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Georgiou, K., McGhee, K.E., Jayaprakash, R. orcid.org/0000-0002-2021-1601 et al. (1 more author) (2021) Observation of photon-mode decoupling in a strongly coupled multimode microcavity. The Journal of Chemical Physics, 154 (12). 124309. ISSN 0021-9606

https://doi.org/10.1063/5.0038086

Supplementary Material

Observation of Photon-Mode Decoupling in a Strongly Coupled Multimode Microcavity

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1. Rabi-splitting energy Vs. TDBC concentration

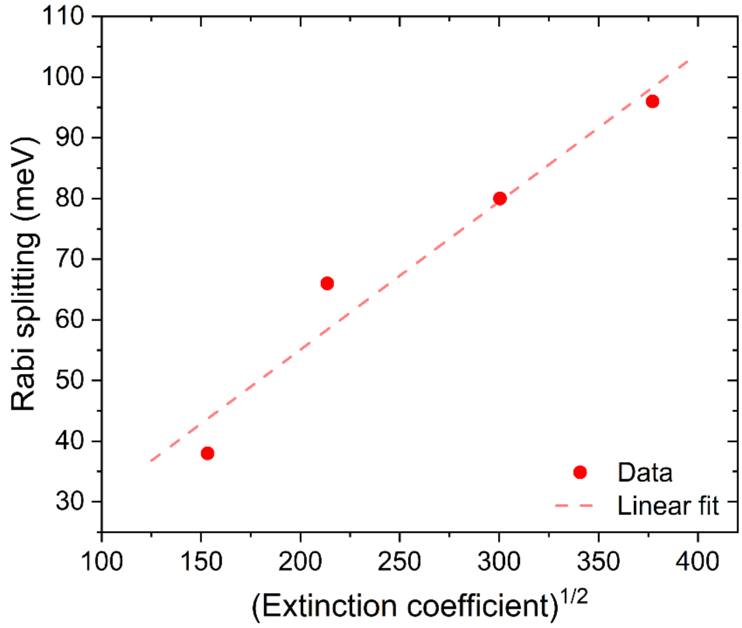

Figure S1. Rabi splitting energy against square root of extinction coefficient for the four cavities studied (red circles). The dashed line is a linear fit to the data.

2. Hopfield coefficients of coupled and decoupled microcavities

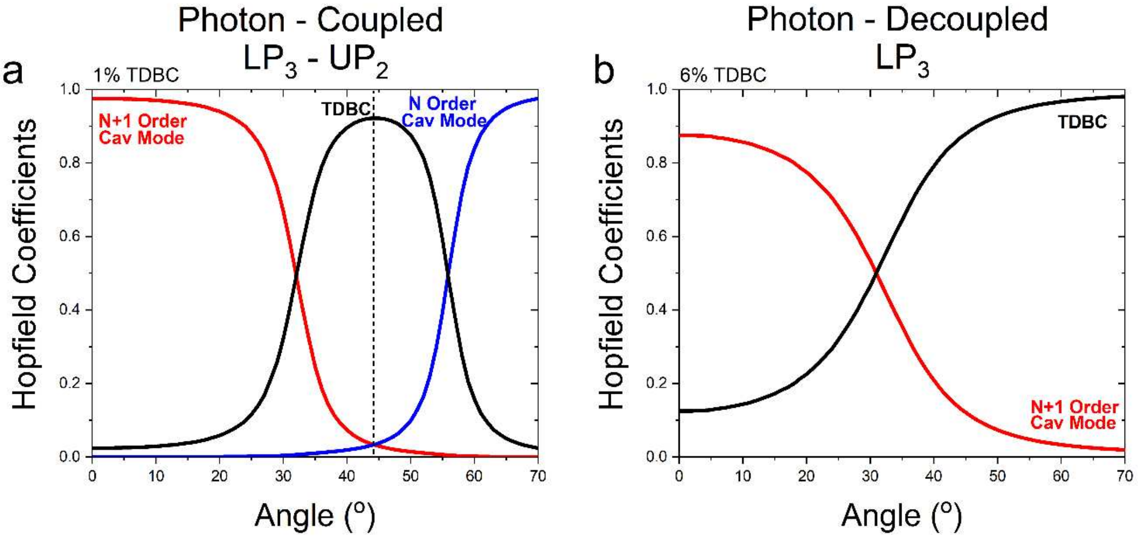

Figure S2. Hopfield coefficients extracted from the coupled oscillator models for (a) a photon-coupled and (b) a photon-decoupled microcavity. The vertical black dashed line in (a) at angle of around 45° indicates the point at which the polariton mode crosses the exciton energy; a point where the mixing between the two photon modes is maximized.

3. Comparison of photon-coupled and photon-decoupled models

Figure S3 plots experimental reflectivity data from two cavities containing 4% and 6% TDBC in gelatine by weight. Data in Figure S3 (a) and (c) is fitted with a (2*N)*×(2*N)* photon decoupled Hamiltonian model while Figure S3 (b) and (d) plots data when fitted with a conventional (*N*+1)×(*N*+1) photon-coupled Hamiltonian model. As it can be seen, the photon-decoupled model fits the data relatively well as compared to the photon-coupled model, particular in the spectral region of the exciton. Therefore, as it is evident from the experimental data and the simulations the cavities have transitioned and operated in the photon-decoupled regime for both TDBC concentrations.

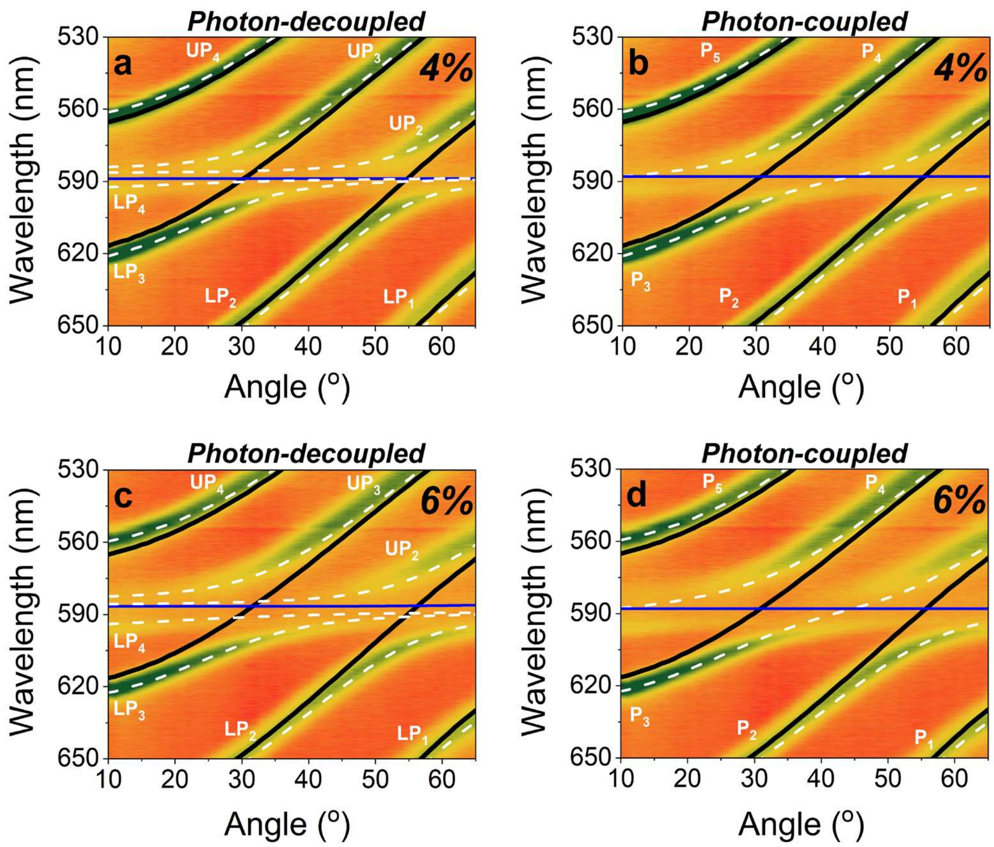

Figure S3. (a) and (b) Experimental angle-resolved white light reflectivity maps of a cavity containing 4% TDBC by weight in a gelatine matrix when fitted with a (a) 2*N*×2*N* photon-decoupled and (b) (*N*+1)×(*N*+1) photon-coupled Hamiltonian model. (c) and (d) Experimental angle-resolved white light reflectivity maps of a cavity containing 6% TDBC by weight in a gelatine matrix when fitted with a (c) (2*N*)×(2*N*) photon-decoupled and (d) (*N*+1)×(*N*+1) photon-coupled Hamiltonian model. The blue solid line corresponds to the peak wavelength of the TDBC absorption, the black solid lines show the uncoupled cavity modes extracted by a TM model and the white dashed lines represent the polariton modes as predicted by the Hamiltonian models.

4. Refractive index and optical constants of electric field TM model

Figure S4 plots the refractive index as a function of wavelength and distance from the substrate for two cavities containing 1% and 6% TDBC by weight in a gelatine matrix. Note that by altering the concentration of TDBC we cause a change of the refractive index in the active layer. Therefore, here we omit refractive index data for cavities containing 2% and 4% TDBC in gelatine by weight as this is directly related to the extinction coefficient of the active layer. The absorption spectra of the TDBC films have been simulated inputted into the TM model using a series of Lorentz oscillators. In addition, Table S1 summarizes thickness and refractive index data used in the TM modelling for all four microcavities discussed here.

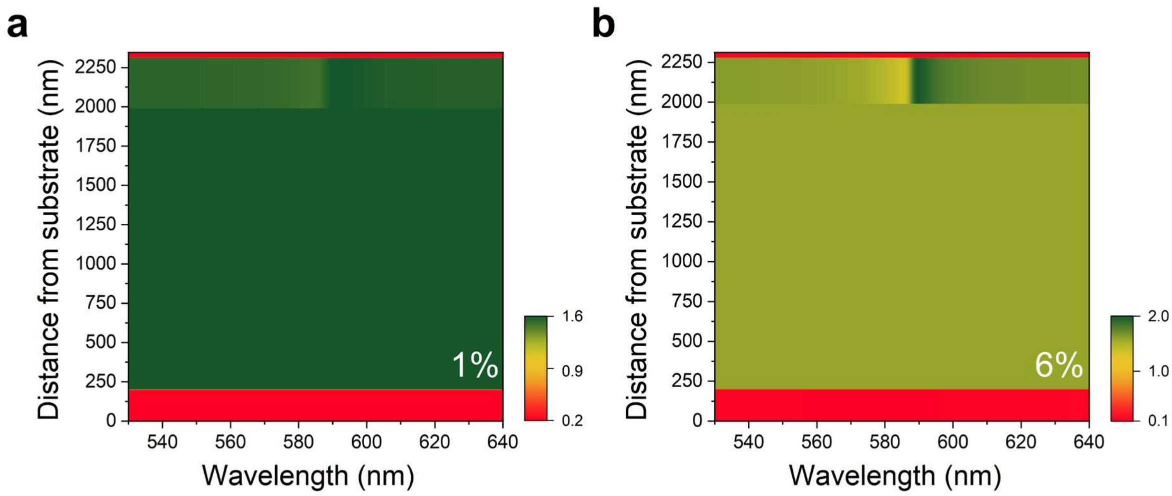

Figure S4. Refractive index in each layer of a microcavity as a function of wavelength and distance from the substrate for (a) 1% and (b) 6% TDBC in gelatine by weight.

Layers	Concentration							
	2%		3%		4%		6%	
	$d\ (nm)$	$n(\lambda)$	$d\ (nm)$	$n(\lambda)$	$d\ (nm)$	$n(\lambda)$	$d\ (nm)$	$n(\lambda)$
Top silver	34	n_{Ag}[1]	34	n_{Ag}[1]	34	n_{Ag}[1]	34	n_{Ag}[1]
TDBC layer	322	1.55	318	1.55	313	1.55	288	1.66
PS layer	1790	1.59	1790	1.59	1790	1.59	1790	1.59
Bottom silver	200	n_{Ag}[1]	200	n_{Ag}[1]	200	n_{Ag}[1]	200	n_{Ag}[1]
Glass substrate	1.1×10^6	1.5	1.1×10^6	1.5	1.1×10^6	1.5	1.1×10^6	1.5

Table S1. Thicknesses (*d*) and refractive indices (*n*) used in the TM models used to calculate the electric field distribution shown in Figure 4 of the manuscript.

5. References

[1] P. B. Johnson and R. W. Christy, Phys. Rev. B **6**, 4370 (1972).