# Ultrafast optical probing of the vibrations of gold nano-dishes

Au ナノディッシュ構造における振動モードの超高速光学測定

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## 1. Introduction

Surface plasmon polaritons (SPP), responsible for dramatic enhancements of electromagnetic fields on nanometer scales, are under intense investigation thanks to the development of metal processing techniques on tiny length scales. Applications of SPPs have been demonstrated in near-field microscopy, surface-enhanced Raman spectroscopy, and nano-photonic circuit design, for example. In particular, miniaturized photonic circuits are expected to allow much faster information processing on smaller length scales. For the practical application of SPPs there is a need for dynamic modulation systems. One possibility for achieving this modulation is the use of acoustic waves at frequencies ~GHz-THz, high enough to respond to requirements of tomorrow's information processing.

In this study, we investigate the GHz acoustic vibrational modes of a plasmonic structure in the form of an array of gold nano-dishes, as shown in **Figure 1**, whose plasmonic characteristics have already been investigated in the past [1, 2, 3, 4]. As this structure can confine both SPPs and phonons at the same time, it works as an effective testbed for SPP-phonon interactions.

## 2. Samples

The gold nano-dish array structures were prepared by a technique involving self-assembled templating and electrochemical deposition [5, 6]. By slowly evaporating a colloidal solution of polystyrene latex spheres onto a glass substrate on which a gold thin film of thickness 360 nm is deposited, a self-assembled hexagonal close-packed structure is formed. After electrochemically depositing gold on the template, polymer spheres

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**Fig. 1** (a) Scanning electron microscope image of a similar dish structure with a 700 nm dish diameter, and (b) schematic image of dish structure used in the present experiment (1600 nm diameter and height 760 nm).

were dissolved completely in toluene. By changing the total charge passed one can control the dish thickness. In this study, we concentrate on the vibrational modes of a sample with a dish diameter of 1600 nm, a dish height of 960 nm and a dish center-to-center separation of 1576 nm.

## 3. Experimental set-up

The experiment is performed with an ultrashort optical technique. A mode-locked Ti:sapphire laser with a wavelength of 820 nm, a repetition rate of 80 MHz and a pulse duration of ~ 200 fs is used as a light source. Pump pulses of wavelength 820 nm are used to excite the vibrations, and delayed probe pulses of wavelength 410 nm generated in a second harmonic generation crystal (SHG) are used for detection of the vibrations. Focusing both beams on the same position on the sample to  $\sim 1 \mu m$  spots, we detect the vibrations as reflectivity changes of the probe light beam. By scanning the sample position with a piezoelectric actuator with nanometer spatial resolution, the two-dimensional spatial distribution of the detected amplitude at different frequencies is mapped to characterize the vibrational modes.

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**Fig. 2** Vibrational spectra of the center of a dish in the dish array obtained by (a) experiment and (b) numerical simulation.

#### 4. Results and discussion

Figure 2 (a) shows the frequency spectrum obtained from the temporal Fourier transform of the experimental reflectivity change detected at the center a dish in the structure, showing a main peak at  $\sim 0.8$  GHz and a few other peaks.

We also performed time-domain finite element simulations of the elastic response using an impulsive force as a source. The vibrational spectrum corresponding to the out-of-plane displacement at the centre of a dish is shown in **Figure 2 (b)**. A main peak at essentially the same frequency as the experimental data is observed. **Figure 3** shows images of simulated vibrational modes at 0.8 GHz and 3.2 GHz. The vibrational mode at 0.8 GHz shows confinement of the phonons in a single dish whereas the mode at 3.2 GHz appears to be a more extended mode of the phononic structure.



Fig. 3 Simulated normalized modulus of the vertical (z -directed) displacement for frequencies 0.8 GHz and 3.2 GHz. The colour scale is given in arbitrary units.

#### 5. Conclusions and future work

In conclusion we have investigated GHz acoustic vibrations in a nano-dish array structure by experiment and by simulation.

A principal phononic resonance at  $\sim 0.8$  GHz was observed for optical excitation in the center of a single dish, corresponding to confined vibrations. This was verified by numerical simulation. In future the wavelength of the probe light will be scanned independent of the pump light in order to pinpoint effects arising from the excitation of surface plasmon polaritons.

#### References

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