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Post-fabrication control of evanescent tunnelling in photonic crystal molecules

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The post-fabrication control of evanescent tunnelling in photonic crystal molecules is demonstrated through the combination of selective infiltration and oxidation. By laser non thermal oxidation, we reduce the photonic coupling by more than 30% while by means of water micro-infiltration, we increase it by 28%. Fine-tuning of the photonic coupling is achieved by low-power laser oxidation and forced evaporation, opening the route to post-fabrication control of array of coupled cavities. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4767216>]

Photonic microcavities (PMC) coupled through their evanescent field are used for a large variety of classical and quantum devices. Electromagnetically induced transparency like effects can occur in coupled optical resonators due to the coherent interference.^{1–3} Moreover, quantum effects based on coupled PMC such as the quantum phase transition in a PMC array,^{4,5} and coupled-cavity quantum electrodynamics^{6–8} are rising the interest of the photonics community. The basic building block is a system of two adjacent PMCs (denominated photonic molecule). In such systems, a molecular-like spatial delocalization of the coupled modes is achieved by an evanescent tunnelling, whenever the frequency matching between the resonances of each single PMC is fulfilled. This requirement is hampered by disorder associated with the tolerance of fabrication technologies and several post fabrication methods have been developed for compensating this kind of detuning such as tip^{9,10} and thermal tuning,¹¹ wet chemical digital etching,¹² atomic layer deposition,^{13,14} anodic and laser nano-oxidation,^{15,16} and liquid micro-infiltration.^{17–19} The tunnelling rate depends on the height and depth of the photonic barrier between two adjacent resonators and therefore it is sensitive to the fabrication-induced disorder present in the center of the molecule. Moreover, the tunnelling rate is the main parameter in the tight-binding theoretical model for photonic arrays, therefore, its control lead to great progress in the field of quantum information processing.

In this letter, we address the problem of developing a post fabrication control of the tunnelling rate in photonic crystal coupled PMCs. The value of the photonic coupling (proportional to the tunnelling rate) is directly measured by the molecular mode splitting at the anticrossing point. By exploiting a combination of tuning techniques such as local infiltration of water, micro-evaporation, and laser induced non thermal micro-oxidation,^{19–21} we are able to either increase or decrease the detuning and the photonic coupling, independently.

We use a 320-nm-thick GaAs membrane with three central layers of high-density InAs quantum dots (QDs) emitting

at about 1300 nm at room temperature. The structure consists of a two-dimensional triangular lattice of air holes with lattice parameter $a = 311$ nm and filling fraction equal to 35% where the single cavity is formed by four missing holes organized in a diamond-like geometry, denominated D2 cavity as shown in the inset of Fig. 1(a).²² The photonic molecules here considered consist in two adjacent D2 cavities along the K axis of the photonic crystal, as the one reported in the inset of Fig. 1(b). A room temperature commercial scanning near-field optical microscope (SNOM) (Twinsnom, OMICRON) is used in illumination-collection configuration.

FIG. 1. PL near-field spectra of a single D2 cavity (a) and of a K-aligned photonic molecule (b) measured by a SNOM set up. The relative SEM images are shown in the insets. The resonant mode M1 (M2) of the single cavity reported in (a) gives rise to the slightly (widely) split modes labelled as P1 and P2 (P3 and P4) of the photonic molecule shown in (b). In (c) are reported, in wavelength units, the experimental splittings Ω_{P1-P2} (blue circles) and Ω_{P3-P4} (red dots) measured for ten nominally identical K-aligned photonic molecules.

In addition, photoluminescence spectra of the samples were also collected in a confocal microscope using a 50 $\times$ microscope objective with numerical aperture of 0.7. The sample is excited by an Ar⁺ ion laser ($\lambda = 514$ nm) and the emitted PL signal is coupled to a single-mode optical fibre of 6 μ m core diameter, acting as a confocal pinhole, connected to a spectrometer and is finally collected by a liquid nitrogen cooled InGaAs array. Numerical calculations were performed with a commercial three-dimensional finite-difference time domain (FDTD) code (CrystalWave, Photond).

The PL spectra measured with the SNOM set up for a typical single D2 cavity (with two main peaks labelled by M1, M2) and a typical K-aligned D2 photonic molecule (with four main peaks labelled by P1-P4, for decreasing wavelength) are reported in Figs. 1(a) and 1(b), respectively. In the insets, the scanning electron microscopy (SEM) images of the photonic cavities are shown. The experimental splittings Ω_{P1-P2} , between the peaks P1 and P2, and Ω_{P3-P4} , between the peaks P3 and P4, are the most relevant parameters in the PL spectra of the photonic molecule. It is also useful to introduce other two relevant quantities: the photonic coupling g and the mode detuning Δ . Both Δ and g are important in determining the splitting Ω . The simplest coupling model (that also neglects losses) gives the following expression: $\Omega^2 = \Delta^2 + 4g^2$ for the splitting as a function of Δ and g .²³

Going back to the experimental data, it has been already demonstrated²⁴ that the slightly split modes P1 and P2 derive from the M1 resonance of the single D2 and experience a weak coupling with g of the order or smaller of 0.5 nm in wavelength units (the evidence is that each of these modes is spatially localized only on one single cavity of the photonic molecule); while the widely split modes P3 and P4 originate from the M2 mode of the single D2 and on the contrary experience a strong coupling characterized by $g \sim 7$ nm (in fact, these modes are spatially delocalized on the whole system). Therefore, we expect that Ω_{P1-P2} has to be ascribed to the energy detuning (Δ) due to the fabrication induced disorder, and Ω_{P3-P4} is mainly attributed to the photonic coupling g .

We measured the PL spectra on 10 nominally identical coupled PMCs. A summary of the mode splitting Ω in wavelength units for these photonic molecules is reported in Fig. 1(c). As discussed above, the large variation of Ω reported in Fig. 1(c) is due either to variation of the mode detuning or of the mode coupling, both given by dielectric disorder. The value of Ω_{P1-P2} , which mainly reflects the mode detuning, spans from almost 0 to 5 nm. In the case of P3 and P4, the measured variation of Ω_{P3-P4} from 9 nm to 16 nm is a signature of the relevant effects of dielectric disorder on the photonic coupling g in nominally identical photonic molecules. In fact, if it would be incorrectly assumed that the photonic coupling g is constant and independent from the disorder, taking into account the previous formula $\Omega^2 = \Delta^2 + 4g^2$ with $\Omega \sim 16$ nm and $2g \sim 9$ nm, one would get $\Delta \sim 13$ nm, which is more than twice the value obtained for the case of P1 and P2, and this is not a reasonable prediction. The fact that also the photonic coupling g is strongly affected by the fabrication induced disorder would limit the possibility of extended array of identically coupled PMC, unless a post fabrication control of g is implemented.

As a first approach to the control of g , we used the non-thermal laser assisted oxidation by SNOM.^{21,25} We exposed the spatial region in the middle of a photonic molecule in order to perform a symmetric oxidation that should prevent the mode detuning. In addition, we choose a photonic molecule which show a very small value of Ω_{P1-P2} , where the disorder induced detuning is likely quite small. The main effect of the laser exposure is to blue shift all the photonic modes, as shown for the mode P3 in Fig. 2(a). More interestingly, Ω_{P3-P4} shows a large reduction from (14.7 ± 0.2) nm to (10.2 ± 0.2) nm, as shown in Fig. 2(b). This demonstrates that the laser induced micro-oxidation is a powerful tool for adjusting the photonic tunnelling rate.

The laser induced oxidation of GaAs, due to the smaller index of refraction of the oxide, is equivalent to an effective reduction of the GaAs material (giving a blue shift of the mode),²¹ that is a reduction of the slab thickness and an effective enlargement of the crystal pores. In order to understand the physical origin of the g reduction, we performed 3D FDTD calculations either by reducing the slab thickness or by enlarging the diameter of the pores in the central region of the photonic molecule. A summary of the results for P3 and Ω_{P3-P4} are given in Fig. 3, where the slab is reduced from 320 nm to 280 nm [Figs. 3(a) and 3(b)] and the diameter of 11 central pores is enlarged from 193 nm to 213 nm [Figs. 3(c) and 3(d)]. In both cases, all the photonic modes are blue shifted, as shown for P3 in Figs. 3(a) and 3(c), where the reduction of the P3 resonant wavelength is (24.0 ± 0.2) nm and (17.6 ± 0.2) nm, respectively. However, while the slab reduction is almost ineffective in changing Ω_{P3-P4} , a reduction of the calculated Ω_{P3-P4} comparable with the experimental data is obtained when enlarging the pore diameters. We found a decrease of (4.5 ± 0.4) nm that

FIG. 2. The P3 resonant mode (a) and the experimental splitting Ω_{P3-P4} (b) are reported as a function of the Ar⁺ laser exposure time. The exposition is performed through the probe of the SNOM set up kept on the central region of the K-aligned photonic molecule.

FIG. 3. FDTD calculations performed in order to simulate the effect of green laser assisted micro-oxidation on an ideal K-aligned photonic molecule, where no disorder induced detuning is present. The P3 mode and the splitting Ω_{P3-P4} as a function of the effective GaAs slab thickness reduction are reported in (a) and (b), respectively. The P3 mode and the splitting Ω_{P3-P4} as a function of the diameter increase of the eleven central pores of the photonic molecule are reported in (c) and (d), respectively. The error bars correspond to the spectral resolution of the FDTD calculations.

perfectly matches the experimental value. On the contrary, the only enlargement of the pore diameters cannot explain the P3 experimental blue shift of (28.0 ± 0.1) nm. So we need to take into account also the slab thickness reduction whose main effect is to blue-shift the resonant modes.

We conclude that the laser induced oxidation is effective in decreasing the photon tunnelling as a consequence of a reduction of the dielectric filling fraction in the photonic barriers between the two adjacent PMCs. Therefore, we expect that the increase of dielectric in the spatial region between the PMCs would result in increasing the coupling strength g .

In order to confirm this prediction, FDTD simulations have been performed assuming that the central pores of the molecule are completely filled by a medium of index of refraction $n = 1.33$ (the value of water). We performed three different simulations by infiltrating 1, 5, and 11 central pores of the K-aligned photonic molecule, as reported in the schemes of Figs. 4(a)–4(c), respectively.

The photonic molecule modes undergo a red shift due to the increase of the effective dielectric constant of the molecule environment as highlighted for P3 in Fig. 4(d). In Fig. 4(e), the calculated splitting Ω_{P3-P4} is reported as a function of the number of infiltrated pores. We found that g can be increased and that the main contribution is given by filling the single central pore, while increasing the number of infiltrated pores the resonant modes show a continuous red shift, as reported for P3, but do not reveal a sizable variation of Ω_{P3-P4} . This not intuitive effect can be ascribed to the oscillatory behaviour of the evanescent waves in photonic crystals, which makes photonics tunneling quite different from quantum mechanics.²⁶ A simple way to understand the major role of the central pore is to consider the spatial distribution of the electric field associated to the P3 and P4 modes: the P3 mode has a maximum of the electric field at the position of the central pore, while, at that position, the electric field of the P4 mode is almost zero.²⁴ On the contrary, the spatial distribution of the P3 and P4 modes in the other pores is

FIG. 4. (a)–(c) Schematic views of the refractive index profile used in FDTD calculations for the same photonic molecule geometry where 1, 7, and 11 central pores, respectively, are simulated as infiltrated with water: grey represents $n = 3.48$ (GaAs), light blue $n = 1.33$ (water), and black $n = 1$ (air). The P3 resonant mode (d) and the splitting Ω_{P3-P4} (e) obtained by FDTD calculations are reported as a function of the number of the infiltrated pores. The error bars correspond to the spectral resolution of the FDTD calculations.

quite similar. Therefore, the filling of the central pore red shifts only P3 with a net increase of Ω_{P3-P4} , while the filling of the others pores has a similar effect on both modes with not substantial modification of Ω_{P3-P4} .

We therefore performed the micro-infiltration experiment with a diluted solution of water and rhodamine, which helps by removing air bubbles inside the pipette (as can be seen by optical inspection). Heuristically, we observed that this strongly improves in filling efficiently the photonic pores. The micro-infiltration experiments, being associated with the increase of g , are more difficult to control and interpret because the increase of Ω can also be produced by a non symmetric water deposition with consequent increase of Δ . Therefore, we need to directly measure g by anticrossing experiments by acting on Δ in order to reach the zero-detuning condition, because only in this condition $\Omega = 2g$, and thus, we get a direct measure of the coupling strength. For this purpose, we implemented a detailed experimental procedure schematized in Fig. 5(a). In the first step [point 1 in Fig. 5(a)], we infiltrated the right side external region of the photonic molecule, then with the micro PL set up we exposed the same spot with the green laser forcing the water evaporation and inducing local photo-oxidation. This allows to measure the anticrossing curve and thus to extract the initial value of g . In the second step [point 2 in Fig. 5(a)], we performed a symmetric infiltration in the central region of the photonic molecule in order to change g , and finally [point 3 in Fig. 5(a)], we induced a second non symmetric micro-oxidation on the left side of the photonic molecule in order to determine the final value of g .

Experimentally, the first infiltration step was implemented on the very right side of the photonic molecule because the higher energy atomic-like mode (P2) was initially localized on the right cavity, as shown in the micro PL spatial distribution (not shown here). Concerning the first

FIG. 5. (a) Schematic view of the three experimental steps performed to measure the increase of the coupling strength. (b) Ω_{P3-P4} is reported as a function of the exposure time of subsequent lateral micro-evaporation and micro-oxidations, as described in (a). The red dot at negative time corresponds to the initial value before the right-side infiltration. The blue circles represent Ω_{P3-P4} after the right-side infiltration followed by a right side laser illumination. The black dots represent Ω_{P3-P4} after the central infiltration followed by a left side laser exposition.

two modes P1 and P2, after the right-side infiltration, the mode localized on the right cavity is red shifted by 4 nm while the left cavity mode resonance remains almost unchanged, confirming that the micro-infiltration is local and selective. Therefore, the mode on the left cavity, which was the fundamental mode P1 before the infiltration, became the first excited state P2 as due to the infiltration mode tuning; That is, the spatial localization of P1 and P2 is inverted. In this way, we know that the modes of the right cavity are red shifted and we can perform anticrossing measurements by blue tuning the right cavity modes.²⁷ As expected the detuning of P3 and P4 is increased, as shown in Fig. 5(b), where the experimental values of the mode splitting Ω_{P3-P4} are reported before and after the right side micro-infiltration, by the comparison of the first two points (the red dot and the first blue dot). Then, we forced the water evaporation and we induced micro-oxidation by illuminating the same right-side spatial region, where now the P1 mode is localized, with the micro PL set up. In this way, we still act on a single cavity and, by blue shifting the P1 mode, we observed an initial reduction of Ω_{P1-P2} then, once the detuning is perfectly compensated, Ω_{P1-P2} starts to increase. The control of Δ allows us to collect an anticrossing curve [see blue dots of Fig. 5(b)] and therefore to extract very precisely the original value of the coupling strength. We note that Ω_{P3-P4} reaches a minimum after an exposure time of about 130 min that gives $g = (6.6 \pm 0.1)$ nm. At the end of this oxidation, after about 370 min of laser exposure, the detuning for P1 and P2 is 4.5 nm which nicely agrees with the detuning between P3 and P4 calculated from the increase of Ω_{P3-P4} from 13.2 nm to 14.1 nm following the relationship $\Omega^2 = \Delta^2 + 4g^2$. In this

configuration, in order to increase the g value, we infiltrated the central region of the molecule [point 2 in Fig. 5(a)] and Ω_{P3-P4} increases from 14.1 nm to 17.4 nm [see the comparison between the last blue dot and the first black one in Fig. 5(b)]. The new tunnelling rate is then determined by means of a second anticrossing experiment obtained with micro-oxidation of the left-side region of the photonic molecule [point 3 in Fig. 5(a)].

The results are reported by the black dots of Fig. 5(b) where we kept the oxidation time as measured by the beginning of the three steps in order to stress the unity of the entire procedure. Measuring the experimental minimum splitting Ω_{P3-P4} after the micro-infiltration of the central pores we get a coupling strength equal to $g = (8.5 \pm 0.1)$ nm, with an increase of 28%. The values of the photonic coupling g , both before and after the infiltration are in good agreement with the FDTD calculations reported in Fig. 4. However, the FDTD calculations shown in Fig. 4(d) indicate that the photonic tunnelling is very sensitive to infiltration of the single central pore, while the filling of the more external pores has very little effects, and possibly it gives a small reduction of g . Let us remark that this latter effect points out that the increase (reduction) of g by increasing (reducing) the effective dielectric constant of the photonic barrier is not likely a universal result and different behaviours can be expected in different geometries.²⁸

In conclusion, we have achieved a deterministic control of the photon evanescent tunnelling rate in a photonic crystal molecule. By water micro-infiltration, we were able to increase the photon coupling by 28%. On the contrary, by laser induced non thermal oxidation, we got a reduction of g by 30%. The combination of the two methods would therefore give a complete control of g with excellent accuracy. This could make possible the realization of array of photonic cavities with on demand tunnelling rate between each pair of coupled resonators. We believe that this peculiar engineering of photonic crystal molecules would open the road to possible progress in the exploitation of coherent interference between coupled optical resonators both for quantum information processing and optical communication.

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