.2a$, and it has a maximum for $t=1.55a$. As reported in Figs. 1(b) and 1(c), the photonic bandgap (PBG) existing for $t=0.7a$ disappears when t is increased to $1.55a$, in agreement with the closing-bandgap principle presented in [3]. This lets us infer that the electromagnetic confinement in the xz plane is not due to the PBG but can be assigned to the mismatch in the momentum space between the cavity mode and the second guided mode in the PhC slab, as described by Tandaechanurat *et al.* [3]. The increased thickness of the slab leads to slight variations of the x - and z -pole modal profiles along y . For this reason, V grows steadily as a function of t , as shown in Fig. 1(a). However, because the modal extension in the xz plane is preserved, these variations of V are negligible with respect to the increase in Q , and thus the Purcell factor [Fig. 1(d)] follows the Q -factor behavior: for $t=1.55a$, F is maximized to $F\sim 78$ with $V\sim 0.68(\lambda/n)^3$ and $Q\sim 700$. A similar trend has been found for the z -pole mode.

The engineering of x - and z -pole modes would foster many applications based on H1 nanocavities operating at visible wavelengths. For instance, the degeneracy of x - and z -pole modes may be useful for

entangled photon generation or quantum cryptography [9]. Other applications, such as single-photon sources or PhC-based optical read out of lab-on-chip devices [10], require well-defined and linearly polarized nondegenerate resonances. Several solutions have been reported in past years to break the degeneracy or to recover it [8,11–13]. A versatile strategy to break the degeneracy is displayed in Fig. 1(e): by moving two cavity neighboring holes, it is possible to change the resonant frequency of the x -pole mode (f_x) significantly without affecting the z -pole mode. This finding is ascribed to the selective modification of the wavevector $\mathbf{k}=[k_x, k_y, k_z]$ along a specific direction. Indeed x - and z -pole modes have the strongest component of $\mathbf{k}$ oriented along the x and z axes, respectively. When two holes are moved closer to the center of the cavity along the x axis ($S<0$, see Fig. 1(e) for definition), thus changing k_x while keeping k_z constant, f_x increases and f_z is unchanged. In the same way f_x decreases for $S>0$, while keeping f_z constant. Figures 1(f)–1(i) show the x - and z -pole modal profiles for an unmodified H1 cavity and for a cavity with $S=0.2a$. The electric field component along x (E_x) of the z -pole mode profile remains unchanged when the holes are moved far from the center [Figs. 1(g) and 1(i)]. The shift instead results in the elongation of the x -pole modal function along x [Figs. 1(f) and 1(h)], thus modifying its resonant frequency.

Such modifications of the field distributions also lead to variations of the Q factors of the modes [Fig. 1(e)]. If $S<0$, abrupt changes are introduced near the maximum of the electric field [1] of the z -pole mode function, resulting in an increase in radiation losses and in a smaller Q factor ($Q\sim 557$ for $S=-0.057a$). In contrast, if $S>0$ these abrupt variations in the modal profile are avoided, the radiative energy in the light-cone minimized, and the Q factor of the z -pole mode enhanced with a V and f_z almost unchanged. The optimized Q factor turns out to be $Q\sim 810$ for $S=0.075a$, and the Purcell factor is assessed as $F\sim 90$. We have therefore verified that momentum space engineering, a strategy exploited to improve confinement of defect states localized within the PBG [1,7], can also be effective for cavity resonances that do not need a PBG.

These theoretical findings have been experimentally demonstrated by exploiting a 2D PhC technology in a Si_3N_4 -air membrane. The PhC geometry was defined in a 400-nm-thick Si_3N_4 film [see Fig. 2(a)], and both cyanine 3 (Cy3) fluorophore and core/shell CdSe/CdS colloidal dot-in-rod nanocrystals [14] have been deposited on the resonator. Room temperature microphotoluminescence characterization was therefore performed.

In order to tune the resonance of the structure, the scalability of the designed PhC was exploited, and several nanocavities, with $r=0.308a$ and different values of a , were fabricated: Fig. 2(b) shows three resonances for three different values of a . The resonant peaks are well approximated by a Lorentzian function [Fig. 2(b)]. For $a\sim 265$ nm, a $Q\sim 620$ is observed for an unmodified H1 cavity.

Fig. 2. (Color online) (a) Bird's eye view of the realized air-bridge structure and (inset) its in-plane view. (b) Resonances obtained by drop casting colloidal nanocrystals on the structure for three different values of a and $r=0.308a$. The inset shows the Lorentzian fitting of the resonant peak for $a=265$ nm. (c)–(f) Resonance of x - and z -pole modes for different values of the hole shift and $a=270$ nm. (g) Resonant frequencies of x - and z -pole modes as a function of the hole shift.

To explore the mode shifting over a wide spectral range, an organic fluorophore (Cy3) with broad emission spectrum was immobilized on the device. Figures 2(c)–2(f) report the microphotoluminescence spectra for different values of S . The z -pole mode is almost unaffected by the shift of the holes, whereas the x -pole mode becomes broadly tunable by changing S . The two modes were identified by performing polarization-resolved measurements, and their resonant wavelengths (λ_x and λ_z , respectively) are displayed in Fig. 2(g) for several values of S . In agreement with the theoretical results of Fig. 1(e), the x -pole mode is tunable over a range of about $\Delta\lambda_x \sim 40$ nm. Small discrepancies between experimental results and theoretical calculations have been observed in terms of slight variations of λ_z and weak nonlinearity of λ_x ; since these variations do not show a clear dependence on S , they could be reasonably attributed to unavoidable fabrication imperfections. The theoretical findings about the influence of the hole's position on the z -pole Q factor have been confirmed by the experiments. For $S=15$ nm $Q \sim 750$ has

been measured, while for $S=-20$ nm the z -pole Q factor reduces to a value of ~ 200 .

In summary, we have reported theoretical and experimental optimization in the visible spectral range of the H1 PhC resonator. The theoretical Purcell factor of the two degenerated modes has been increased up to ~ 90 without introducing high-order states or resorting to other kinds of cavities, such as an $L3$ cavity or double heterostructure, together with a maximum Q factor of ~ 750 .

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