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Charge-transfer chemical reactions in nanofluidic Fabry-Pérot cavities

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Abstract

We investigate the chemical reactivity of molecular populations confined inside a nanofluidic Fabry-Pérot cavity[1] (see Figure 1). Due to strong light-matter interactions developing between a resonant electromagnetic cavity-mode and the electric dipole moment of the confined molecules, a polariton is formed[2]. The former gets dressed by environmental vibrational and rotational degrees of freedom of the solvent[3]. We call the resulting polariton dressed by its cloud of environmental excitation a "*reacton*", since it further undergoes chemical reactions. We characterize how the *reacton* formation modifies the kinetics of a photoisomerization chemical reaction involving an elementary charge-transfer process. We show that the reaction driving-force and reorganization energy are both modulated optically by the reactant concentration, the vacuum Rabi splitting and the detuning between the Fabry-Pérot cavity frequency and targeted electronic transition. Finally, we compute the ultrafast picosecond dynamics of the whole photochemical reaction. We predict that despite optical cavity losses and solvent-mediated non-radiative relaxation, measurable signatures of the *reacton* formation can be found in state-of-the-art pump-probe experiments.

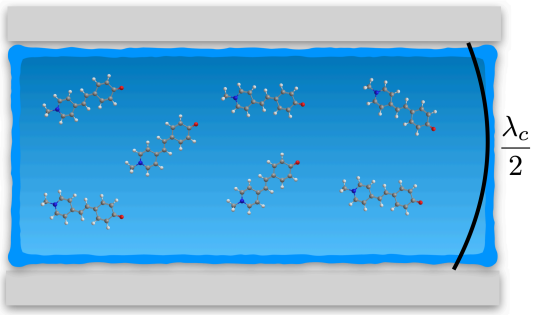

Figure 1: Pictorial representation of molecules of (E)-4-[2-(1-methylpyridin-1-ium-4-yl)vinyl]phenolate, in solution inside a nanofluidic Fabry-Pérot cavity.

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