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# Design of plasmon fields using nano-fabrication techniques and femtosecond light pulses

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## 1. Introduction

Light harvesting and local field enhancements are the key role of local surface plasmon resonances (LSPRs). Metal nano structures, such as metal nano particles and nano rods, pairs of particles, aggregated particles, and complicated nano patterns engineered by electron-beam lithography, are widely used for this purpose. Tunings of resonance frequencies and lifetimes of LSPR eigenmodes are essential to obtain the maximum efficiency of the field enhancements. Designs and fabrications of plasmonic structures require nanometer-scale precision. Because surface plasmons are confined and possess the maximum intensity at metal-insulator interfaces or at metal-metal gaps, SP fields are sensitively affected by local inhomogeneity and fluctuations.

Design of excitation light field in ultrafast time-domain is also an important aspect to determine the intensity and the spatial distribution of SP fields. Because SPs are coherent oscillations of electron density, they can interfere with external light fields. Utilizations of this feature enable to control time evolutions of SP fields, which can be applied to developments of SP devices and diagnoses of them.

In this paper, we demonstrate fabrications and characterizations of LSPRs formed by arrays of metal nano particles.

## 2. LSPRs of Au nanoparticle chains and the time-resolved autocorrelation

We investigate eigenmodes and coherent features of LSPRs excited on Au nanoparticle chains by microscopic scattering spectroscopy and femtosecond time-resolved autocorrelation. Nanoparticle (NP) chains are fabricated on an ITO-coated glass substrate by using a nano-manipulation method: NPs are aligned with nano-scale precision by a piezo motor driven micro-tip. Fig.1 shows chains consists of three NPs (100nm in diameter): Gap distances between neighboring NPs are zero for the structure A and 10~20 nm for the structure B. The plasma resonance of a *single* NP show a peak at 590 nm, which tends to split when other NPs are placed within <100 nm gap. Such plasmon hybridization effect can be seen in the structure B, which shows two peaks at 574 nm and 640 nm. The hybridization effect induces much larger spectral change when neighboring NPs make contact: The structure A shows completely separated two peaks at 590 nm and 813 nm.

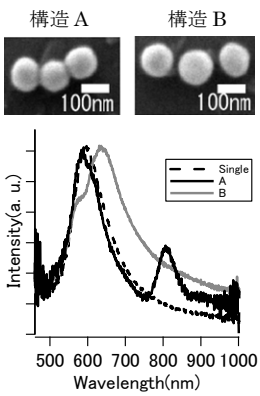


Fig.1 Structure and scattering spectrum of Au nano-particle chains.

Interestingly, the lifetime of the mode appear at 813 nm (11 fs) is 4-times longer than that of a Mie plasmon of single Au NP of which resonance locates at the near infrared region. The elongation of lifetime is caused by a suppression of radiation dumping. As a result of coupling between three NPs, the newly generated eigenmode at 813 nm possesses a feature of a dark mode.

We also take an autocorrelation of plasmon scattering from the structure A and B using a 9 fs ultrafast laser as the excitation source (Fig.2). The carrier wavelength of the laser is 820nm. Therefore thus the mode at 813 nm (structure A) is resonantly excited, while the mode 640 nm (sture B) is off-resonantly excited. The difference in the eigen frequency induces phase shifts of plasmon oscillations which can be seen in >6 fs delay times.

The oscillation phase shift can be applied to a control of plasmon intensities. The structure A shows stronger intensity than the structure B for the micrograph taken at the delay $\tau$ = 8.5 fs, while it is inversed for $\tau$ = 10.0 fs.

## 3. Conclusions

Two-types of Au nanoparticle arrays are investigated by microscopic scattering spectroscopy and femtosecond time-resolved scattering autocorrelation. Only a ~10 nm difference in gap distances between neighboring NPs induces drastic change in the plasmon eigenmodes, which can be attributed to the change of the coupling intensity. The difference in eigen frequencies of plasmon modes induces oscillation phase shift in the autocorrelation, which can be applied to control of plasmon excitation by external light fields.

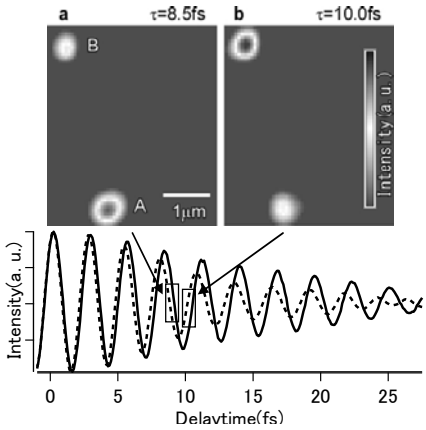


Fig. 2 Plasma scattering autocorrelation of two plasmonic structures.