Plasmonic Platforms for Polaritonic Chemistry

Abstract: We are developing experimental platforms to probe vibrational strong coupling (VSC) between molecules and resonant infrared (IR) nanophotonic architectures, in order to understand how this coupling can fundamentally control chemical reactivity, as well as enable new classes of light-matter interaction. This method of altering the potential energy surface of a chemical process via coherent, electromagnetic perturbation of vibrating bonds has also been termed “polaritonic chemistry”. We employ a combined experimental strategy leveraging expertise in (1) the design of IR “metasurfaces” composed of plasmonic metal substrates that provide tailorable VSC to molecules within their optical near-field; and (2) multiple continuous wave (CW) spectroscopic techniques that enable analysis of several non-equilibrium, dynamic electronic effects in the metal substrate. Taken together, these tools allow studies into new regimes of spectral bandwidth (e.g. simultaneous multi-mode coupling), coupling strength, and time domains (e.g. studies of long lived and steady-state phenomena) that have been inaccessible using conventional optical cavities and time-resolved spectroscopies performed to date. Vibrational strong coupling is fundamentally interesting because it is a coherent interaction between radiation and molecular motion. The direct manipulation of a molecular process using externally controlled forcefields to obtain a desired outcome, i.e. “coherent control” or “quantum control”, has been a long-standing goal connected to the central aims of chemical science. Thus, this presentation will discuss the limits of chemical analysis and chemical control at interfaces leveraging a framework based on coherent interactions between controllable features of the engineered surface geometry and the molecular systems under study.

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