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WITHDRAWN - Spectroscopy and Imaging of Strongly-Coupled Plasmon-Phonon Modes

Strong plasmon-phonon (pl-ph) coupling is a key process in nanoscale light-matter interaction physics. Optical probes have dominated strong-coupling studies, but they offer limited spatial resolution. Nowadays, atom-wide sub-5 meV electron beams generated in transmission electron microscopes allow us probing infrared (IR) excitations (e.g. phonons, plasmons, etc.) with better spatial sensitivity [1,2]. We present a spectroscopy study of strong pl-ph coupling in mid-IR antennas using spatially-resolved electron energy loss spectroscopy (EELS). We imaged the spatial distribution of strongly-coupled ph-pl modes, measured their dispersion and coupling constant, revealing unique spatial behavior of polaritonic hybrid modes in mid-IR antennas [3].
The mid-IR structures consist of double-antennas composed by a single rod-like Al plasmonic antenna attached to a rod-like a-SiO2 phononic antenna, side by side. We adjusted the length of the double-antenna in such a way that the plasmon response of the Al antenna lies within the upper Reststrahlen band (RB) of the SiO2. Optimal pl-ph coupling conditions are achieved for antennas of 3 μm in length. The EELS spectra of the Al/SiO2 antennas show double-peak resonances as a result of the pl-ph coupling. Each peak in the double-peak resonance is associated with the formation of one hybrid pl-ph mode. We determined the dispersion curve associated with the polaritonic modes and found Rabi splitting as large as 26 meV within the RB. Our simulations indicate the appearance of only two coupled pl-ph modes, despite the large variety phonon polariton modes involved in the coupling. We measured the coupling constant of the pl-ph interaction (g = 18 meV), meeting strong coupling requirements. Our work suggests that geometry plays a relevant role in the development of strong-coupling hybrid platforms. We also imaged the spatial distribution of strongly-coupled pl-ph modes and found that is akin to the distribution of dipole optical modes in rod-like antennas.
Our spectroscopy and imaging work enables a broader understanding of strong coupling phenomena between elementary excitations for nanophotonics.

[1] M. J. Lagos, et al, Nature 543 (2017) 529
[2] M. Lagos, et al, Microscopy 71 (2022), i174
[3] M. J. Lagos, et al, ACS Photonics 8 (2021) 1293

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