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Plasmon Coupling and Coherent Acoustic Phonon Dynamics of Periodic Gold Pair Nanocuboids by Near-IR Laser Spectroscopy

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Introduction Surface plasmon resonance (SPR) bands of assembled metal nanostructures are shifted by near- and far-field interaction of dipole couplings, as compared with the same sized single. The assembled metal nanoarrays prepared by nano-lithographic mehods are helpful to provide the quantitative information between the SPR shift and the configuration [1]. In the present study, we prepared 2D gold nanocuboid arrays with electron beam lithography (EBL) method and examined the effect of plasmon coupling on the coherent acoustic phonon with near-IR transient absorption (TA) spectroscopy. The TA experimental results were discussed on the basis of finite-different time-domain (FDTD) simulation.

Experiments Two types of gold pair nanocuboid (size: 150 × 150 × 20 nm^3) arrays were prepared by the EBL method. One was designed for near-field coupling effect with the constant separation and seven different nanogaps of 0, 1.8, 3.5, 5.3, 7.1, 8.8 and 10.6 nm. For the first type, two groups were included and named as S400nm and S600nm according to their constant separations. The other was designed for far-field interaction effect with the constant nanogap and seven different separations from 400 nm to 700 nm with 50-nm increasing interval. For the second type, three groups were included and named as G0nm, G5nm and G10nm according to their constant nanogaps. The scheme is shown in the inset of Fig. 1 and ± 45 º represents the polarization direction of the detection pulses.

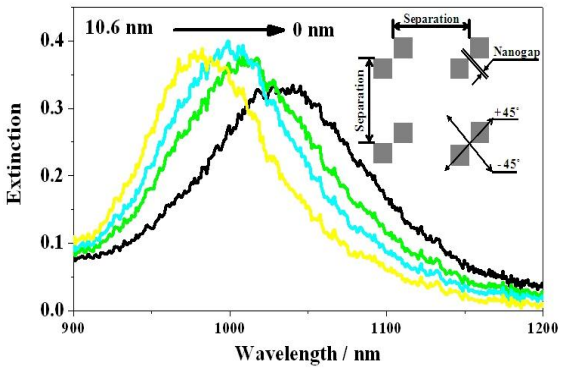

Fig. 1. SPR bands of the gold nanocuboid arrays with various nanogaps from 10.6 nm to 0 nm and the representative scheme of the arrays in the inset.

Results and discussion The SPR bands in Fig. 1 show a remarkable red-shift from 975 nm to 1050 nm with the decrease of the nanogap from 10.6 nm to 0 nm. With ultrafast excitation, coherent acoustic phonon vibration combined with thermal relaxation can be detected, which follows after electron-electron scattering and electron-phonon coupling processes. The

coherent acoustic phonon vibration presents a bleaching oscillation around the SPR band. In Fig. 2 the oscillation of bleaching peaks of the gold nanocuboid arrays shows a similar behavior irrespective of the nanogap from 10.6 nm to 0 nm. This kind of oscillation of bleaching peaks for all the arrays were fitted with three-component damped cosine functions and the three periods. Two symmetric breathing modes of coherent acoustic phonon were mainly detected for the gold nanocuboids with the displacements from edge to edge and from tip to tip. The corresponding oscillation periods for the two modes are around 84 and 114 ps which are very similar with the theoretical values 93 and 131 ps ($T = 2d / c_{\mathrm{L}}$, d is the length and c_{L} is the longitudinal sound velocity in the metal nanostructures). The third oscillation period (46 ps) was assigned to the second harmonic of the edge-to-edge breathing modes. Fig. 3 presents the oscillation amplitudes of the bleaching peaks with the variation of the nanogaps and an exponential relationship was observed. A similar relationship between the spectral shift and the structural deformation of the pair was observed by the simulation with FDTD method as shown in the inset, which was assigned to the nanogap sensitivity for the same structural deformation.

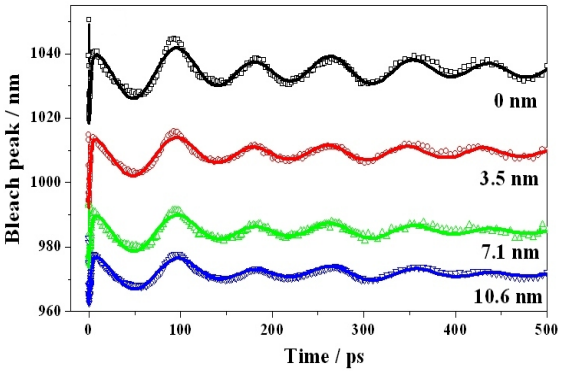

Fig. 2. The oscillation of bleaching peaks for the corresponding arrays with the decrease of the nanogap from 10.6 nm to 0 nm.

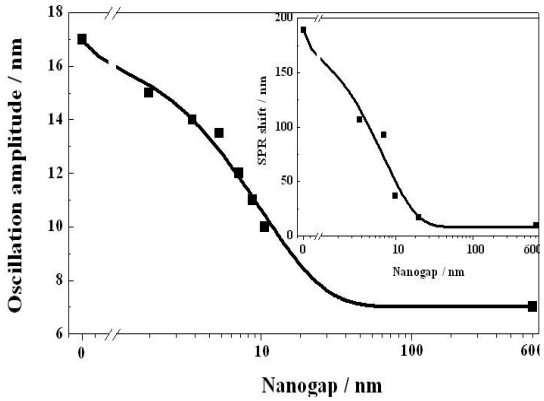

Fig. 3. The oscillation amplitudes of the bleaching peaks for the gold nanocuboid arrays S600nm. The SPR shift corresponding to ± 1 nm size change on the tips and edge centers by FDTD simulation in the inset.

The TA experimental results revealed that the oscillation periods of coherent acoustic phonon vibration were not influenced by the different nanogaps and separations, even though SPR bands were shifted due to the near- and far-field dipole couplings. Moreover, the oscillation amplitude of coherent acoustic phonon vibration was exponentially decayed with the increase of nanogap while the damping was not much influenced.

[1]. L. Wang, Y. Nishijima, K. Ueno, H. Misawa, N. Tamai, Appl. Phys. Lett. 95 (2009) 053116.