Reversible Switching of Ultrastrong Light-Molecule Coupling

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Strong coupling of light and matter can give rise to a multitude of exciting physical effects [1]. Following the first observation of strong coupling between a cavity and dye molecules [2], organic molecules have been increasingly used for the study of strong coupling, since their large transition dipole moment permits the observation of Rabi splitting in the range of a few hundreds of meV at room temperature. However, organic molecules, as we show here, can also be used for combining light-matter interaction with molecular functionality [3], and for the study of strong coupling at the interface between physics and chemistry [4]. Remarkable, the light-molecule interaction in our system enters the regime of ultrastrong coupling [5], where the energy splitting is substantial fraction of the coupled transition energies, and new phenomena such squeezed vacuum state and generation of entangled photon pairs are predicted.

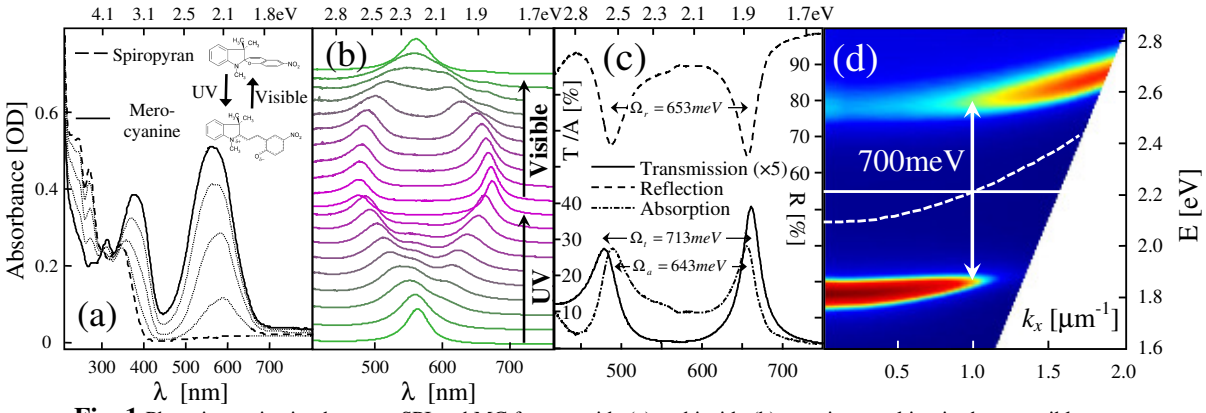

Fig. 1 Photo isomerization between SPI and MC form outside (a) and inside (b) a cavity, resulting in the reversible switching of ultra-strong coupling. (c) Normal incidence transmission, reflection and absorption spectra for the cavity at maximal coupling. (d) Cavity polariton dispersion diagram in the coupled state. The dashed line marks the "empty" cavity dispersion in the SPI state, and the horizontal line marks the molecular peak absorption energy.

Our system is a low-Q cavity made by two semi-transparent silver mirrors and a polymer gap between them. The gap is filled with photochromic Spiropyran molecules, which can be transformed between two chemical states – transparent Spiropyran (SPI) form, and visible-absorbing Merocyanine (MC) form, of which absorption spectrum shown in Fig. 1a. By tuning the cavity mode to match the peak absorption of the MC form, we resonantly couple the cavity and the molecules. Moreover, transformation into the MC state is carried by UV illumination, and the back reaction can be carried out by visible light, or thermally. Thus, this system offers all-optical control over the coupling strength, demonstrated in Fig. 1(a). Initially, the molecules are in the SP form, and the "empty" cavity peak is observed at 560nm. Upon UV irradiation, the single peak gradually splits, as the system goes into the strong coupling regime. At the final state, a Rabi splitting of 713meV is observed. This value amounts to 1/3 of the transition energy of the molecule, placing our system in the ultrastrong coupling regime. The process can be reversed, by using visible light (Fig. 1a). The observation of strong coupling is validated by the observation of similar splitting in the reflection (653meV) and absorption (643meV) spectra, shown in Fig. 1b. Finally, we measure the angle-dependent dispersion for the cavity (Fig. 1d, tuned such that resonant coupling occurs at an angle of ~35°), where the appearance of the upper and lower polariton branches is clearly visible, with a separation of 700meV between them at resonant coupling.

To conclude, our system is an ideal model to study strong light-matter interactions at the interface between physics and chemistry, exhibiting a remarkable coupling strength of 1/3 of the molecular transition energy, which brings molecular systems into the realm of ultrastrong coupling.

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