Polariton emission of strongly coupled non-oriented molecules with low dipole moments in a tunable optical microcavity

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Light-matter interaction of dipole transitions in ensembles of molecules is of special interest today [1]. Hybridization of energy states of molecular excitons and electromagnetic modes in cavities or in vicinity of plasmonic nanostructures leads to the formation of new polaritonic states, whose properties are significantly different from the properties of the original uncoupled states. The strong dependence of the polaritonic state properties on the electromagnetic mode characteristics ensures the possibility to control them using tunable microcavity modes [2].

In our previous studies we have developed a tunable microcavity cell (TMC) based on an unstable Fabry-Perot resonator that makes it possible to precisely control the cavity length at the nanometer scale and, hence, the mode volume, quality factor, and spectral position of the mode [3]. These properties of the TMC allow us to directly measure the dispersion of polaritons in the course of transmission as well as emission of the light. In most studies, ensembles of molecules with large dipole moments or highly oriented aggregates are used in order to increase the light-matter coupling strength [1]. This allows one to achieve strong coupling even with the use of optical microcavities with quite large mode volumes. However, majority of organic molecules, including biological one, have rather low values of dipole moments and do not form oriented aggregates. Therefore, the strong coupling regime has been previously reached mostly with the use of extremely localized plasmonic modes [4], which significantly limits the potential of practical applications for such coupled systems.

Here, we have encapsulated Rhodamine 6G (R6G) in a polymer matrix and placed it in the previously developed TMC. Prepared samples are non-oriented organic molecules with relatively low dipole moments. Nevertheless, an analysis of fluorescence emission and its dependence on the properties of the microcavity modes has **demonstrated emission from both upper and lower polariton state** with Rabi splitting energy as large as 225 meV [5]. Anticrossing behavior of the polariton dispersion curves has been demonstrated as well despite of the broad emission spectrum of R6G molecules. We speculate that the unusual phenomena of the emission from an upper polariton is due the large Rabi splitting energy, which have approximately the same value as the highest vibrational energy level. Demonstrated efficient emission from the "more energetic" upper polariton state can extend practical applications of strong coupling phenomena and also involve in these studies a wide variety of non-oriented organic (including biological) molecules with relatively low dipole moments.

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