Strong Light-Matter Coupling Using a Robust Non-Cyanine Dye J-aggregate Material

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We demonstrate strong light-matter coupling using a novel J-aggregate material based on a dibenz[a,j]anthracene macrocycle that is robust under high power optical excitation. Organic materials inserted in optical microcavities have been used to achieve strong light-matter coupling at room temperature with coupling strength, i.e. Rabi-splitting, exceeding 100 meV. Strong light-matter coupling leads to polaritonic resonances for the coupled system that are superpositions of the underlying excitonic and photonic states. The resulting polaritonic states, due to their hybrid nature, possess remarkable optical properties, and in theory can exhibit laser-like coherent light emission at remarkably low excitation densities due to polariton condensation. A key hindrance to achieving polariton condensation thus far using organics has been exciton-exciton annihilation, which quenches excitations from the polaritonic states before they can condense. This process has been observed in polaritonic devices based on a variety of organic materials including single crystal anthracene and J-aggregates of cyanine dyes, and is a property of many organic materials in thin film. Here we demonstrate strong light-matter coupling using J-aggregates of a promising new dibenz[a,j]anthracene-based macrocycle that does not show exciton-exciton annihilation until very high excitation densities exceeding 20 MW/cm^2, while in thin films of the J-aggregated TDBC (cyanine dye) annihilation appears at 10 kW/cm^2. Thin films were prepared by spin coating a 6 mg/ml solution of the dye in chlorobenzene, yielding layers that were 15 nm thick with an RMS roughness of less than 1 nm. J-aggregation of the dye in these films was evidenced by the appearance of a narrow absorption line at 465 nm of FWHM = 15 nm and the concomitant disappearance of the monomer absorption band as the dye concentration was increased. The films absorb nearly 27% of the incident light at the J-aggregate absorption peak wavelength of 465 nm, which implies an absorption coefficient of $2.1 \times 10^5\ cm^{-1}$. The films show good photochemical stability and photoluminescence quantum yield exceeding 90%. Strong coupling was observed when thin films were incorporated into a $\lambda/2n$ planar optical microcavity. The microcavity consisted of a silver mirror and a dielectric Bragg reflector and had a modest empty cavity Q ~ 200. The J-aggregate layer was carefully situated at the cavity anti-node to maximize the light-matter coupling. Polaritonic devices using the new material exhibit Rabi-splitting of 130 meV at room temperature. Polaritonic dispersion was observed both in angular reflectance and photoluminescence measurements. Experiments are underway to optimize the cavity design and demonstrate organic based polariton condensation.