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Antibonding ground state in photonic crystal molecules

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We employ a far-field analysis of the angular emission pattern to experimentally assess the symmetry of localized modes in coupled photonic-crystal cavities. We demonstrate that the spatial distribution of localized modes in photonic-crystal nanocavities may change from a bonding to an antibonding orbital, a feature that is unusual in quantum mechanical coupled systems. Experimental data are confirmed by numerical calculations and interpreted in terms of the peculiar oscillatory behavior of the evanescent waves in photonic-crystal band gaps.

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The natural and fertile analogy between photonics and quantum mechanics is currently driving the achievement of novel optical and optoelectronic devices based on photonic crystals (PCs). Using PCs, it is possible to tailor the photons in very similarly to the manipulation of electrons in electronics.[1] Photonic band gaps (PBGs), i.e., frequency intervals where light cannot propagate, are the direct counterpart of electronic band gaps. Dielectric defects in photonic crystals introduce strongly localized states in the PBGs similarly to the case of substitutional impurities in crystals, and more generally in the case of atoms.[1,2] The peculiar properties of these dielectric defects in photonic crystals [also known as photonic-crystal cavities (PCCs)] are also very similar to atomic states and they are playing a relevant role in the development of light emitters or optical waveguides.[2]

Following the analogy with quantum mechanics, coupled PCCs are also denominated photonic molecules.[3–5] Both homoatomic and heteroatomic photonic molecules have recently been realized.[6] In the case of homoatomic molecules, the coupling between identical localized photonic modes gives a frequency splitting into two modes of different parities. In analogy to real molecules and to recent reports for quantum dots (QDs),[7] we call the even (usually nodeless) molecular state a bonding state, while the odd molecular state (which always has a node) is called the antibonding state. Intuitively, the molecular ground state is expected to have bonding orbital character and, on the contrary, the first excited molecular state is expected to have antibonding orbital character. Recently it has been shown that the spin-orbit coupling can lead to antibonding ground states in quantum dot molecules.[7] Similarly, it has been theoretically predicted that the ground state may change from bonding to antibonding in photonic-crystal systems by varying either the distance between the PCCs or their alignment.[8,9]

Here we give a direct experimental proof of the ground-state bonding or antibonding nature in coupled PCCs. The challenge is that the parity of the coupled modes refers to a phase property which is quite difficult to probe. Phase-sensitive techniques have been developed both in the near field[10] and in the far field,[11] by interferometric methods and resonant elastic scattering. Recently we have demonstrated that a reconsidered photonic Young's-like experiment, based on photoluminescence (PL), where the slits are replaced by two identical photonic nanocavities with embedded QDs, can directly probe the photonic mode symmetry by simply using far-field (FF) photoluminescence analysis.[12] In this paper we use the reconsidered Young's-type experiment to demonstrate that the ground state in photonic crystals may actually change from bonding to antibonding character depending on the spatial alignment of the two isolated cavities, as theoretically anticipated by numerical predictions.[9] However, the physical interpretation of this striking effect has apparently been quite overlooked in the literature, although evidence for the antisymmetric nature of the ground state in coupled PCCs has been recently reported.[13] The effect is interpreted in terms of the photonic energy functional.

The investigated sample consists of a 320-nm-thick GaAs membrane with three layers of high-density InAs QDs emitting at 1300 nm grown by molecular-beam epitaxy at the center of the membrane. The photonic structure is a two-dimensional triangular lattice where the single cavity, denominated $D2$, is formed by four missing holes.[14] The photonic molecules are designed in two different configurations, depending if the coupling line lies along the principal M or K axis of the photonic crystal. Henceforth we will also refer to vertically (horizontally) aligned $D2$ cavities if the coupling line lies along the principal M (K) axis of the photonic crystal [see the scanning electron microscope (SEM) images in Figs. 1 and 2]. The photonic molecules were characterized in a microphotoluminescence setup using a microscope objective of numerical aperture of 0.7. The external cone of view is then 45° with respect to the normal to the sample surface and the experimental angular resolution is of the order of 8°.[15] For excitation we used a solid-state laser emitting at 532 nm. PL emission from the sample was collected with a fiber, dispersed by a spectrometer and detected by a cooled InGaAs array; the spectral resolution is of the order of 0.1 nm. Finally, numerical simulations were carried out with

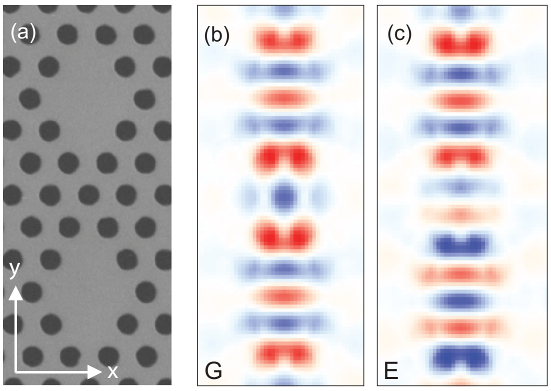

FIG. 1. (Color online) (a) SEM image of the M-coupled PCC; (b) FDTD near-field map of the ground-state E_x component; (c) FDTD near-field map of the first-excited-state E_x component.

a finite-difference time-domain (FDTD) solver package, and performing guided-mode-expansion (GME) calculations.

Figures 1(a) and 2(a) show the SEM images of the M- and K-aligned photonic molecules, respectively. The coupling strength between the fundamental modes of the two $D2$ cavities depends on the alignment configuration, mainly due to the spatial mode elongation. This is shown in Figs. 1(b), 1(c), 2(b), and 2(c) where the x component of the electric field calculated by the FDTD method is reported for the ground (G) and first excited (E) states of the M and K photonic molecules, respectively; the color scale indicates the positive (red) and negative (blue) amplitude. Both G and E modes arise from the coupling of the ground modes of the two $D2$ cavities, which turns out to be even along both the M and K axes. A large mode overlap is found for the M coupling, while very little overlap is observed in the case of K alignment.[5] As a matter of fact, we find from numerical calculations a frequency splitting between the G and E modes of the photonic molecules of 16 and 0.5 nm (in wavelength units) for the M and K alignments, respectively. For the M molecules the parity of the modes has to be considered for spatial inversion with respect to the horizontal K axis of the photonic crystal and the G (E) mode is predicted to be even (odd). This corresponds to the intuitive expectation on the basis of the quantum mechanical counterpart, that the ground state has a bonding character. In the case of the K molecules the parity of the modes has to be considered for spatial inversion with respect to the vertical M axis of the photonic crystal and the G (E) mode is predicted to be odd (even). Therefore the ground state of the K molecules is predicted to have an antibonding character, which is in contrast with the intuitive expectation on the basis of the quantum mechanical analogy.

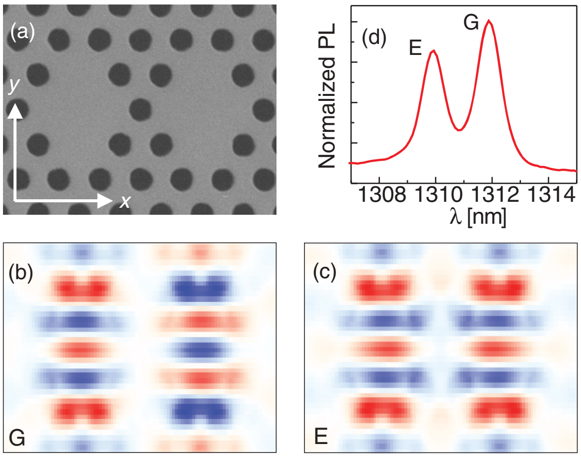

FIG. 2. (Color online) (a) SEM image of the K-coupled PCC; (b) FDTD near-field map of the ground-state E_x component; (c) FDTD near-field map of the first-excited-state E_x component; (d) PL spectrum showing an initial disorder-induced detuning of 2.1 nm.

Unavoidable dielectric disorder associated with the fabrication tolerance makes the two PCCs of the photonic molecules not identical. For example, the photonic pores are not all identical and this introduces a local dielectric perturbation that shifts the microcavity modes. For the uncoupled microcavities the disorder does very little to the mode distribution, since usually the shift is much smaller than the mode separation. Let us now consider the photonic molecule. The main role of disorder is to introduce a frequency detuning Δ between the two modes of the individual microcavities. The resulting molecular modes have a frequency splitting given by $\Omega = \sqrt{\Delta^2 + \Omega_0}$ where Ω_0 is the splitting for identical microcavities. Whenever Ω_0 is not much larger than Δ, the disorder strongly modifies the mode distribution of the molecular states. For $\Omega_0 \gg \Delta$ the disorder is negligible and we expect to have molecular modes delocalized over the whole photonic molecule, with a spatial distribution very similar to that in the ideal case of zero disorder. On the contrary, for $\Omega_0 \ll \Delta$ the modes are localized on each microcavity. While the detuning from resonance may be negligible for the M alignment where the mode splitting Ω_0 is as large as 16 nm, it turns out to be an important point for the K alignment where the mode splitting Ω_0 is expected to be only 0.5 nm. In order to experimentally prove the antibonding nature of the ground state in photonic crystals, the first step is therefore to tune the photonic molecules to perfect resonance $\Delta = 0$. We take advantage of the laser-assisted oxidation of the PCCs for compensating the photonic disorder,[16,17] which is schematically drawn in Fig. 3(a). We have shown that laser exposure with 1 mW at 514 nm on a spot of 1 μm allows a controlled shift of the frequency of the modes of PCCs, due to photoinduced oxidation of the GaAs membrane.[17] Since the oxidation produces a blueshift of the photonic modes, we need to expose the red-tuned nanocavity of the photonic molecules. Then spatially resolved PL is used to image the photonic mode of the cavity and monitor the spatial localization of the photonic modes. We exposed the red-shifted PCC of the K photonic molecules at steps of the order of half an hour with a power of 1 mW. The anticrossing curve of the G and E states of the molecule is shown in Fig. 3(b). The initial splitting [see Fig. 2(d)] was of the order of 2.1 nm and after 150 min we were able to obtain the minimum splitting (i.e., the anticrossing) of the order of 1.2 nm.

The FF intensity patterns directly give information on the mode parity. In the case under consideration, the even molecular mode arises from two in-phase single-cavity modes. Following the Young's analogy, we expect to observe constructive interference along the axis perpendicular to the alignment direction.[12] On the contrary, the odd mode arises from two out-of-phase single-cavity modes, and we expect to find destructive

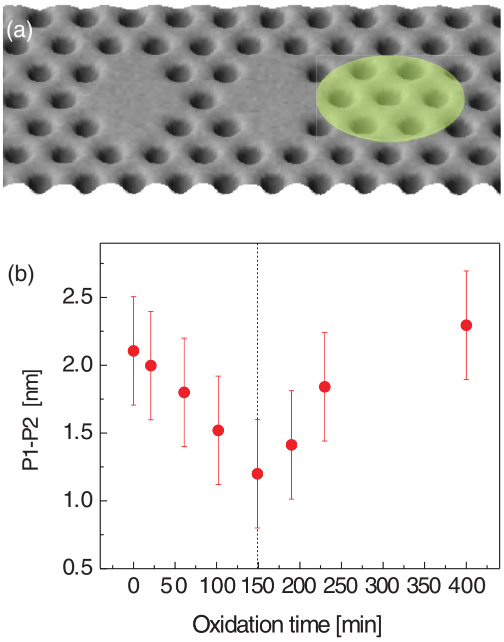

FIG. 3. (Color online) (a) Laser exposure (green circle) on the SEM image of the K-coupled PCC for controlling the mode detuning via light-induced oxidation; (b) anticrossing curve of the G and E states of the molecule.

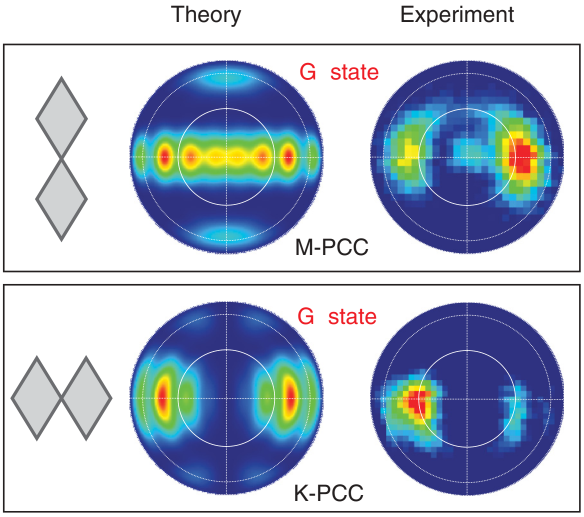

FIG. 4. (Color online) Comparison between the FDTD calculations (left) and experimental data (right) for the G state of both M- and K-coupled PCCs. The white circles indicate the aperture angles of 30° and 60°, respectively.

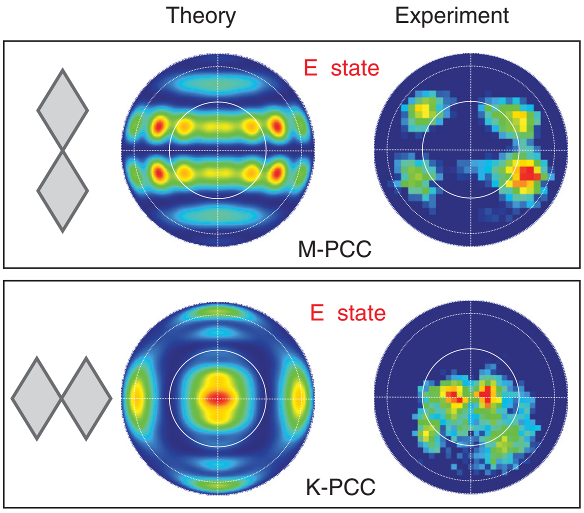

FIG. 5. (Color online) Comparison between the FDTD calculations (left) and experimental data (right) for the E state of both M- and K-coupled PCCs. The white circles indicate the aperture angles of 30° and 60°, respectively.

interference along the axis perpendicular to the alignment direction. It is also worth noticing that the FF pattern of the G mode of the single $D2$ cavity is essentially a bright horizontal band.[12] With these data in mind, the Young's analogy explains the differences between the two modes: while a bright horizontal line is the signature for evenness of the G mode in the vertical coupling, the presence of a large vertical dark fringe is the fingerprint of oddness for the G mode in the horizontal coupling. The experimental and calculated results for the G state for both photonic molecules are reported in Fig. 4. The calculation clearly explains the differences between the two modes. The bright horizontal line is the signature for evenness of the G mode in the M coupling, while the presence of a large horizontal dark fringe is the fingerprint of oddness for the G mode in the K coupling. The experimental data nicely agree with the theoretical predictions. Therefore the G state is bonding for the M coupling and antibonding for the K coupling.

In order to confirm these findings, let us demonstrate that in both coupling configurations the E modes have opposite parity with respect to the relative G modes. The experimental and calculated results for the E state for both photonic molecules are reported in Fig. 5. Again, the calculation clearly explains the differences between the two modes. The oddness of the E mode for the M coupling is demonstrated by the presence of a dark horizontal fringe in the FF pattern. Similarly, the bright central spot in the FF pattern in the case of the E mode in the K coupling arises from the product of the horizontal band of the single $D2$ cavity by the vertical constructive bright fringe due to the evenness of the E mode in the K coupling. Again, the experimental data nicely agree with the theoretical predictions.

The counterintuitive change of the photonic molecule ground state from a bonding to an antibonding orbital is also confirmed by performing GME calculations reported in Fig. 6, where we analyze the mode symmetry of the

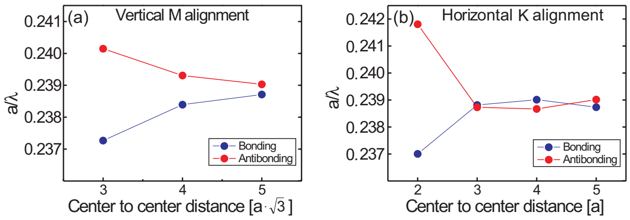

FIG. 6. (Color online) Calculated energies with the GME method for the vertically (a) and horizontally (b) coupled $D2$ cavities as a function of center-to-center separation (in units of the hole-to-hole distance).

photonic molecules as a function of the cavity-to-cavity distance. For these calculations, we used supercell dimensions of up to $16a \times 7w_0$ for K alignment (where a is the period of the photonic lattice and $w_0 = a\sqrt{3}$), and $10a \times 13w_0$ for the case of M alignment. The experimental data discussed in Figs. 4 and 5 are related to the results corresponding to a center-to-center distance equal to $3\sqrt{3}\,a$ and $3a$. Figure 6(a) shows that the ground molecular state for the M alignment, as a function of the center-to-center distance, is always bonding and the bonding-antibonding energy splitting decreases monotonically as expected in standard coupled systems. On the contrary, in the case of horizontal coupling, Fig. 6(b), the ground-state symmetry does not follow a trivial behavior. The fundamental mode can have either an even or an odd character as a function of the cavity separation.

An antibonding ground state has been recently observed in coupled InAs QDs.[7] The heavy-hole—light-hole mixing and the spin-orbit interaction causes this counterintuitive reversal for particular values of the barrier thickness. In photonics the origin for the antibonding ground state is quite different and relies on the basic difference between the evanescent waves in atoms and photonic crystals.[8] In order to understand the physics involved, let us recall that the eigenstates of an arbitrary photonic system satisfy the variational principle related to the minimization of the energy functional U:[1]

$$U = \frac{\int |\vec{\nabla} \times \vec{E}(\vec{r})|^2 dV}{\int \varepsilon(\vec{r})|\vec{E}(\vec{r})|^2 dV}. \qquad (1)$$

The ways to minimize U are two: (i) by reducing the number of mode nodes and (ii) by increasing the field concentration in the high-dielectric regions. As in quantum mechanics, bonding states always satisfy (i) and this is the reason why the ground states usually have bonding character. However, while the wave function in atoms decays exponentially with distance from the nucleus, the localized electric field in a photonic-crystal microcavity shows an exponentially damped oscillation with distance from the dielectric defect. The interference between the oscillating evanescent electric field of the two PCC modes in the region of the mode overlap produces constructive or destructive interference. In the case of constructive (destructive) interference, the electric field is enhanced (reduced), and this creates a reduction (enhancement) of the energy functional. Therefore the presence of antibonding ground states in photonics has to be associated with constructive interference in the defect region for the odd state, which may enhance the electric field concentration in the dielectric regions.

In conclusion, we have experimentally demonstrated that photonic molecules support both bonding and antibonding ground states, depending on the spatial arrangement of the two individual dielectric defects. The oscillating nature of the evanescent waves in the photonic band gaps allows the design of photonic molecules with ground states of different parities. Following our results, it could be possible to change the bonding into antibonding character by a dielectric perturbation of the PC which, controlling the tunneling of the modes, may tailor the evanescent waves. In addition, there has been a growing interest in quantum simulators based on coupled cavity arrays, recently. Theoretical models often assume a tight-binding formulation, with an effective intercavity coupling parameter J that is assumed, in analogy to electronic systems, to be positive as in the case of bonding ground states. With our results, we have shown that J, in these effective tight-binding models, can also be negative (as in the case of an antibonding ground state), depending on the structural geometry of the underlying photonic structures, with interesting conclusions for the development of novel theoretical models. We believe that these peculiar properties will open the road to possible progress in quantum information processing.

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