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Imaging Hybrid Plasmon-Phonon Modes in Mid-Infrared Antennas

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Coupling between elementary excitations is ubiquitous in quantum matter. Strong plasmon-phonon (pl-ph) coupling can modify the energy landscape in the mid-infrared range, providing opportunities to alter the energy states of infrared polaritons beyond the standard tunability approaches. Optical probes have dominated strong-coupling studies but they offer limited spatial resolution. Highly monochromatic atom-wide electron beams allow us to probe collective excitations (plasmon, phonons) with better spatial resolution [1]. For instance, plasmon coupling was investigated in particles [2], bulk and surface phonons were imaged in individual nanostructures [3], and pl-ph modes were probed in hybrid platforms [4]. In this work, we present an experimental EELS study of pl-ph coupling in double-antennas using low-cost materials (Al and SiO_2) with Rabi splittings as large as 26 meV. The spatial distribution of strongly-coupled modes was imaged, revealing unique spatial behavior of polaritonic excitations in mid-infrared hybrid antennas.

We used a Nion UltraStem microscope equipped with an aberration corrector and monochromator to study the mid-infrared response using a ~ 1.5 - 2 Å probe with an energy spread of ~ 9 meV, at 60 kV. To study the interaction between surface plasmon and phonon polaritons, we fabricated suspended rod-like Al/SiO_2 double-antennas with extreme aspect ratio (up to 40). Each component consists of a rod-like plasmonic and a phononic antenna, which are attached side by side (see schematic in Fig. 1a) minimizing the interaction with the support. Single and double-antennas were fabricated using standard methods of FIB preparation under low-dose conditions to prevent irradiation damage.

We initially probed single SiO_2 antennas of different lengths and found that the surface phononic response is confined within the Reststrahlen bands, as expected in finite systems sustaining surface optical phonons modes [5]. By changing the length of the SiO_2 antenna, we found that the scattering also remains constrained within the Reststrahlen bands. In contrast, Al plasmonic antenna response can be tuned over a wider range by adjusting the antenna length [6]. For instance, the dipolar modes of single aluminum antennas of 1 and 6 μm in length have energies at ~ 460 meV and ~75 meV, respectively. This clearly shows plasmonic tunability over a wider spectral range (ΔE ~ 540 meV), in comparison to the phononic antenna (ΔE ~ 20 meV).

To study the pl-ph interaction, the plasmonic response of the aluminum structure was tuned by adjusting its length in such a way that its response lies within the upper Reststrahlen band of the SiO_2. This condition was achieved with double-antennas of ~ 3 μm in length. Figure 1a shows several EEL spectra acquired in a double-antenna (3.1 μm in length) revealing a strong scattering variation due to the presence of several polaritonic excitations. Notice that some EEL spectra exhibit a resonance splitting of ~ 26 meV within the Reststrahlen band. This split is a very robust phenomenon, which reveals the formation of new hybrid pl-ph excitations due to the coupling between the modes of each antenna, one due to the collective motions of electrons (plasmon) in the aluminum and other due to the collective motion of ions (phonons) in the silica. Each peak in the double-peak resonance indicates the excitation of one of the new generated pl-ph modes. They are denominated symmetric and anti-symmetric modes due to their charge configurations (see inset Fig. 2b).

We measured the dispersion relationship between the oscillation frequency and the wave-vector of the hybrid pl-ph mode (Fig. 1b). We notice that a split in the dispersion curve appears within the upper Reststrahlen band as an indication of pl-ph coupling, where the plasmon and phonon bands cross. Our simulations also show similar repulsive behavior between the coupled modes. The coupling constant of the pl-ph interaction was determined (g = 18 meV), indicating that the coupling conditions are in the strong regime. In spite of the modest quality-factor materials, the fabricated Al/SiO_2 double-antenna equals performance of platforms of different geometries that capitalize on novel materials [4]. Our work suggests that geometry plays a relevant role in the development of strong-coupling hybrid platforms.

Furthermore, the scattering associated with the excitation of strongly-coupled pl-ph modes was imaged (Fig. 1c) and we found that its spatial distribution is similar to the distribution of dipole modes in rod-like antennas. The maps of the symmetric and anti-symmetric modes share several similarities along the double-antenna but some minor variations can be noticed in the transverse direction likely associated with their charge configurations. Other surface polaritonic modes were also imaged showing different spatial distributions. Spatially-resolved EELS measurements across SiO_2/Al interfaces revealed a drastic scattering variation within sub-10 nm distances due to the charge symmetry of each pl-ph mode.

In summary, we have probed a hybrid antenna composed by one plasmonic Al structure attached to a phononic SiO_2 structure. We found evidence of strong coupling between plasmon and phonons, as revealed by the energy split in the dispersion curve. We have imaged strongly-coupled pl-ph modes bringing important physical insights into the local density of electromagnetic states and into the energy exchange over a Rabi oscillation cycle. Our results suggest that geometry plays an important role in the design of hybrid strong-coupling platforms [7].

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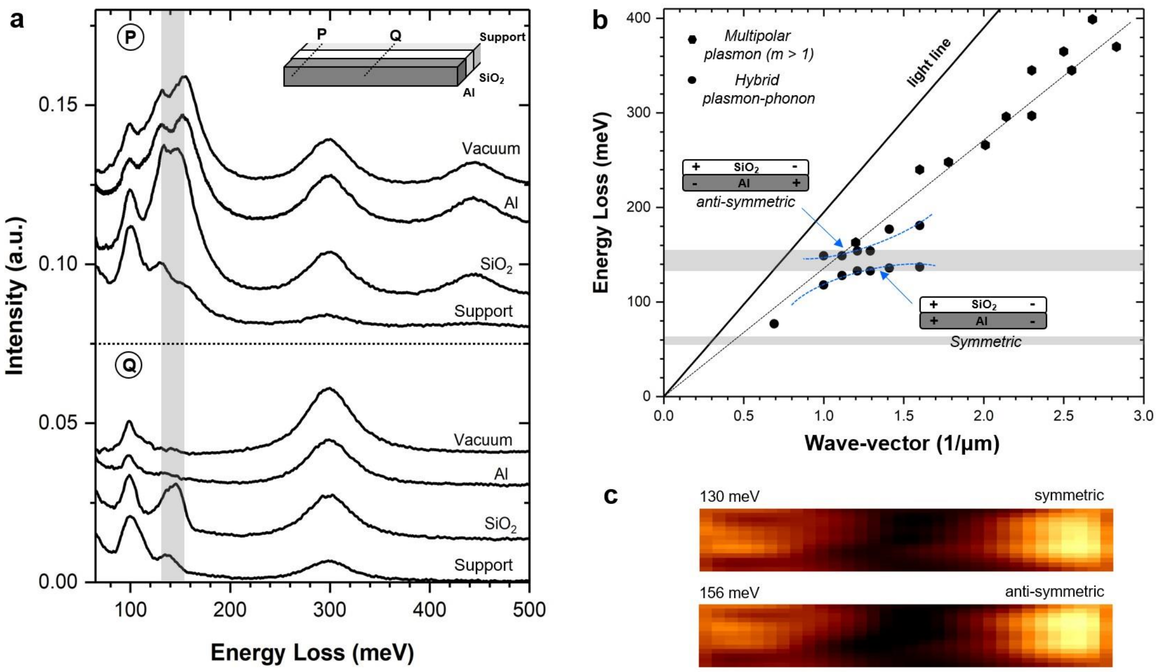

Figure 1. a) Spatially-resolved EELS spectroscopy of polariton modes in an Al/SiO2 double-antenna. Inset shows schematic of the suspended double antenna. EEL spectra were acquired across the antenna interfaces at two different locations (P and Q) indicated in the inset. Gray band indicates the Reststrahlen band. b) Dispersion curve of coupled plasmon-phonon modes and plasmon polaritons. Notice the energy split formed in the upper Reststrahlen band. Charge configurations of each plasmon-phonon mode are indicated in the dispersive branches. The black line represents the light line and the dotted black and blue curves was plotted to guide the eyes. c) Maps of the strongly-coupled plasmon-phonon modes. A dipole-like spatial distribution of scattering signal is evident.

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