Polaritonic Devices Utilizing Nanoscale Films of J-Aggregates

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Exciton-polariton based photonic devices are a novel platform for realizing low threshold lasing [1] and optical switching [2] in a scalable integrated architecture. Use of organic materials as the excitonic component facilitates room temperature operation of such devices [3, 4]. Here we report room temperature exciton-polariton devices consisting of layer-by-layer (LBL) assembled thin films of polyelectrolyte and the J-aggregates of the cyanine dye TDBC [4] inserted in a resonantly-tuned planar $\lambda/2n$ optical microcavity with metal mirror and dielectric Bragg reflector (See Figure 1). Rabi-splitting and polaritonic dispersion are observed in the reflectance, transmittance, and photoluminescence measurements of the device (See Figure 2a,b). The devices exhibit Rabi-splitting of $\Omega_R = 125 \pm 7$ meV with a polyelectrolyte/J-aggregate layer that is only 5.1 ± 0.5 nm thick [6]. Furthermore, the linewidth of the lower energy polariton state, measured on resonance, is $\Gamma = 12.1$ meV. Hence, the ratio $\Omega_R/2\Gamma = 5.1$ indicates devices operating in a limit where the light-matter coupling (Ω_R) significantly exceeds competing dephasing processes (Γ). These figures of merit are achieved by virtue of the nanostructured film's large absorption coefficient of $\alpha \sim 1.0\text{x}10^6$ cm^{-1} and location of the layer at the microcavity anti-node. Because strong coupling is achieved with thin films of nanometer scale thickness control, the majority of the microcavity modal volume is available for integrating optically active materials such as colloidal quantum dots and fluorescent polymers in structures that can be used to investigate fundamental physical phenomena such as non-radiative energy transfer, and ultimately leverage the coherent properties of the strongly coupled states.

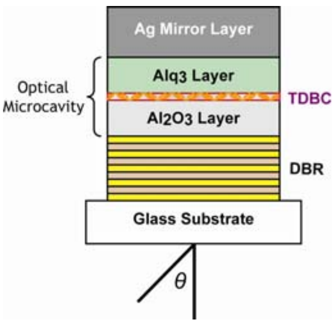

Figure 1: Device design of polaritonic structure. Microcavity consists of a DBR and a silver mirror layer. On top of the J-aggregate layer, a film of the small molecule Alq_3 is thermally evaporated.

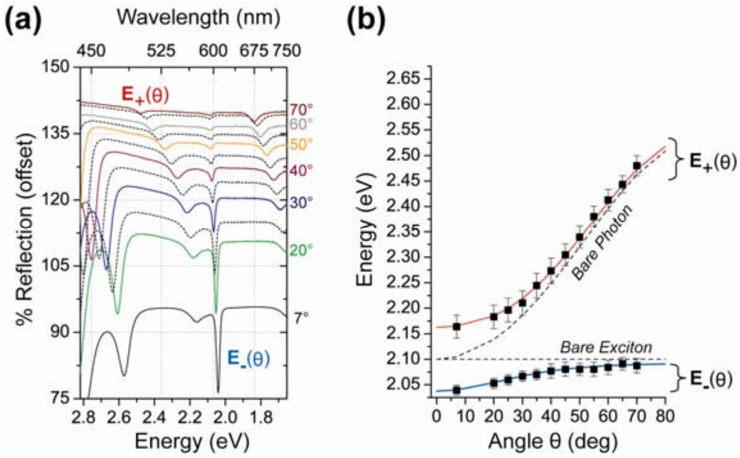

Figure 2: (a) Angularly resolved reflectance measurements and (b) dispersion relation for resonantly tuned polaritonic structure. The microcavity is resonantly tuned at normal (at $\theta = 0°$) to the exciton resonance of 2.10 eV. $E_-(\theta)$ denotes the lower energy polariton states, $E_+(\theta)$ the higher energy polariton states. Reflectance measured with TE polarized light. Measurements from $\theta = 20°$ to $70°$ are relative values. Data at $\theta = 7°$ is absolute reflectance.

References

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[5] TDBC's chemical formula is (5,6-dichloro-2-[3-[5,6-dichloro-1-ethyl-3-(3-sulfopropyl)-2(3H)-benzimidazolidene]-1-propenyl]-1-ethyl-3-(3-sulfopropyl) benzimidazolium hydroxide, inner salt, sodium salt).

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