

# Optical Probing of Acoustic Vibration through Coupled Plasmon Resonance

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Vibration in nanostructures is of considerable interest in recent years, because plasmon resonances and mechanical vibrations are coupled together that leads to stronger modulation of optical response<sup>1</sup> and it plays a nonnegligible role in detecting mechanical properties of matters in many fields such as material science and biology. However, most of published work only studied two-dimensional in-plane vibration and seldom covered vertical vibration modes. In order to study how vertical vibration modulates optical response, we demonstrate a practical method to optically probe three-dimensional vibration modes (both in-plane and vertical modes) using Ultrafast Transient Absorption Spectroscopy.

There are infinite numbers of modes in vibration and each mode exhibits its own modulation on optical spectra. Each optical mode is sensitive to different acoustic vibration mode. Therefore, the shifts of each peak in the spectrum can be used to probe the corresponding acoustic vibration. Let's use Au-SiO<sub>2</sub>-Au multilayer nanostructure with various shapes on glass substrate as an example, such 3-layer structures that form a magnetic dipole resonator is sensitive to the vertical vibration. According to our simulation results (Figure 2a), the right-side peak is expected to show a blue-shift with increased SiO<sub>2</sub> thickness while it experiences a red-shift once feature size enlarges. In this case, vertical vibration and in-plane vibration exhibit opposite optical modulations on spectra that makes it possible to distinguish different modes based on peak shifts. If we define a vector  $A$  and each component in  $A$  represents one vibration mode, its induced optical response  $B$  can be measured. A matrix  $T$  is calculated through the relationship  $B = A * T$ ,  $T$  can be utilized to predict original vibration modes based on resultant optical responses in the future.

Nanostructures with various shapes are fabricated (Figure 1) and first characterized by UV-VIS Spectrometer, from the measured transmission spectra (Figure 2b, 2c, 2d), we observed that the peak positions of experimental results are clearly consistent with calculation. Then, the next step is to achieve transient spectra through transient absorption spectroscopy. The samples are first excited to vibrate by a high-power pump pulse, then a weak probe pulse after a time delay  $\tau$  is sent to measure instant absorption spectra. With ultrahigh time resolution of 150 fs, it is possible to explore transient effects of vibration on optical spectra. Figure 3a shows one transient spectrum we got, if measuring time-dependent absorption spectra at 685 nm and 714 nm wavelength around 700 nm peak (Figure 3b, 3c), a  $\pi$  phase shift between two absorption oscillations is exhibited on the spectra. Right now, we are still working on this part, new data will be presented in the conference.

1. Ahmed, A., et al. (2017). "Understanding how acoustic vibrations modulate the optical response of plasmonic metal nanoparticles." **11**(9): 9360-9369.

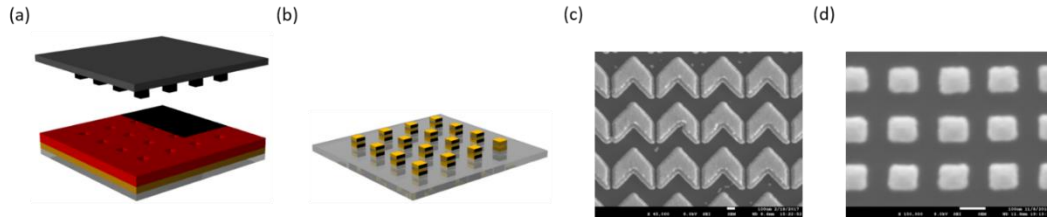


Figure 1. Schematic of fabrication process: (a) One UV-curable resist layer and one lift-off layer on a glass substrate and Nanoimprinting by a pillar array mold. (b) After etching residual layer, E-beam evaporation and lift-off, Au-SiO<sub>2</sub>-Au structure was left on glass substrate. (c) SEM image of chevron shape sample. (d) SEM image of square shape sample.

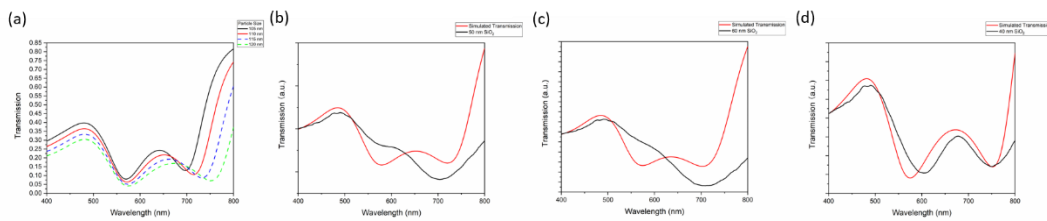


Figure 2. (a) Simulation result of varying size nanostructures. (b) Experimental transmission spectrum of 200 nm pitch, 115 nm feature size, 30 nm Au thickness and 40 nm SiO<sub>2</sub> thickness. (c) Experimental transmission spectrum of 200 nm pitch, 115 nm feature size, 30 nm Au thickness and 50 nm SiO<sub>2</sub> thickness. (d) Experimental transmission spectrum of 200 nm pitch, 115 nm feature size, 30 nm Au thickness and 60 nm SiO<sub>2</sub> thickness.



Figure 3. (a) Broadband transient absorption spectrum (b) Time-dependent absorption spectrum at 685 nm wavelength. (c) Time-dependent absorption spectrum at 714 nm wavelength.