Tunable polaritonic molecules in an open microcavity system
S. Dufferwiel, Feng Li, A. A. P. Trichet, L. Giriunas, P. M. Walker, I. Farrer, D. A. Ritchie, J. M. Smith, M. S. Skolnick, and D. N. Krizhanovskii

Citation: Applied Physics Letters 107, 201106 (2015); doi: 10.1063/1.4936092
View online: http://dx.doi.org/10.1063/1.4936092
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/107/20?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Strong exciton-photon coupling in open semiconductor microcavities

Effect of magnetic field on polariton emission characteristics of a quantum-well microcavity diode

Estimating the conditions for polariton condensation in organic thin-film microcavities

Boundary effects on the dynamics of exciton polaritons in semiconductor microcavities

Polariton quantum boxes in semiconductor microcavities
Tunable polaritonic molecules in an open microcavity system

S. Duffnerwiel, F. Li, A. A. P. Trichet, L. Girinias, P. M. Walker, I. Farrer, D. A. Ritchie, J. M. Smith, M. S. Skolnick, and D. N. Krizhanovskii

Department of Physics and Astronomy, University of Sheffield, Sheffield S3 7RH, United Kingdom
Department of Materials, University of Oxford, Oxford OX1 3PH, United Kingdom
Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom

(Received 24 August 2015; accepted 6 November 2015; published online 17 November 2015)

We experimentally demonstrate tunable coupled cavities based upon open access zero-dimensional hemispherical microcavities. The modes of the photonic molecules are strongly coupled with quantum well excitons forming a system of tunable polaritonic molecules. The cavity-cavity coupling strength, which is determined by the degree of modal overlap, is controlled through the fabricated centre-to-centre distance and tuned in-situ through manipulation of both the exciton-photon and cavity-cavity detunings by using nanopositioners to vary the mirror separation and angle between them. We demonstrate micron sized confinement combined with high photonic Q-factors of 31 000 and lower polariton linewidths of 150 μeV at resonance along with cavity-cavity coupling strengths between 2.5 meV and 60 μeV for the ground cavity state. © 2015 AIP Publishing LLC.

Photonic molecules are two or more coupled electromagnetically interacting microcavities. In the most simple case, two zero-dimensional microcavities are brought together so that their photonic modes interact, leading to hybridisation and the formation of bonding (B) and antibonding (AB) modes with an energy splitting given by the hybridisation and the formation of bonding (B) and antibonding (AB) modes with an energy splitting given by the coupling strength, J. The initial demonstrations of lithographically fabricated photonic molecules coupled two adjacent cavities leading to a direct analogy with a simple diatomic molecule. Alternative configurations of photonic molecules exist such as square-shaped photonic dots coupled via a semiconductor bridge, coupled defects in photonic crystal membranes, whispering-gallery microdisks coupled via a small air gap, and planar Fabry-Perot cavities coupled through a partially transparent distributed Bragg reflector (DBR). More complicated structures such as photonic graphene have been developed where a honeycomb lattice of coupled micropillars displays polariton dispersions which are analogous to the electronic bandstructure of its chemical counterpart. Coupled micropillars have found applications in the development of ultrabright entangled photon sources along with Josephson oscillations between two linked polariton condensates. Recent theoretical proposals have shown that a weak nonlinearity in a coupled cavity system can lead to a strongly antibunched single photon source in a process termed unconventional polariton blockade. A coupled cavity system based upon coupled micropillars containing quantum wells (QWs) is likely to be unsuitable to observe this effect since the etching required to achieve micron sized lateral confinement, and hence enhance the nonlinear polariton-polariton interaction to practical values, leads to significant degradation of the polariton linewidth. Consequently, to experimentally demonstrate this blockade effect in III-V QW systems an alternative coupled cavity system which can concurrently achieve narrow polariton linewidths, strong lateral confinement, and controllable cavity tunneling times is required.

In this study, we demonstrate a suitable coupled cavity system based upon spectrally tunable open access polaritonic molecules. The system is formed through reducing the centre-to-centre distance between adjacent zero-dimensional cavities during mirror fabrication so that hybridisation of the modal structure occurs. The cavity resonances of the coupled cavity system are strongly coupled to QW excitons, present in a cavity region above the bottom semiconductor DBR, giving rise to hybrid light-matter eigenstates and the formation of a polaritonic molecule. In-situ manipulation of both the exciton-photon detuning and cavity-cavity detuning using nanopositioners allows full control over the cavity-cavity coupling strength. In addition, the small concave mirror radius of curvature (RoC) of 6 μm gives rise to micron sized polariton confinement in the hybridised modes, which combined with the narrow polariton linewidths at resonance of 150 μeV, may allow the blockade regime for coupled cavities to be reached.

The shape of the coupled cavities is given by the surface formed from the isophase profile of two Gaussian modes calculated for a cavity length of 3 μm and RoC of 6 μm, separated by a distance d. A 13 × 13 array of coupled cavity features was fabricated using focused ion beam (FIB) milling, where d was incrementally reduced from 10 μm to 7.5 μm as shown in the microscope image in Fig. 1(a). Note that open cavities can also be realised using laser machining. The coupled cavity array was then coated with 11 pairs of SiO2/TiO2 quarter-wave stacks. The individual hemispherical cavities of the molecules laterally confine the photonic field with a confining potential that is nearly harmonic giving rise to zero-dimensional cavity eigenstates which can be described by Hermite or Laguerre Gaussian profiles depending on the exact symmetry of the confining potential. The RoC of the single cavity concave feature of 6 μm gives an expected Gaussian beam waist of the fundamental mode at the QW position of around 0.89 μm. In the

http://dx.doi.org/10.1063/1.4936092
the polaritonic molecule has a centre-to-centre distance of 8.08 μm and a linear polariser was used to select modes polarised along the long axis of the molecule. The four labelled spectral resonances correspond to the symmetric/antisymmetric modes which arise from coupling of the individual cavity ground states (s-B/s-AB) and the first transverse modes (p-B/p-AB) as discussed below. A distinct anti-crossing is observed between all eigenmodes and the QW exciton with clear lower and upper polariton branches with a Rabi splitting of 3.4 meV. The B/AB modes of the polaritonic molecule system can be described by a 4-level coupled-oscillator model with the Hamiltonian

\[
H = \begin{pmatrix}
E_X & \Omega_{\text{Rabi}}/2 & 0 & 0 \\
\Omega_{\text{Rabi}}/2 & E_{c1} & J/2 & 0 \\
0 & J/2 & E_{c2} & \Omega_{\text{Rabi}}/2 \\
0 & 0 & \Omega_{\text{Rabi}}/2 & E_X 
\end{pmatrix}
\]

(1)

in which \(E_X, E_{c1},\) and \(E_{c2}\) are the energies of the bare exciton and cavity modes, \(\Omega_{\text{Rabi}}\) is the exciton-photon coupling, and \(J\) is the cavity-cavity coupling strength. The eigenenergies that describe the upper (UP) and lower (LP) polariton states are given by diagonalising the Hamiltonian of Eq. (1)

\[
E_{\text{UP/LO}} = \frac{2E_X + (\Delta \pm J/2)^2}{2} \pm \frac{1}{2} \sqrt{(\Delta \pm J/2)^2 + \Omega_{\text{Rabi}}^2},
\]

(2)

where \(\Delta\) is the exciton-photon detuning given by \(E_c - E_X\), where we have assumed the cavities are in resonance \((E_c = E_{c1} = E_{c2})\). The calculated eigenenergies as a function of detuning, plotted in Fig. 1(d), exhibit a reduction in the coupling strength (B-AB splitting) as the detuning is tuned to zero due to the coexistence of light-matter and light-light couplings. We observe this reduction experimentally in Fig. 1(c), with a B-AB splitting of ~1 meV at large negative detunings, which gradually diminishes when approaching the exciton energy. Since the B-AB splitting between polaritons is contributed by their photonic component increasing the excitonic component leads to reduced photonic coupling and a smaller B-AB splitting.

Real space images of the s-B and s-AB modes shown in Fig. 2(a) clearly exhibit the symmetric and antisymmetric nature of the mode profiles. Note that the parameter \(d\) only determines the nominal separation between the overlapping isophase Gaussian profiles of the constituent cavities which define the fabricated coupled cavity shape. The actual mode position is determined by the photonic potential induced from this shape and leads to a modal structure of the coupled cavities with lateral sizes much smaller than the nominal separation.13 For example, the s-AB mode in Fig. 2(a) has a nominal separation of 8.0 μm, whereas in the experiment the measured separation between mode maxima is 2.96 μm with full width half maxima of 1.3 μm. The lateral extent of the higher order modes is typically larger for increasing transverse numbers leading to stronger modal overlap between these modes in coupled cavities. This leads to much higher B-AB splittings in comparison to the ground state coupled s-type modes in the same photonic molecule. As an example, these B-AB modes with higher transverse orders are shown in Figs. 2(b)–2(d), respectively.
Fig. 2. Tomographic images of the first four pairs of bonding/antibonding states. (a) S-AB/s-B modes ground state mode coupling (b) p-AB/p-B modes due to first transverse mode coupling. (c) AB/B modes due second transverse mode coupling. (d) AB/B modes due to third transverse mode coupling. The scale bar is 4 μm.

A typical PL spectrum at large negative detuning is shown in Fig. 3(a). Due to the elongated geometry (compared to circular-shaped cavities) of the photonic molecule and the TE-TM splitting induced by the DBR surfaces, we observe two orthogonally linearly polarised sets of s-B and s-AB modes. Fig. 3(b) shows the s-B state at a negative detuning of −9.6 meV. A Lorentzian fit yields a linewidth of 47 μeV corresponding to a photonic Q-factor of around 31 500. This is most likely limited by the stability of the system to low frequency acoustic vibrations. Fig. 3(c) shows the lower polariton s-B and s-AB states at zero exciton-photon detuning. Fitting the lower polariton s-B state reveals a linewidth of 150 μeV which is limited by the inhomogeneously broadened exciton linewidth of 650 μeV. Narrower polariton resonances can be expected for a semiconductor half cavity containing a narrower linewidth QW.

The coupling strength of the coupled longitudinal modes is plotted against nominal centre-to-centre distance in Fig. 3(d). The transition between uncoupled and coupled cavities occurs abruptly when the centre-to-centre distance is smaller than around 8.1 μm. At a centre-to-centre distance of 8.26 μm, we measure a coupling strength of 63 μeV which increases continuously to 2.45 meV at d = 7.90 μm. Hence the precise control of the coupled cavity separation provided by the FIB milling fabrication process allows the selection of any arbitrary coupling strength between these ranges.

The ability to manipulate the relative angle between top and bottom mirrors in-situ using goniometer nanopositioners provides a further degree of freedom to tune the B-AB splitting. This provides high flexibility in choosing the cavity-cavity detuning δ = E1 − E2 constituting a distinct advantage of open cavities. Furthermore, this allows for the correction of any fabrication imperfections which can cause slight detunings between adjacent cavities even when the planar areas of the mirrors are parallel. The detuning dependent B-AB splitting for coupled cavities is characterized by

$$\Delta E = E_{AB} - E_B = \sqrt{\delta^2 + J^2}. \quad (3)$$

In the experiment, tilting the mirrors tunes δ, allowing to “decouple” the photonic molecule by reaching a |δ| large enough to significantly reduce the tunnelling probability. Fig. 4(a) displays the spatial profiles of the modes corresponding to the configuration in the panels above. At zero-angle where δ = 0 (middle panel), the polaritonic molecule exhibits s-B and s-AB modes each of which fills both cavities symmetrically, whereas at large negative (left panel)
and positive (right panel) angles, the cavities begin to “decouple” and the modes are more localized in each individual cavity. The B-AB mode splittings are plotted as a function of the relative angle for two exciton-photon detunings corresponding to photonic ($\Delta = -14.0 \text{ meV}$) and excitonic cases ($\Delta = +0.75 \text{ meV}$) in Fig. 4(b). For both cases, there is clear anti-crossing behaviour due to the strong cavity-cavity coupling, but the B-AB splitting is greater for the photonic-like case at all angles due to the smaller exciton fraction. The reduction from $1.07 \text{ meV}$ at $\Delta = 0$ to $0.37 \text{ meV}$ at $\Delta = 0.75 \text{ meV}$ is consistent with a reduction in the photonic component from 99.6% to 27.6% and is complementary to the tuning by the $z$-nanopositioner in Fig. 1(c).

In summary, we present polaritonic molecules with full tunability of the cavity-cavity coupling strength through controlled fabrication of the centre-to-centre distance combined with in-situ tunability of the exciton-photon and cavity-cavity detunings. This tunability is likely to find a number of applications. First, it may allow the coupling of both the exciton and biexciton line of a pre-selected single QD to the cavity detunings. This tunability is likely to find a number of applications. First, it may allow the coupling of both the exciton and biexciton line of a pre-selected single QD to the optical modes of the photonic molecule in an efficient manner leading to bright source of entangled photon pairs. Second, the system exhibits excellent potential to observe polariton blockade in coupled cavities where we demonstrate a number of prerequisite conditions such as experimentally observable cavity-cavity tunnelling times (>2 ps), micron sized confinement, and narrow polariton linewidths. For optimum antibunched emission, we estimate a required non-linearity of $U \sim 5 \mu \text{eV}$ using the experimentally measured parameters of $\gamma_{LP} = 150 \mu \text{eV}$ for the ground state bonding mode and $J = 0.5 \text{ meV}$. In this system, the nonlinearity is manifested as the repulsive polariton-polariton interaction which is inherited from the excitonic component. The required value is comparable to the nonlinear polariton-polariton interaction measured in 10 μm micropillars, which is expected to be larger in open cavity systems due to the increased lateral confinement.

This work has been supported by the EPSRC Programme Grant No. EP/J007544, ERC Advanced Investigator Grant EXCIPOl and by the Leverhulme Trust.