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 promising new J-aggregate material based on a dibenz[a,j]anthracene macrocycle [1], Figure 1a, that is robust under high power optical excitation. Strong light-matter coupling leads to polaritonic resonances that are superpositions of the underlying excitonic and photonic states [2] and can exhibit laser-like coherent light emission at remarkably low excitation densities due to polariton condensation [3]. A key hindrance to achieving polariton condensation thus far using cyanine dye J-aggregates has been exciton-exciton annihilation [4], which quenches excitations from the polaritonic states before they can condense. The J-aggregates of the dibenz[a,j]anthracene-based macrocycle show no signs of exciton-exciton annihilation until optical excitation densities exceeding 20 MW/cm^2, while in thin films of a typical J-aggregated cyanine dye, TDBC, annihilation appears at 10 kW/cm^2. Thin films of the macrocycle 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. The 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, Figure 1b, and the concomitant disappearance of the monomer absorption band as the dye concentration was increased [1]. The films possess an absorption coefficient of 2.1 x 10^5 cm^{-1} at the J-aggregate absorption peak wavelength of 465 nm, show good photochemical stability, and have photoluminescence quantum yield exceeding 90%. Strong coupling was observed when thin films of the macrocycle were situated in a $\lambda/2n$ planar optical microcavity consisting of a silver mirror and dielectric Bragg reflector. Devices exhibit polaritonic dispersion with a room temperature Rabi-splitting of 130 meV, Figure 2. Experiments are underway to demonstrate organic-based polariton condensation.

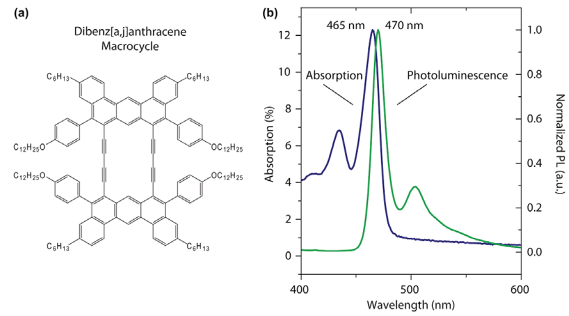

FIGURE 1: (a) Chemical structure of dibenz[a,j]anthracene-based macrocycle. (b) Optical absorption and photoluminescence spectra from a thin film prepared by spin-coating the compound onto a glass substrate. Thin film roughness of less than 1 nm was observed in AFM.

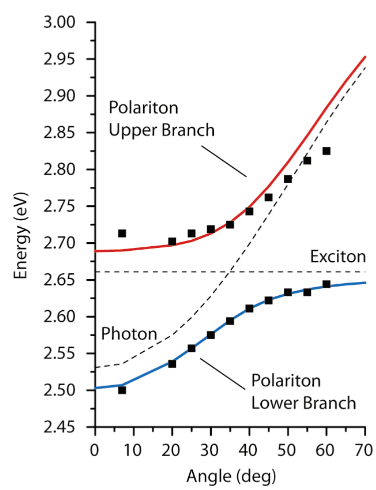

FIGURE 2: Polaritonic dispersion relation derived from angular device reflectance data. Anti-crossing of energy levels is observed at 35º from normal.

REFERENCES

[1] J. M. W. Chan, J. R. Tischler, S. E. Kooi, V. Bulović, T. M. Swager, "Synthesis of J-Aggregating dibenz[a,J]anthracene-based macrocycles", *Journal of the American Chemical Society*, vol. 131, pp. 5659-5666, April 2009.

[2] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, "Observation of the coupled exciton-photon mode splitting in a semiconductor quantum microcavity", *Physical Review Letters*, vol. 69, pp. 3314 – 3317, May 1992.

[3] J. Kasprzak J, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. J. Keeling, F. M. Marchetti, M. H. Szymanska, R. Andre, J. K. Staehli, V. Savona, P. B. Littlewood, B. Deveaud, L. S. Dang, "Bose-Einstein condensation of exciton polaritons", *Nature*, vol. 443, pp. 409-414, September 2006.

[4] J. Moll, W. J. Harrison, D. V. Brumbaugh, A. A. Muenter, "Exciton annihilation in J-aggregates probed by femtosecond fluorescence upconversion", *Journal of Physical Chemistry A*, vol. 104, pp. 8847-8854, October 2000.