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COUPLED MULTIPLE ORGANIC MICROCAVITIES

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ABSTRACT. Recently, organic based microcavities became a focal point in the boundary photonic research due to the special features of the organic active molecules whose properties may be easily modulated. By using the thermal evaporation we realized double and triple coupled-microcavity structures containing one or more active layers of light emitting organic molecules. The resonators are tuned at the same wavelength and coupled by low reflectivity mirrors. In particular we grew an asymmetric double system with only one active organic layer embedded in one microcavity. The reflectivity measurements show that the coupling between the two resonator results in a splitting of the optical modes. The photoluminescence spectra display a double wavelength emission. The comparison of the diagonalized effective Hamiltonian with the observed resonances further confirms the strong coupling between the two cavities. A quantum statistical approach for interacting quantum systems in the strong coupling regime reproduces with very good agreement the experimental results. The triple structure has been designed to improve the coupling among the resonators. The device shows three emission peaks at different wavelength and represents a very interesting start up to realize white organic based LEDs and ultrafast optical amplifiers.

1. Introduction

The simultaneous confinement of electronic states and optical fields in microcavity [1] (MC) structures has opened up a novel and very promising methods to study and control the light-matter interaction [2, 3] in advanced optical devices [4, 5]. The quantized photon modes of a MC and the exciton states of a quantum well (QW) embedded in the MC are interesting physical examples of coupled oscillators systems. Strong coupling between the two oscillators arises in the resonance regime leading to the formation of new exciton-photon coupled modes, termed cavity polaritons [6]. A photonic analogue of cavity polaritons resulting from QW excitons strongly coupled to cavity photons is represented by the photon modes in multiple coupled MCs. During the last decade the systems of multiple coupled MCs have been investigated as a way to further increase the flexibility in controlling both radiation and material degrees of freedom [7, 8, 9, 10, 11]. In a single cavity, the optical field strength in the cavity is enhanced relative to that of the external photon field. In two coupled cavity the same phenomenon arises, but in addition there is coupling between the photon fields of the two cavities, that is controlled by the transmission of the

central mirror. The mode interaction may leads to strong mode pushing; when the Fabry-Pérot modes of the individual MCs are resonant, the observed modes of the coupled system are shifted to either side of the resonance wavelength. The amount of pushing, or splitting, depends on the degree of coupling between the two resonators. The reflection spectrum of such coupled MCs has two dips in the photonic band gap, which correspond to resonances of incident light with the modes of coupled MCs [12]. Optical multiple MCs have been proved to be a very successful system to study the light-matter interaction [8, 9, 10] and to observe some phenomena such as polarization splitting [7] and parametric oscillation [13, 14]. In addition, these structures have several characteristics that make them excellent candidate for the design of new optoelectronic and photonic devices [15]. A theoretical analysis of coupled non-linear Fabry-Pérot cavities showed that bistable and multistable behavior may occur, therefore these systems can be used to realize devices including laser oscillators, a range of optical and electro-optic switches and optical parametric oscillations [13, 14, 16, 17, 18]. To obtain and enhance non-linear optical processes, monolithic semiconductor MCs with non-linear material clad between two highly reflecting Bragg mirrors have been proposed [19]. In this framework, organic materials were used as active medium in optical MCs to realize many different non linear optical devices (e.g., waveguide second harmonic generation, electro-optic modulators, directional Couplers). In particular, porphyrins [20] are a well known class of materials which unique characteristics make them very promising materials for optical application, which include optical limiting, optical switching, optical data processing and optoelectronic device fabrication [21, 22]. A vertical MC structure consisting of two mirror, separated by a cavity containing an active inorganic or organic medium such as (In,Ga)As QWs or a organic molecules thin film [23]. In this work, we report on the study, modeling and realization of multiple organic MCs. The structures were entirely grown by thermal evaporation in ultra high vacuum (UHV). The samples are characterized by reflectivity and photoluminescence measurements both at normal incidence. Their behavior have been simulated by coupled oscillator model as proposed in Ref. [27].

2. Microcavities Design and Experimental Techniques

The samples were evaporated in UHV chamber, working at pressure below 4×10^{-6} Torr and equipped with three crucible and a quartz based thickness monitor (Sycon-STM100). The films were deposited onto Corning 7059 glass substrates. The organic thin film deposition required a homemade macor based crucible and the organic layer was evaporated at a rate of 0.5 Å$/s$. The structures are composed by $\lambda/2$ thick Lithium Fluoride (LiF) cavities coupled via a central Distributed Bragg Reflector (DBR). In the case of organic MCs also a thin film (10 nm) of tetrakis (4-methoxyphenyl) porphyrin (TMPP) is embedded [24, 25] in the center of the structure. The ensemble is enclosed between a bottom and a top Bragg mirrors. The DBRs are made of quarter wavelength layers of LiF ($n_1 = 1.392$) and Zinc Sulfide ($n_2 = 2.352$). The double MC [26] shown in Fig.1(left), consists of a bottom DBR of 8 periods, an active regions called "bottom cavity" containing the organic material, a middle DBR mirror of 3.5 periods, a second cavity completely dielectric ("top cavity"), and finally a top DBR realized with only 4 periods. Triple MC (See Fig.1 (left)) consist of 3 organic cavities enclosed between a bottom and a top DBR of 6 and 3.5

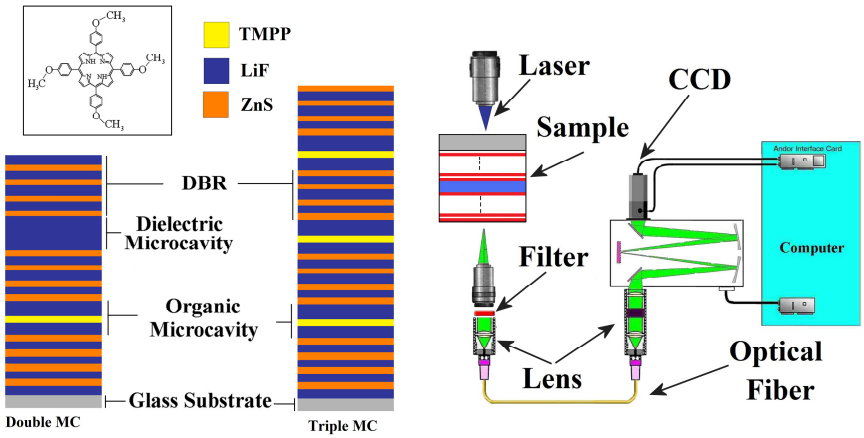

FIGURE 1. (Left) Double MC and triple MC structure. In the top is shown the organic molecule structure. (Right) PL setup

periods respectively. The three cavities have a $\lambda = 330$ nm thickness of LiF with a 10 nm thin TMPP film at their centre. The MCs are coupled by two DBR each consisting of 3.5 periods. The samples are been characterized by photoluminescence measurements and optical reflectivity taken stepwise while growing the structure. The reflectivity spectra was obtained with a spectrophotometer Perkin Elmer mod. Lambda 2. The PL setup (Fig. 1 (right)) consists of a $\lambda = 473$ nm laser focused by means of an objective on the bottom of the samples. The PL signal was collected by a lens that focused the light on the end face of a single-mode optical fiber. A long pass filter cutoff the laser signal. The spectrum emitted by the sample is analyzed using a monochromator coupled to a charge coupled device detector. In the case of double MC, the PL spectra was obtained in two different conditions: first by pumping the bottom of the sample and collecting the light emitted from the top side of the cavity; second pumping the top cavity and collecting the light emitted from the bottom side of the sample. All measurements were carried out at room temperature and in air atmosphere.

3. Double MC

The normal-incidence optical reflectivity (Fig. 2 (left)) shows the reflectivity of the bottom cavity containing the organic layer (dot line), in which a single resonance dip at about 1.962 eV is evident, and the reflectivity of the whole device (continuous line). The two coupled cavity modes can be easily identified as the dip at 1.898 and 1.980 eV. The PL spectra display two peaks at the same energy of the reflectivity dips despite only one cavity contains the active layer, thus confirming the effective coupling of the two cavities (Fig. 2 (right)). The coupling of the two cavities is also confirmed experimentally by the blueshift of the bottom MC resonance. Besides, the hypothesis of strong coupling regime in this structure is supported by theoretical analysis. The coupled energies are obtained diagonalizing the Hamiltonian [8]:

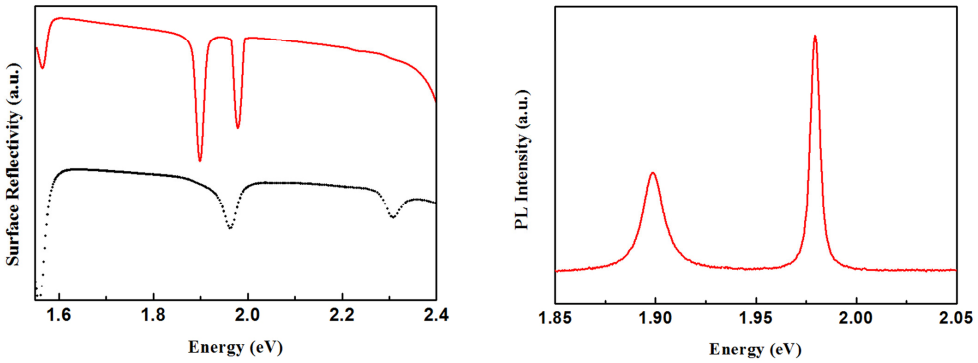

FIGURE 2. (Left) Experimental reflectivity spectra and (Right) experimental PL spectra of double MC.

$$\begin{pmatrix} E_1 & V_{opt} \\ V_{opt} & E_2 \end{pmatrix} \tag{1}$$

where $E_1 = 1.962$ eV is the energy of the uncoupled mode of the bottom cavity, E_2 is the unknown energy that the top cavity would exhibit in absence of coupling, and

$$V_{opt} = \eta_c \frac{\sqrt{1 - R_c}}{(2nL_{eff} \cos\theta_c)} \tag{2}$$

is the optical coupling between the cavities, determinate by the transmission $1 - R_c$ of the central mirror [8]. The values of E_2 and V_{opt} can be obtained from the experimental data setting the eigenvalues of the above matrix equal to the experimental resonance energies obtained from the reflectivity spectra. We obtain $E_2 = 1.916$ eV and $V_{opt} = 34$ meV and we note that the V_{opt} obtained is much larger than the half width at half maximum which is of about 9 meV, hence the system is in the very strong coupling regime. In this regime, each of the two reflectivity dips comes from both the cavity modes. The active structure can be considered as the photonic model of cavity polaritons resulting from QW excitons strongly coupled to cavity photons. PL from polaritons is obtained by pumping incoherently one subsystem (QW excitons) and detecting quanta from the other subsystem. Analogously, in the present device, we detect light from the passive top cavity while pumping the bottom cavity and exciting the incoherent PL from the active organic layer. A quantum statistical approach for interacting quantum system in the strong coupling regime reproduces with very good agreement the experimental PL spectra [27].

4. Theory

We study theoretically the PL properties of the coupled double MC by exploiting a quantum statistical approach for interacting quantum systems in the strong coupling regime

[27]. We start considering a system of two coupled MCs each of them can have an embedded layer of luminescent organig layer. Here we address the case where cavity photons and the organic excitations are in the weak coupling regime, corresponding to the present experimental realization.

The master equation for this system can be written as

$$\dot{\rho} = i[\rho, H_S] + \mathcal{L}_A^R + \mathcal{L}_B^R \,, \tag{3}$$

where the system Hamiltonian reads

$$H_S = \omega_a a^\dagger a + \omega_b \, b^\dagger b + g(a^\dagger \, b + a \, b^\dagger) \,, \tag{4}$$

with g being the coupling strength between the two single mode cavities (with annihilation operator a and b respectively) depending on the transmission of the central mirror. The superoperators $\mathcal{L}_A^R$ and $\mathcal{L}_B^R$ describe the interaction of the two cavity modes with reservoirs providing both damping and (when present) pumping mechanisms. Each of these terms consists of two contributions arising from two independent reservoirs: $\mathcal{L}_C^R = \mathcal{L}_C^{R1} + \mathcal{L}_C^{R2}$ ($C = A, B$). Transmission and diffraction losses of the cavity modes can be modeled, within a quasimode picture, as an effective coupling with a continuous ensemble of electromagnetic modes through the output mirrors [28]. When dealing with optical frequencies it can safely be regarded as a zero temperature reservoir. The resulting Liouvillian term can be written as

$$\mathcal{L}_C^{R1} = \frac{\gamma_c^{\mathrm{T}}}{2}(2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c) \,. \tag{5}$$

where γ_c^{T} describes the transmission and diffraction losses of the cavity mode. The incoherent optical pumping origins from light emission from the optically active transitions of the cavity embedded organic layer providing the characteristic emission band. Such levels get populated after laser excitation of the absorption band at higher energy. This mechanism can be described by an additional reservoir at a given effective temperature depending on the pumping rate. We address the weak excitation regime where saturation effects are not present. In this case it is possible to describe the optically active transitions as an ensamble of harmonic oscillators coupled to the resonant light mode of the surrounding cavity. The interaction Hamiltonian can be written as

$$H_I = \sum_m g_m c^\dagger \, d_m + H.c. \,, \tag{6}$$

where $c = a$ or b, and d_m is the annihilation operator for the m-th mode at energy ω_m. After the usual approximations leading to the master equation and neglecting the small Lamb and Stark shift terms [29, 30], the corresponding Liouvillian term takes the form,

$$\begin{aligned} \mathcal{L}_C^{R2} &= \frac{\gamma_c^{\mathrm{L}} + P_c}{2}(2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c) \\ &+ \frac{P_c}{2}(2c^\dagger \rho c - cc^\dagger \rho - \rho cc^\dagger) \,, \end{aligned} \tag{7}$$

where $P_c = \pi \sum_m g_m^2 \delta(\omega_c - \omega_m) \left\langle d_m^\dagger d_m \right\rangle$. The decay rate of the cavity mode is $\gamma_c^{\mathrm{L}} = \pi \sum_m g_m^2 \delta(\omega_c - \omega_m)$. The resulting total Lindblad-like Liouvillian term for the cavity

mode $\mathcal{L}_C^R = \mathcal{L}_C^{R1} + \mathcal{L}_C^{R2}$ can be written as,

$$\begin{aligned}\mathcal{L}_C^R &= \frac{\gamma_c + P_C}{2}(2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c) \\ &+ \frac{P_C}{2}(2c^\dagger \rho c - cc^\dagger \rho - \rho cc^\dagger)\,, \end{aligned} \tag{8}$$

where $\gamma_c = \gamma_c^{\mathrm{T}} + \gamma_c^{\mathrm{L}}$ describes the total decay rate of the cavity photon-number $\langle c^\dagger c\rangle$. In Eq. (8) γ_c contains contributions from both the two reservoirs and P_c is the pumping rate depending on the populations of the organic energy levels weakly coupled to the cavity mode. In the weak excitation regime: $P_c = \sum_i \gamma_i^c \langle n_i^c\rangle$, where $\langle n_i^c\rangle$ is the population of the i-th level and γ_i^c is the corresponding spontaneous emission rate into the cavity mode. Starting from Eq. (3) a closed system of equations for the intracavity photon numbers $\langle c^\dagger c\rangle \equiv \mathrm{Tr}[c^\dagger c\rho]$ can be easily obtained,

$$\begin{aligned}\frac{d}{dt}\langle a^\dagger a\rangle &= -\gamma_a \langle a^\dagger a\rangle + 2g\,\mathrm{Im}\langle a^\dagger b\rangle + P_a \\ \frac{d}{dt}\langle b^\dagger b\rangle &= -\gamma_b \langle b^\dagger b\rangle + 2g\,\mathrm{Im}\langle b^\dagger a\rangle + P_b \\ \frac{d}{dt}\langle a^\dagger b\rangle &= \left[i(\omega_a - \omega_b) - \frac{\gamma_a + \gamma_b}{2}\right]\langle a^\dagger b\rangle \\ &+ ig\left(\langle b^\dagger b\rangle - \langle a^\dagger a\rangle\right)\,, \end{aligned} \tag{9}$$

with $\langle b^\dagger a\rangle = \langle a^\dagger b\rangle^*$. We are interested in calculating the steady-state emission spectra: $S_c(\omega) = \lim_{t\to\infty} 2\mathrm{Re}\int_0^\infty \langle c^\dagger(t)c(t+\tau)\rangle e^{i\omega\tau}d\tau$. According to the quantum regression theorem [29], two-time correlations $\langle A_n(t)A_m(t+\tau)\rangle$ follow the same dynamics of one-body correlation functions $\langle A_m(\tau)\rangle$ but with the one-time correlation $\langle A_n(t)A_m(t)\rangle$ as initial conditions. In our specific case the initial conditions are provided by the steady-state cavity occupations $\lim_{t\to\infty}\langle c^\dagger c\rangle$ and by $\lim_{t\to\infty}\langle a^\dagger b\rangle$. Hence we also need to derive the dynamics of one-body correlation functions:

$$\begin{aligned}\partial_t\langle a\rangle &= -i\tilde{\omega}_a\langle a\rangle - ig\langle b\rangle \\ \partial_t\langle b\rangle &= -i\tilde{\omega}_b\langle b\rangle - ig\langle a\rangle\,, \end{aligned} \tag{10}$$

where $\tilde{\omega}_c = \omega_c - i\gamma_c/2$. According to the input-output formulation of optical cavities [30], the PL spectra obtained collecting the light escaping from the cavity a (or b) are proportional to $S_a(\omega)$ and $S_b(\omega)$ respectively. In particular the output field can be determined by the following relationship:

$$c_{\mathrm{out}}(t) = c_{\mathrm{in}}(t) + \sqrt{\gamma_c^{\mathrm{T}}}c(t)\,, \tag{11}$$

where $c_{\mathrm{in}}(t)$ describes the input field operator. We are interested to the case where only one cavity has an embedded organic active layer ($P_a = 0$). In this case, by applying the quantum regression theorem, we obtain the following compact analytical expressions,

$$S_a(\omega) = \gamma_a^{\mathrm{T}}\frac{P_b}{\sqrt{2\pi}}\frac{g^2}{|(\omega-\Omega_1)(\omega-\Omega_2)|^2}\,, \tag{12}$$

$$S_b(\omega) = \gamma_b^{\mathrm{T}}\frac{P_b}{\sqrt{2\pi}}\frac{|\omega-\tilde{\omega}_a|^2}{|(\omega-\Omega_1)(\omega-\Omega_2)|^2}\,, \tag{13}$$

where Ω_i $(i = 1, 2)$ are the complex Rabi resonances:

$$\Omega_{(1,2)} = \frac{\tilde{\omega}_a + \tilde{\omega}_b}{2} \mp \frac{1}{2}\sqrt{4g^2 + (\tilde{\omega}_a - \tilde{\omega}_b)^2}\,. \tag{14}$$

We observe that the input field operators do not provide any contribution to the PL spectra since we are considering the case where there is no input light field exciting resonantly the cavity i.e. in the frequency range of interest where the spectrum is calculated. As it can be easily inferred from the obtained expressions, the two spectra differ each other significantly even at resonance $\omega_a = \omega_b$. It is also interesting to investigate the limit case of very small coupling $g \to 0$ or even $g << |\omega_a - \omega_b|$, Eqs. (12, 13). In this case, from Eqs. (12, 13), we easily obtain: $S_a(\omega) = 0$, and

$$S_b(\omega) = \gamma_b^T \frac{P_b}{\sqrt{2\pi}} \frac{4}{4(\omega - \omega_b)^2 + \gamma_b^2}\,. \tag{15}$$

As Eqs. (12) and (13) show, the coupling of the active cavity with an additional passive one can modify significantly the spectral properties of the emitted light. Equations (12) and (13) can be useful for the theoretical descriptions of PL measurements in double cavities. The procedure described in this section to derive Eqs. (12) and (13) can be easily generalized to the case of multiple cavities beyond two.

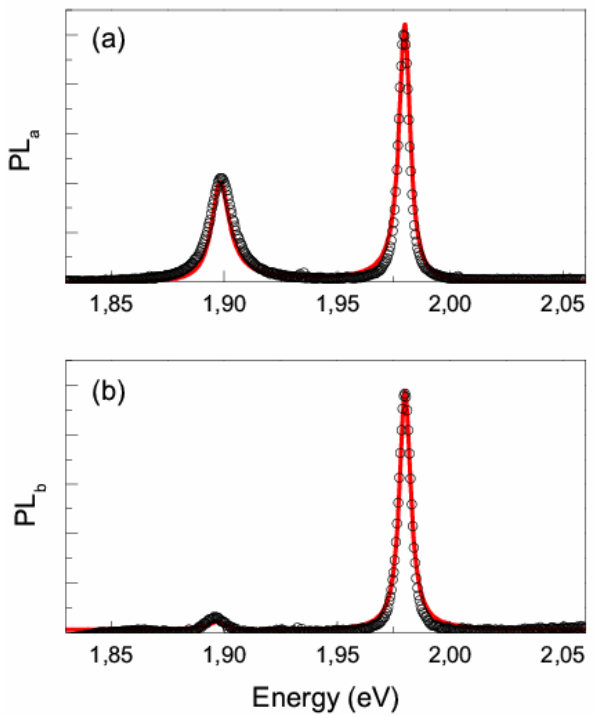

FIGURE 3. Comparison between experimental and theoretical results.

This theoretical approach, allows us to calculate the PL spectra which can be obtained for two cases. First by collecting the light emitted from the top side of the cavity (PL_a); second collecting the light emitted from the bottom side of the sample (PL_b). Figure 3a displays PL_a. The circles represent the experimental data collected at normal incidence from the side of the empty cavity, while the continuous line describes the best fit obtained by using Eq. (12). The obtained best fit parameters are: $\gamma_a = 4.5$ meV, $\gamma_b = 11$ meV, $\omega_a = 1.962$ eV, $\omega_b = 1.916$ eV. The theoretical spectra have been obtained using a coupling strength $g = 34$ meV, previously obtained from the reflectivity spectra at normal incidence. Figure 3b displays PL_b. The circles represent the experimental data collected from the side of the cavity with the embedded organic layer. The continuous line shows the corresponding theoretical calculation according to Eq. (13). In particular after fitting the PL_a data (Fig. 3a) we use the obtained fit parameters as input to describe the PL_b signal without using any other fit procedure (Fig. 3b). The agreement of the PL_b data with the theoretical calculation with no free parameters is rather impressive. The calculation procedure is fully reversible, i.e. it is possible to fit the PL_b and calculate the PL_a. The results show the ability of the theoretical model to fully catch the physics of the energy transfer between two strongly coupled subsystem under incoherent excitation. On the other hand this very good agreement confirms the good quality of this hybrid active device and the strong coupling between the two coupled MCs.

5. Triple Microcavities

The reflectivity spectra of the whole structure (see continuous line in Fig. 4 (left)) shows the three dips of the cavity modes at 1.78 eV, 1.88 eV and at 1.95 eV having a similar linewidth of 10 meV (full width at half maximum), corresponding to a quality factor Q of about 165. The Fig.4 (left) shows also the reflectivity of the single MC (dot line), centered at 1.88 eV, and of the double structure (dashed line), positioned at 1.77 and 1.88 eV.

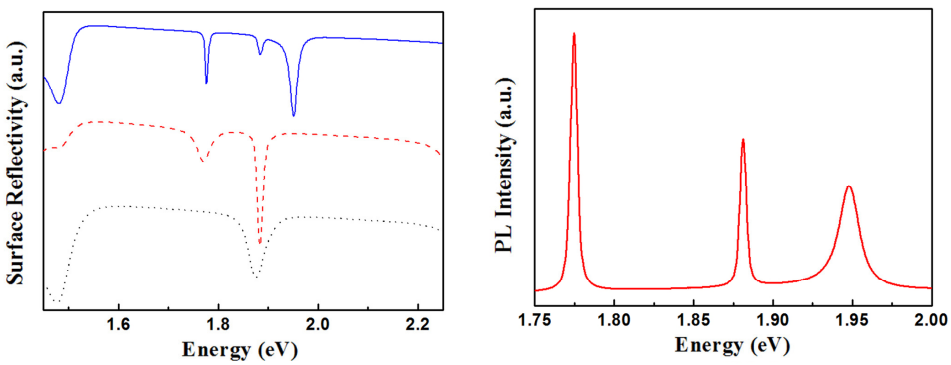

FIGURE 4. (Left) Reflectivity spectra for single (dot-dark), double (dash-red) and triple (continuos-blue) MCs. (Right) PL spectra of the triple coupled MCs.

These spectra clearly show a shift respect to the design energy of the single cavity, that, again, is a sign of the coupling between the cavities. The shift of the two side modes is not symmetric with respect to the energy of the single cavity mode. This effect may be attributed to the rapid wavelength dependence of the refractive index of the organic layer. At normal incidence (see Fig.4 (right)), the device emits three and well separated peaks each one corresponding to the optical modes of the three coupled cavities. The peaks are at the same energy of the reflectivity dips. This feature can be very interesting for the realization of devices capable of emitting multiple wavelengths that are desirable in several applications such as two-wavelength interferometry for distance measurements, terahertz difference signal generation, and frequency mixing. Furthermore, one other interesting application could be the realization of white light emitting device. In fact, controlling the coupling between the cavities and the width of the emitted lines, the device may emit large peaks at Red, Green and Blue wavelength resulting in a white emission.

6. Conclusions

The UHV thermal deposition on a Corning glass substrate was used to fabricate multiple active organic MCs. We investigated and simulated the behavior of two light emitting, double and triple, coupled MC structure working at room temperature. Their emission arises from an organic ultrathin layer at the center of the cavities. The reflectivity and the PL spectra clearly show that the cavities are coupled and that the structures emits over a large range of wavelength. These optical devices are considered the photonic analogue of a MC embedded QW in the strong coupling regime. In particularly, in the case of double MC, we report photoluminescence spectra obtained exciting incoherently the bottom cavity by its embedded organic layer. In this way we were able to investigate the strongly coupled system by exciting just one of the two subsystems and probing each of the two subsystems. The experimental results have been compared with the theoretical predictions of a quantum statistical model developed for the analysis of PL from strongly coupling systems. The excellent agreement demonstrates the predicting ability of the model. In conclusion our theoretical model can be very useful for the design of novel light emitting optoelectronic devices based on cavity embedded organic active molecules and these devices are very promising for the study of the rich physics and applications promised by active media in multi-cavity systems.

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