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Spontaneous emission enhancement in planar metal-organic structures

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Abstract. We provide an experimental investigation of a Purcell enhancement in a silver-organic periodic structure with an organic material CBP (4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl). We fabricated two structures using a thermal evaporation technique with a different thickness ratio between layers. A rate of a fluorescence intensity decay of the periodic structures was experimentally measured in a near ultraviolet region. We show experimentally, that the rate of CBP molecule fluorescence decay increasing. A maximum registered value of the Purcell factor is close to 10.

1. Introduction

Organic light-emitting diodes (OLEDs) are attracting a great attention of researchers for display and lighting applications [1,2]. Current organic light emitting diodes suffer from the low light extraction efficiency. One of approaches to increase an external (by optimization of light extraction efficiency) and an internal (by tuning light-matter coupling properties) quantum efficiency of organic light emitting systems is using novel periodic dielectric-based and metal-based structures. Complex periodic structures allow to tune the light-matter interaction, i.e. manipulate the spontaneous decay rate of emitters. There are many experimental and theoretical works analyzing an influence of different types of periodic structures on emitters materials lifetime [3,4,5]. Recently, control on dye organic molecules emission lifetime using metal-dielectric structures was demonstrated [6]. Thus, it is very important to investigate light-matter interaction features in case of organic emitters to find ways to improve efficiency of existing organic based emitting systems and to find new applications. 4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl (CBP) is a very common organic material nowadays. Usually, CBP is using as host material in today's OLEDs. Furthermore, frequency of surface plasmon at the interface between CBP and silver is in CBP emission range. This fact makes CBP a promising material for plasmonics.

In this paper we have investigated properties of periodic multi-layered metal-organic (consist of silver and CBP layers) structure. Particularly, influence of presence of periodic structure on a fluorescence decay rate of organic emitting material CBP.



2. Experimental

We created two periodic metal – organic structures, based on two materials (CBP and silver) with a different CBP layer thickness. Using a Kurt. J. Lesker Spectros II system (organic and metal thin film deposition system consisting of a vacuum down to 1×10^{-6} mbar chamber) to fabricate metal-organic structures, 5 periods of CBP/silver layers were deposited on Al_2O_3 (sapphire) substrate with evaporation rates 0.1 nm/s and 0.05 nm/s, respectively. Figure 1 (a, b) shows schemes of considered structures. Also, to compare emission properties of structures with CBP emission properties, was fabricated thin CBP layer of the top of sapphire substrate as a reference sample.

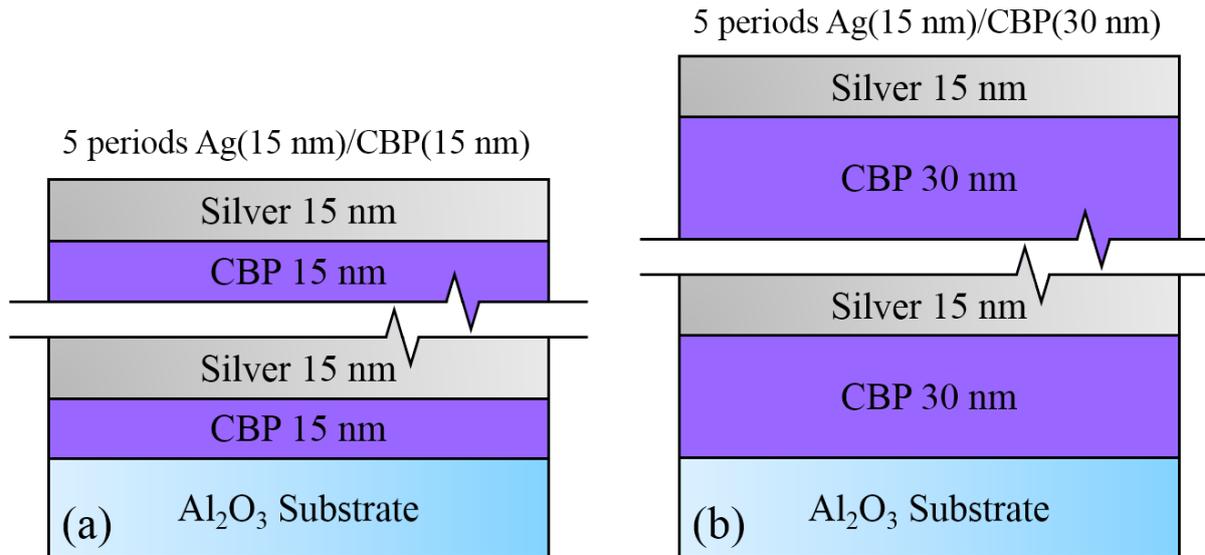


Figure 1(a, b). (color online) (a) Scheme of the periodic Ag 15nm/CBP 15 nm structure (5 pair 15 nm silver and 15 nm CBP). (b) Scheme of the periodic Ag 15nm/CBP 30 nm structure (5 pair 15 nm silver and 30 nm CBP).

A fluorescence spectrum of the CBP at room temperature (blue curve on figure 2) was measured by Jobin-Yvon Horiba Fluorolog FL3-22 spectrometer. Decay dynamics of CBP molecule excited states coupled with periodic structure and in a free space (reference sample) was measured by using a Time Correlated Single Photon Counting (TCSPC) method at room temperature and in 1×10^{-4} mbar vacuum. The structure was excited through the silver layer at 262 nm wavelength, the third harmonic of the output from a Coherent Mira 900F Ti:Sapphire oscillator tuned to 786 nm central wavelength, with a 76 MHz repetition rate. A 365 ± 10 nm bandpass filter with OD (optical density) > 4 outside this region was placed in the beam path between two lenses where the photoluminescence was collimated. Samples were excited at 45° angle to the substrate normal. Emission from sample was collected also at 45° angle to the substrate normal at 3.4 eV.

3. Results and discussion

In the periodic metal – dielectric structures interacting surface plasmons form bands outside the light cone. In this region at the frequencies corresponds to eigenfrequencies of the surface plasmons density of states (local amplitude) is increased. For the CBP/Ag interface frequency of surface plasmon corresponds to the emission band of CBP (violet – near ultraviolet region). The main parameter defining properties of the metal-dielectric structures is ratio between thickness of metal layer and thickness of dielectric layer. Therefore, was fabricated two structures with different ratios: $\eta = d_{\text{Ag}}/d_{\text{CBP}} = 1$ (thicknesses of silver and CBP layers are 15 nm) and $\eta = 0.5$ (thickness of CBP layer is 30 nm thickness of silver layer

is 15 nm). Both structures consist of 5 pairs of layers. Further increase of the thickness of the structure is pointless since the total thickness of Ag in the structure is comparable with light coherence length in silver.

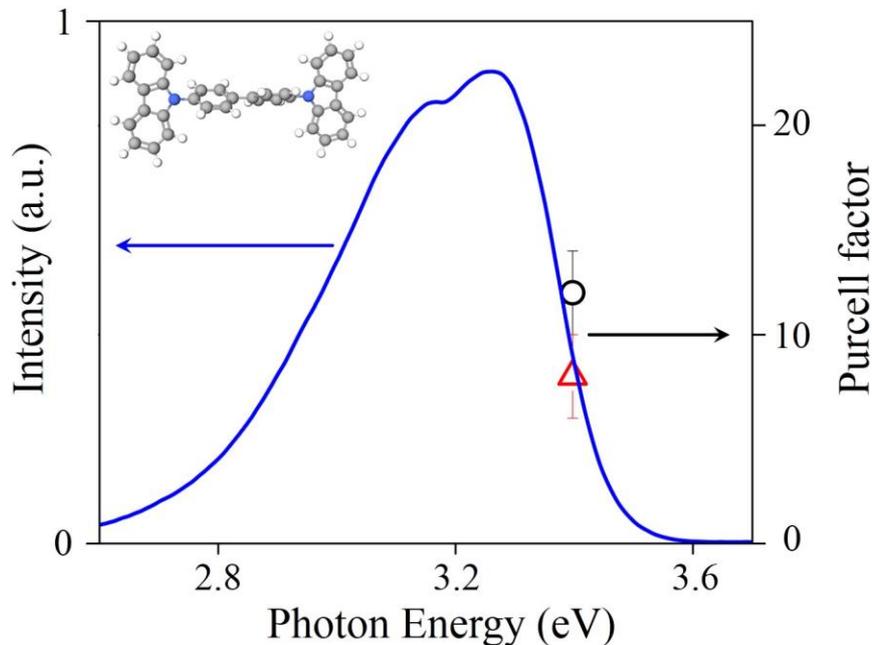


Figure 2. (color online) Blue curve (left axis) demonstrates an emission spectrum of CBP. Black circles and red triangles with error bars demonstrates the experimentally obtained values of the Purcell coefficient for Ag 15 nm / CBP 15 nm structure (right axis). The inset demonstrates molecular structure of CBP material (4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl).

As expected, there is a small difference between experimental results for CBP fluorescence decay rates in structures with the different thicknesses ratio. Figure 3(a, b) shows a comparison between the radiative decay of the periodic metal-organic structures correspond to $\eta=0.5$ (Figure 3 (a)), $\eta=1$ (Figure 3 (b)) and CBP reference film (black squares on both pictures). It can be seen, that the radiative decay rate of the CBP emitter in both periodic metal-organic structures for frequency 3.4 eV increases (an experimentally measured decay times for periodic structures are much smaller than for the bare CBP). Experimental curves could be approximate with the sum of two decay exponential functions with two different decay times (“fast” and “slow”). Both “fast” and “slow” times are less in the periodic metal-organic structures. Ratio between inverse lifetimes of the bare CBP and periodic metal- dielectric structures (a Purcell factor) close to 10. (Figure 2 demonstrates results for Purcell factor, that calculated for Ag 15 nm / CBP 15 nm, estimated using “fast” (circle) and “slow” (triangle) lifetimes).

4. Conclusions

Was provided an experimental investigation of the Purcell enhancement in the metal-organic periodic structures based on organic material (CBP) and silver with two different ratios between layers thicknesses. The rate of the fluorescence intensity decay of investigated structures was experimentally measured. We have demonstrated that in the periodic metal-dielectric structure the Purcell factor is of the order of ten.

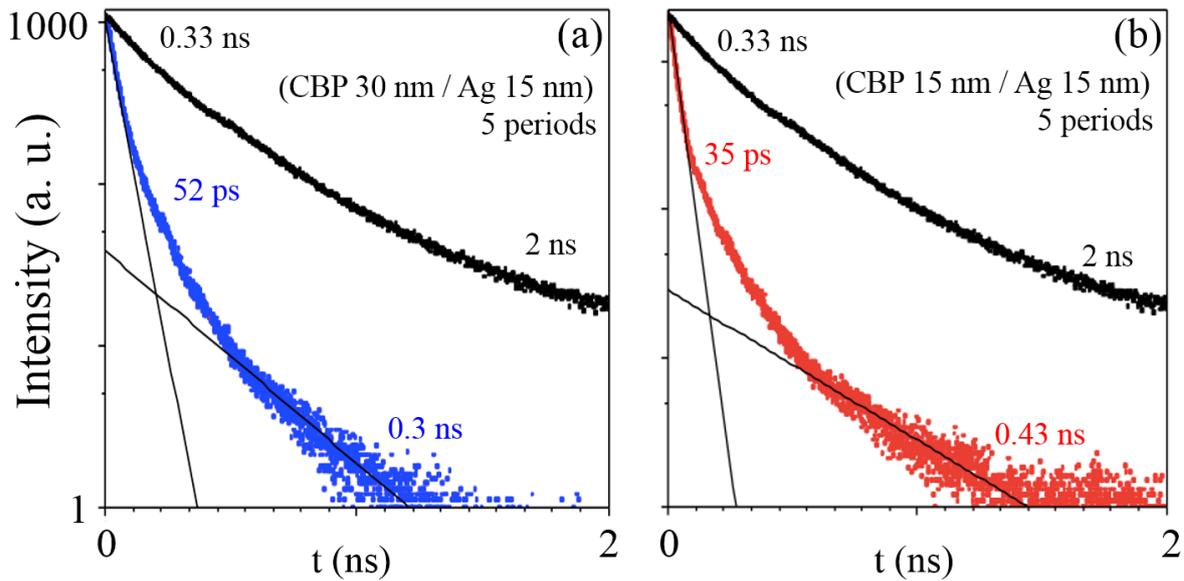


Figure 3(a, b). (color online) TCSPC measurement results. **(a)** Measured PL decay time of bare CBP (black curve) and periodic Ag 15 nm / CBP 30 nm structure (blue curve) at photon energy 3.4 eV. **(b)** Measured PL decay time of bare CBP (black curve) and Ag 15 nm / CBP 15 nm periodic structure (red curve) at photon energy 3.4 eV.

Acknowledgments

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