

Strong coupling in optical microcavities containing the fluorescent molecular dye BODIPY-Br

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A microcavity is a structure in which an optically active semiconductor is placed between two highly reflective mirrors. Such a structure quantizes the local electromagnetic field, and confines photons into a series of discrete optical modes. Within the so-called 'strong-coupling regime' the confined cavity photons and the electronic excitations of the semiconductor (excitons) can no longer be thought of as separate eigenstates, but rather can couple together to form new quasi-particle states termed 'cavity-polaritons'. Polaritons necessarily have very different optical properties from their constituent parts and are a fascinating test-bed for fundamental physics, with non-equilibrium polariton-condensation suggesting potential applications in lasers, optoelectronics and ultra-fast optical switches.

We have explored [1] the optical properties of a series of strongly-coupled microcavities containing the fluorescent molecular dye BODIPY-Br (bromine-substituted boron-dipyrromethene) dispersed into a transparent polystyrene dielectric matrix (see figure 1(a)). Using temperature-dependent photoluminescence emission, white-light reflectivity and measurements of fluorescence quantum yield, we explore the population of polaritons along the lower polariton branch following non-resonant optical excitation (see Figure 1(b)). We find that both the cavity fluorescence quantum efficiency and the distribution of polariton states along the lower polariton branch is a function of exciton-photon detuning. Significantly, we show that in the most negatively detuned cavities, the emission quantum efficiency approaches that of a control (non-cavity) film.

Our previous work on microcavities that contain weakly-fluorescent J-aggregates of cyanine dyes indicated that exciton to polariton scattering processes were facilitated by the emission of energy in the formation of vibrational quanta. In contrast, we find that the population of polariton states along the lower polariton branch in BODIPY-Br microcavities can be most effectively described by a direct 'radiative-pumping' mechanism, in which polaritons are 'pumped' by photons that are spontaneously-emitted by excimer-like states that exist in a weakly-coupled reservoir. Using a simple fitting model we obtain an excellent agreement with measured photoluminescence as a function of temperature and exciton-photon detuning, and qualitative agreement with the measured photoluminescence quantum efficiency. We use our results to discuss the prospects for polariton-lasing and polariton-condensation in microcavities containing BODIPY-core molecular dyes.

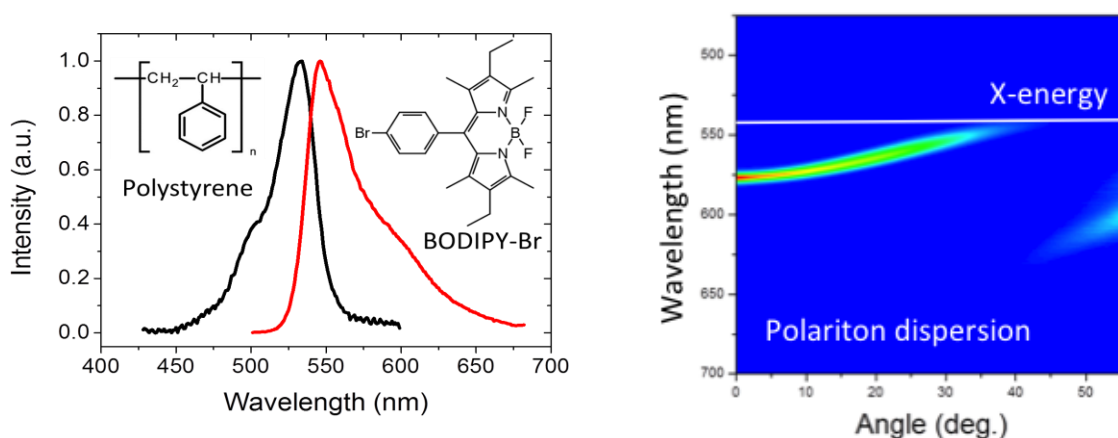


Figure 1: Part (a) (left) shows the absorption and fluorescence of BODIPY-Br when dispersed into a polystyrene matrix. Part (b) (right), shows the angular dependent fluorescence from a microcavity containing BODIPY-Br as a function of external viewing angle.

[1] Richard T. Grant, Paolo Michetti, Andrew Musser, Pascal Gregoire, Tersilla Virgili, Eleonora Vella, Marco Cavazzini, Kyriacos Georgiou, Francesco Galeotti, Caspar Clark, Jenny Clark, Carlos Silva, David G. Lidzey, Submitted 2016.