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Ultrafast imaging of plasmonic-phononic structures

[1] Leader: Name　Oliver B. Wright

Affiliation　Hokkaido University, Professor

Participants:

1) Ronald Ulbricht,　Hokkaido University,　JSPS Fellow

2) Hidenori Mimura, Research Institute of Electronics, Professor

[2]　Purpose of research

We propose to optically image plasmonic nanostructures with sub-micron resolution using tightly focused ultrashort optical pulses that generate coherent phonons with acoustic frequencies up to 100 GHz and wavelengths down to 100 nm. We shall thereby determine the effect of the surface plasmon polariton (SPP) modes on the phonon modes.

The samples consist of gold nano-voids with diameters in the several-100 nm range. Experimental results that we recently published (see T. Kelf et al., N. J. Phys. 15, 023013, 2013) have allowed us to demonstrate that the proposed imaging is possible, but the role of the plasmonic modes has not been properly elucidated.

By collaboration with Shizuoka University we sought to better characterize the sample optical modes by white-light spectroscopy, and thereby use the results to carry out tuned optical pump and probe experiments on ultrashort time scales. This should reveal the role of the plasmonic modes on the optical modulation produced by the phononic modes of the samples.

Research content

Samples:

Nano-void samples of Au void diameter 1600 nm on glass substrates were used (see Adv. Mater. 16, 90, 2004). The samples have a graded height and so the void depth can be varied by choosing the excitation point on the sample

Measurements

We carried out characterization with white-light spectroscopy using a white-light laser spectrometer from the near-IR to visible wavelengths. The spectra were taken for voids with a height-to-diameter ratio ~0.9. This allowed us to indentify several Mie modes representative of the near-hollow spherical voids.

GHz vibrations for spectrally characterized voids of varying diameter heights were measured with coincident 10-micron diameter pump and probe spots (200 fs pump and 1 ps probe pulse durations) at normal incidence with sub-nJ pulses at 80 MHz repetition rate. By temporal Fourier analysis we identified the excited GHz vibrational modes

The dependence of the vibrational mode frequencies and phase on the optical probe wavelength was investigated by optical-wavelength scanning of the probe in the range 720-920 nm with a fixed pump wavelength of 415 nm.

[3]　Results

Figure 1 shows the white-light spectroscopy results, where the Mie mode resonances are clearly visible.

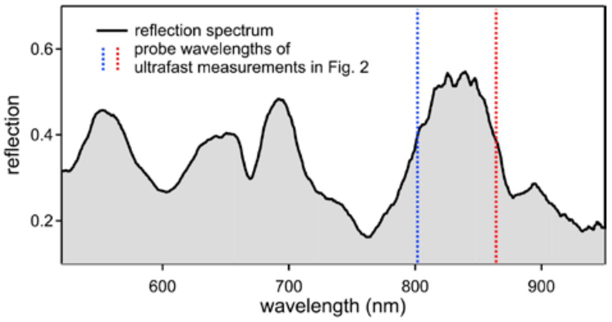

Fig. 1 broadband reflection spectrum of nanovoids (grey-shaded); the ultrafast measurements shown in Fig. 2 were performed on the blue side (800nm) and the red side (860nm) of the plasmon peak at 830nm.

In the pump-probe measurements, which measure the excitation-induced change in the optical reflectivity at the probe wavelength, we found several oscillations that correspond to acoustic resonances in the GHz range, as shown in Fig. 2.

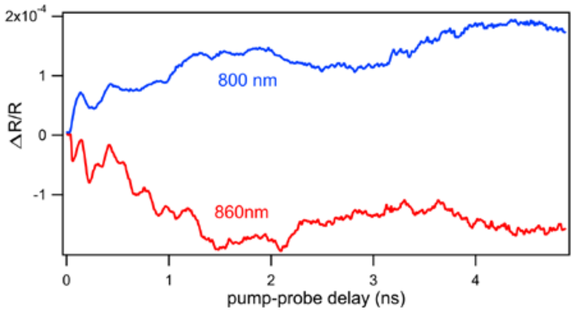

Fig. 2 Pump-probe measurements, yielding the pump-induced differential change in reflection ΔR/R, at probe wavelengths of 800nm (blue) and 860nm (red); several oscillations are visible, originating from the excitation of phonon modes that couple to the plasmon mode at 830nm (see Fig. 1); the modulation of the plasmon peak position due to the induced vibrations is evident from the change of the phase of the oscillations.

We also found that the phase of the acoustic vibration inverted across the localized-plasmonic Mie resonances. In the example shown in the figues the peak wavelength of the resonance is at 830nm, and the phase of the acoustics oscillations flips between probing on the blue side (800nm) and the red side (860nm) of the plasmon resonance. This is as expected from a simple model for acousto-optic coupling.

Future prospects and research

These results show great promise for manipulating the coupling between plasmons and phonons in complex periodic structures. In the future there remains much research to be done to determine the effect of the void size, truncation and to explain the results quantitatively. It is particularly important to try to distinguish localized SPP contributions (probably dominant in the present experiments) and non-localized SPP contributions. The results with different void truncations, availablel with our current sample, should help elucidate this. Also, what determines the maximum opto-acoustic coupling should be determined.

[4] Documentation

A conference paper was submitted to Meta14 in Singapore on the subject of this work entitled "Gigahertz vibrations in a graded plasmonic–phononic crystal."

(Also see T. Kelf et al., N. J. Phys. 15, 023013, 2013)

出張報告

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氏　名：Ronald Ulbricht
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