Vibrational strong coupling between molecular vibration and subwavelength plasmonic cavity supporting gap plasmon mode

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Abstract: We report on strong coupling between molecular vibrational resonances of polymethyl methacrylate (PMMA) molecules and gap plasmon resonance of an ultrathin plasmonic cavity in the mid-infrared range. The strong coupling is achieved when the molecular vibrational mode and plasmonic cavity exchange energy faster than their relaxation rates and it is maximum when two relaxation rates are equal [1]. In this work, we designed, fabricated and characterized a composite medium consisting of a thin PMMA layer sandwiched between the nanoantenna array and a continuous metallic thin film to achieve vibration strong coupling. The spectral position and the relaxation rate of gap plasmonic resonance are tuned through the molecular resonance of the PMMA molecules (at 1730 cm^{-1}) to go from weak to strong coupling regime. Strong coupling between vibrational modes and gap plasmon mode leads to the formation of new hybrid light-matter states called polaritonic states (@ 1690 cm^{-1} & 1810 cm^{-1}), separated by the vacuum Rabi splitting (120 cm^{-1}). Thin film coupled nanoantennas with sub-wavelength gaps have shown great potential in nanophotonic applications because they offer the ultimate electric field confinement in the gap. Our work is complementary to earlier work using microcavities and provides a new approach to achieve strong coupling with a nanoscale plasmonic cavity ($\lambda/25$) and the possibility to modulate the strong coupling regime by changing the gap thickness of the cavity and the lattice period of the nanoantenna array.

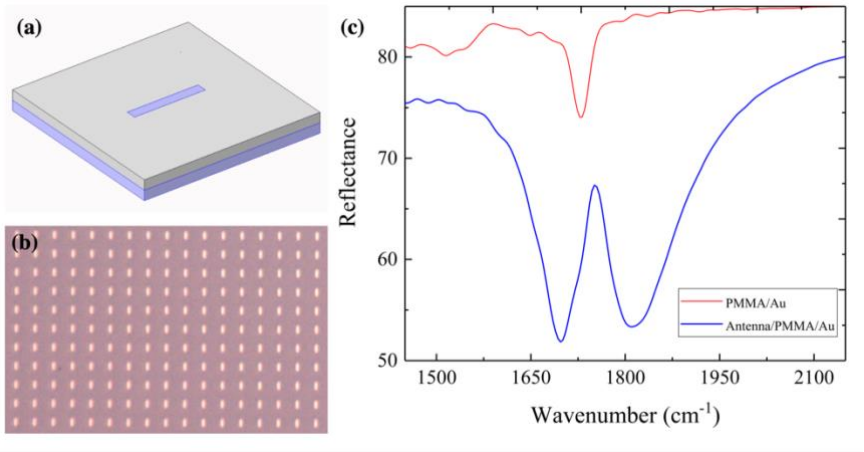

Figure 1.(a) Schematic diagram of the plasmonic cavity consisting of thin film coupled nanoantenna with a sandwiched PMMA layer. (b) Optical microscopic image of the fabricated Au nanoantenna arrays. (c) Reflectance spectra of the PMMA layer with a back reflector with (red curve) and without (blue curve) nanoantenna array on top.

Reference: 1. Shalabney, A. et al. Nature Comm. **6**, 5981 (2015).