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Designing light emission with multiple organic based microcavities

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ABSTRACT

A light emitting structure consisting of three coupled microcavities has been realized and studied. All three cavities contain a luminescent organic thin film of tetrakis(4-methoxyphenyl)porphyrin and they are coupled by means of two same LiF/ZnS Distributed Bragg Reflector. The entire structure is enclosed between a bottom and a top Bragg mirror with different layer number. Reflectivity spectra collected at different growing steps allow us to demonstrate the effective strong coupling between the three bare cavity modes. The reflectivity spectrum of the whole structure shows the presence of three well defined cavity dips. At normal incidence, the device emits three, well separated, peaks each one corresponding to a delocalized optical mode of the coupled system, while at higher angles the peaks result blue shift. The experimental reflectivity spectra are in good agreement with a theoretical model based on the transfer matrix method. The model is applicable to any device, therefore, once the materials to employ as emitters are established, the cavity can be designed in order to amplify just the modes that correspond to the peak wavelengths of the emission spectrum and/or to change completely the spectral shape.

1. Introduction

Optical microcavities (MCs) are able to deeply change the light–matter interaction. For example the spontaneous emission of atoms can be significantly enhanced or reduced compared with its vacuum level by tuning discrete cavity modes in and out resonance with respect to the emitter. The main effects on spontaneous emission are spectral narrowing and intensity and directionality enhancement of the emission. Hence microcavity design is a potential method for obtaining efficient organic light emitting diodes (OLED) emitting pure primary colors for application in full-color display. Considerable attention has been paid to the study of multiple MCs. These structures have demonstrated their potential for developing various optoelectronic devices based on Rabi splitting, Rabi oscillations or other light–matter interaction effects [1–8]. Among these, it is worth mentioning the demonstration of parametric oscillation in a monolithic semiconductor triple MC with signal, pump and idler waves propagating along the vertical direction of the nanostructure reported in 2006 [5].

The operation principle of coupled-cavities is analogous to that of mechanical harmonic oscillators coupled by a spring. It is common knowledge that if two oscillators, having the same resonant frequencies, are coupled by some type of interaction, the resultant system will possess two resonant frequencies positioned symmetrically about the resonant frequency of the noncoupled oscillators and the splitting of the resonant frequencies will increase as the strength of the coupling interaction is increased. In this case n single MCs are coupled by n-1 dielectric mirror. The modes of the single MC in the structure are resonants and their interaction generates a shift of the modes of the coupled system to either side of the resonance wavelength. The coupling and the splitting are controlled by the transmission of the mirror shared between two cavities. Therefore, these devices can be designed to control light polarization, the emission direction and intensity of electromagnetic waves across a wide range of frequencies, accordingly such structures can be exploited to control the emission properties in terms of color, polarization and radiation pattern.

Among the active materials, organic molecules provide many advantages over the inorganic one. In particular the possibility of molecule functionalization plays a fundamental role in the design of devices to solve specific tasks [9–14]. Moreover organic materials are interesting due to their large exciton oscillator strengths and binding energies. These properties lead to a stronger interaction with the cavity mode and a stable, strongly coupled state at room temperature [15]. Therefore the special features and the flexibility of the organic molecules make them very interesting candidates for the development of high-efficiency optoelectronic and photonic devices [16–23]. In particular, the porphyrins provide an extremely versatile synthetic base for a variety of material applications. These materials have found broad applications as field-responsive materials, particularly for optoelectronic applications. For example, the facile substitution of the periphery of various porphyrins has generated a series of unusual liquid crystalline materials. The porphyrin ligand serves as a platform on which one can erect desirable molecular and material properties, including very large dipole

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moments, polarizabilities, and hyperpolarizabilities. The nonlinear optical properties of these materials are of special interest, in part for energy transfer with molecular control, and in part for potential applications in optical communications, data storage, and electro-optical signal processing.

In 2004, Sony reported that a white emitter with pixilated microcavity structures combined with color filters can achieve high brightness and good color saturation in full color active-matrix OLED displays [24]. Recently the technology of white organic light-emitting diodes (WOLEDs) is attracting growing interest due to their potential application in indoor and automotive lighting. Nevertheless, the spectral narrowing induced by high quality resonators reduces the color rendering index (CRI) and requires special design. In this framework, the multiple MCs were found to be a low-cost architecture to realize high color quality WOLEDs for lighting applications.

In this contest, innovative high performance WOLED architecture based on the coupling of organic MCs was realized [25,26].

In this work, we present a comprehensive theoretical and experimental study of the optical performances of multiple organic based MCs. Following above design requirements, we demonstrate that, using a simple model based on the Transfer Matrix Reflectivity (TMR) method, it is possible to simulate the entire structure taking into account both the coupling among the cavities and the optical properties of the active medium. Preliminary experimental results on this sample have been reported in an academic conference [27]. Each MC is designed to work at the same wavelength and contains a porphyrin thin film as active medium. Optically pumping the device, it emits at three different wavelengths. This special and simple design is very flexible in terms of spectral control and offers an easy way to improve the CRI still keeping high optical efficiency. The measured optical reflectivity of the device has been fitted by means of a Matlab code based on the TMR model [28,29]. The good agreement both of the fit and of the parameters obtained by the fitting procedure, confirms the easy optical performances control and the quality and reliability of the growth method.

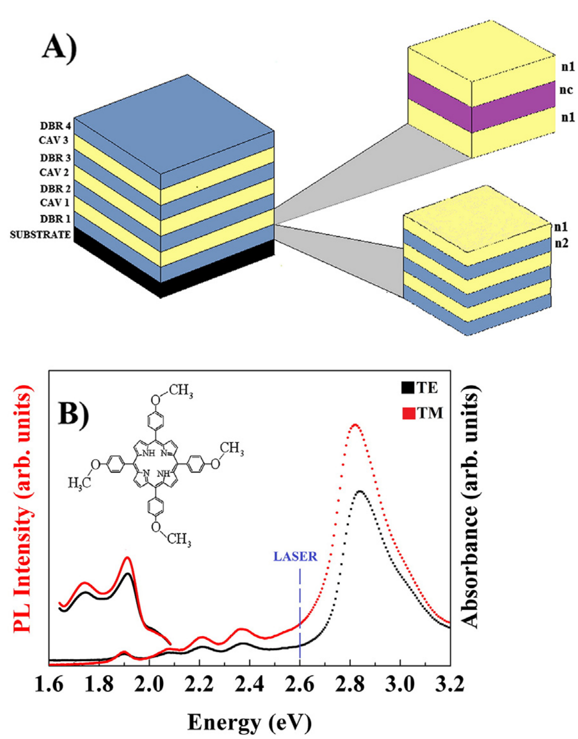

Fig. 1. Device structure consisting of three MCs coupled via two middle mirrors (A) and spectral properties and molecular structure of the tetrakis(4-methoxyphenyl)porphyrin (B).

2. Experimental

The triple MC (see Fig. 1A) consists of three identical structures optically connected by two common central Distributed Bragg Reflectors (DBR) built by a Ultra High Vacuum (UHV) deposition process. The structure is bounded on the outer sides by two mirrors. The design of the DBR requires the choice of two different dielectric media and the knowledge of the refractive index of the used materials. In our samples the DBRs are made of pairs of lithium fluoride and zinc sulfide. The refractive index at the working wavelengths is roughly $n_1 \cong 1.3$ and $n_2 \cong 2$ respectively. The high refractive index mismatch allows us to obtain reflectivity higher than 98% over a wide spectral region with a small number of periods.

The sample consists of pairs of LiF/ZnS in each Bragg mirror (respectively 6 pairs for the bottom mirror and 3.5 pairs for the others mirrors). The optical thickness of the DBR layers is $\lambda/4$. The three cavities consist of a 330 nm thick LiF thin film which embedded at their center a 10 nm thin tetrakis(4-methoxyphenyl)porphyrin (TMPP) layer. The TMPP molecular structure is reported in the inset of Fig. 1B. The optical properties of this molecule are characterized by a very strong Soret absorption band in the near UV (2.94 eV) accompanied by four weaker *Q*-bands at lower energies (Fig. 1B). Moreover the molecule emits an intense band at about 1.9 eV and a weaker one at 1.7 eV. The multilayer dielectric structure has been designed in order to exhibit resonances at wavelengths in the emission band of the organic layer. Of course, to improve the quantum efficiency, the pump frequency should coincide with absorption peak. In our experiment we used a cheap solid state laser whose wavelength is not resonant with the absorption peak but it is however inside the absorption band. Moreover in order to improve pumping efficiency, the DBRs were designed with a stop band lying outside the pumping frequency. In this way the pumping photons can easily reach the organic layer.

Some studies [30–32] have showed that the molecular structure preserves its integrity after the UHV thermal deposition. The organic thin film grows following a planar structure constituted by a single or multi-stacked molecule layer packing. The observed morphology could be considered the reason of the significant polarization dependence of the spectra that is a clear signature of an optical anisotropy [32].

To improve the device performances, the DBRs and the cavities have been designed to be resonant at 1.9 eV, i.e. close to the more intense emission component of the organic material.

The sample has been optically characterized measuring its reflectivity stepwise while growing the structure (Fig. 2). Fig. 3 shows the angle and polarization resolved photoluminescence (PL) spectra. The bottom side of the cavity was pumped by a DPSS laser working at $\lambda = 473$ nm. All the experiments were carried out at room temperature and in air atmosphere. In order to simulate and design the optical behavior of the device, a Matlab program was written using the TMR method [28]. The optical reflectivity of the device is described by means of a model which results from the product of the matrices of the three coupled structures:

$$MC = DBR1 * C1 * DBR2 * C2 * DBR3 * C3 * DBR4 \quad (1)$$

where C1, C2, and C3 are the cavities and DBR1, DBR2, DBR3, and DBR4 are the dielectric mirrors. Every DBR is defined as N pairs of layers with an optical thickness of $\lambda/4$, refractive index n_1 and n_2 and thickness d_1 and d_2. In our device n_1 is the refractive index of the LiF layer, while n_2 is the refractive index of the ZnS. In the working wavelength range of the device the refractive index of the LiF is almost constant, while the refractive index n_2 of the ZnS slightly changes following the semi-empirical relationship [33]:

$$n_2 = a + \frac{b}{\lambda^2} - i * c \quad (2)$$

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