

Sculpting chemical landscapes inside optical cavities

James A. Hutchison, Anoop Thomas, Jino George, Xiaolan Zhong, Thibault Chervy, Atef Shalabney, Eloïse Devaux, Cyriaque Genet and Thomas W. Ebbesen

Université de Strasbourg & CNRS, Institut de Science et d'Ingénierie Supramoléculaires (ISIS), Strasbourg 67000, France

E-mail: hutchison@unistra.fr

Light-molecule interactions can be divided into the weak and the strong. They can involve a single molecule or collective states of many thousands of molecules, and involve optical fields at their zero-point (vacuum) energy, or at very high intensity. Only with control over the gamut of these interactions can we realise the full potential of photocatalysis, photovoltaics, display technology, etc.

In this contribution I summarize our recent work on modifying chemical reaction landscapes by strong coupling the vacuum fields of an optical cavity with collective states of organic molecules. By placing molecules at high density inside optical cavities, coherent exchange of photons between the molecules and the cavity generate new, hybrid light-molecule states with consequences not only for chemical catalysis but also energy and electronic transport.

Previously we showed that coupling of electronic transitions of molecules could influence the rate of a photochemical reaction.^[1] More recently we have shown that vibrational transitions of molecules can also be strongly coupled with light,^[2] leading to changes in the rate of a ground state, thermally-induced dissociation reaction.^[3] The latter studies suggest the ground state Morse potential of the system is reshaped by vibrational strong coupling. The temperature dependence of the reaction rate suggests profound changes also to the nature of the transition state. Further potential for chemistry in optical cavities will be discussed.

Funding: International Center for Frontier Research in Chemistry (icFRC, Strasbourg), the ANR Equipex Union (ANR-10-EQPX-52-01), the Labex NIE projects (ANR-11-LABX-0058 NIE), and CSC (ANR-10-LABX-0026 CSC) and USIAS within the Investissement d'Avenir program ANR-10-IDEX-0002-02.

References:

- [1] J. A. Hutchison, T. Schwartz, C. Genet, E. Devaux, T. W. Ebbesen, *Angewandte Chemie Int. Ed.*, **2012**, 51, 1592
- [2] A. Shalabney, J. George, J. A. Hutchison, G. Pupillo, C. Genet, T. W. Ebbesen, *Nature Communications*, **2015**, 6, 5981
- [3] A. Thomas, J. George, A. Shalabney, M. Dryzhakov, S. J. Varma, J. Moran, T. Chervy, X. Zhong, E. Devaux, C. Genet, J. A. Hutchison, T. W. Ebbesen, *Angewandte Chemie Int. Ed.*, **2016**, 55, 11462