Vibrational strong coupling in ultra-compact nanogap patch antenna cavity Govind Dayalo, Ikki Morichika and Satoshi Ashihara

Institute of Industrial Science, The University of Tokyo, 4-6-1, Komaba, Meguro-Ku, Tokyo

153-8505, Japan

E-mail: dayal@iis.u-tokyo.ac.jp

There is a growing interest in the study of polaritonic systems, mixed states of photons and vibrational excitons, for both, the observation of quantum macroscopic phenomena, and the realisation of all-optical devices that could offer limitless advantages for controlling chemical reactions and molecular sensing [1]. Most experiments on vibrational strong coupling with ensembles of molecules involve wavelength scale planar microcavities that are either metallic or based on distributed Bragg reflectors. Here, we report the design, fabrication and characterization of an ultracompact nanogap patch antenna cavity that operate in the strong coupling regime. In our system, the nanogap patch antenna cavity which is filled with an ensemble of infrared active molecules exhibit ultra-small mode volume $(0.0011\lambda^3)$, strong field confinement, with the advantage of being angle insensitive $(\pm 40^\circ)$. When the nanogap patch antenna cavity mode is tuned with the molecular resonance, the system enters the strong coupling regime resulting in the formation of hybrid half-light half-matter excitations with modified energy levels as shown in the figure below. Our results show that subwavelength size nanogap patch antenna cavity combined with an infrared active molecular layer is an ideal system for achieving vibrational strong coupling at the nanoscale, opening new possibilities to change the chemical reactivity landscape and to use it as a tool in ultrasensitive spectroscopy down to single-molecule level.

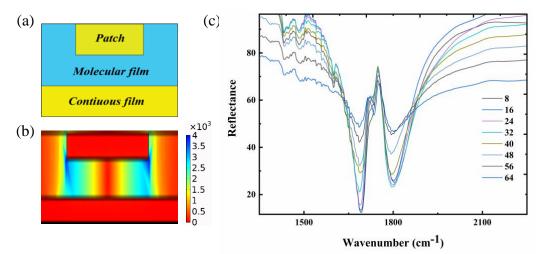


Figure 1 (a): Schematic diagram of the nanogap patch antenna cavity. (b) Simulated electric field of the nanogap patch antenna cavity showing strong field enhancement within the cavity. (c) Measured FTIR spectra of the fabricated cavity for different angle of incidences.

Reference: [1] T. W. Ebbesen, "Hybrid Light-Matter States in a Molecular and Material Science Perspective," Acc. Chem. Res. 49, 2403 (2016).