Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Impact of thermal annealing and laser treatment on the morphology and optical responses of mono- and bi-metallic plasmonic honeycomb lattice

Yi-Ju Chen,[a] Gabriele Schmidl,[a] Andrea Dellith [a], Annett Gawlik [a], Guobin Jia [a], Thomas Bocklitz,[a, b, c] Xiaofei Wu,[a] Jonathan Plentz,[a] and Jer-Shing Huang [a, b, d, e, *]

[a] Leibniz Institute of Photonic Technology, Member of Leibniz Health Technologies, Member of the Leibniz Centre for Photonics in Infection Research (LPI), Albert-Einstein Straße 9, 07745 Jena, Germany
[b] Institute of Physical Chemistry and Abbe Center of Photonics, Friedrich Schiller University Jena, Member of the Leibniz Centre for Photonics in Infection Research (LPI), Helmholtzweg 4, 07743 Jena, Germany
[c] Institute of Computer Science, Faculty of Mathematics, Physics & Computer Science, University Bayreuth, Universitätsstraße 30, 95447 Bayreuth, Germany
[d] Research Center for Applied Sciences, Academia Sinica, 128 Sec. 2, Academia Road, Nankang District, Taipei 11529, Taiwan
[e] Department of Electrophysics, National Yang Ming Chiao Tung University, No. 1001, Daxue Road, East District, Hsinchu 30010, Taiwan

*Email: Jer-Shing.Huang@leibniz-ipht.de

Content

S1: Zoom-in images of Ag and Pd samples prepared by thermal annealing and laser treatment

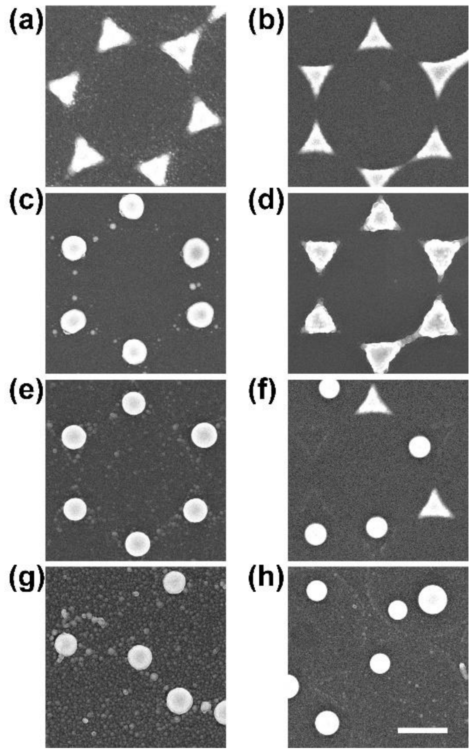

Fig. S1 Zoom-in images of (a) Ag and (b) Pd pristine samples, (c) Ag and (d) Pd particles after thermal annealing, (e) Ag and (f) Pd particles after laser treatment with a single 100 mJ cm^{-2} pulse, and (g) Ag particles treated with a single 200 mJ cm^{-2} pulse, and (h) Pd particles treated with five 100 mJ cm^{-2} pulses. The scale bar is 200 nm and applies to all images.

S2: Investigating the effect of the pulse number and laser fluence on Ag-Pd

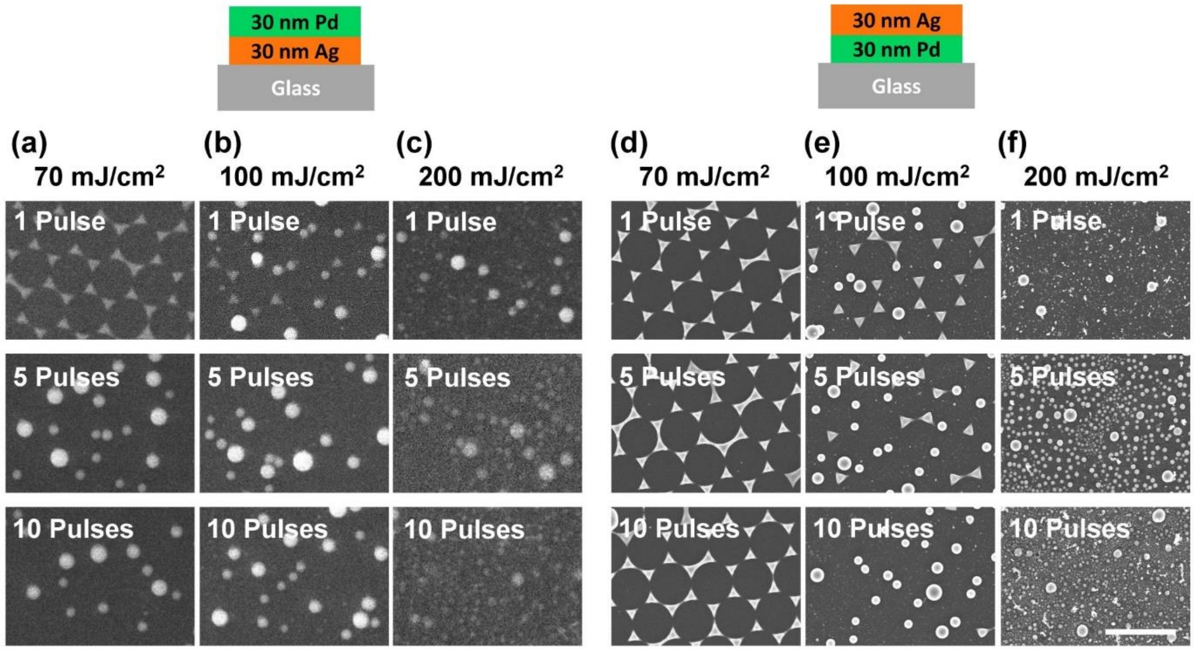

Fig. S2 SEM images of (a) Pd/Ag/Glass (b) Ag/Pd/Glass honeycomb-packed array treated by 1, 5, 10 pulses with 70 mJ cm^{-2}, 100 mJ cm^{-2}, and 200 mJ cm^{-2}, respectively. The scale bar is 1 μm and applied to all images.

In Fig. S2, we investigated the effect of pulse number and laser fluence on both Pd/Ag/Glass and Ag/Pd/Glass samples. We used pulse numbers of 1, 5, and 10 with fluences of 70, 100, and 200 mJ cm^{-2}. To generate ideal spheres, a high pulse number or laser fluence is necessary. The threshold number of the pulse for morphology transition is related to the interface material. For example, when we applied 5 pulses with a fluence of 70 mJ cm^{-2} on the Pd/Ag/Glass sample, spherical particles with a random distribution are generated (shown in Fig. S2a, 5 pulses). On the other hand, when we used Pd as the interface material, the particles remained intact, showing sharp triangular islands in a honeycomb lattice even after applying up to 10 pulses with a fluence of 70 mJ cm^{-2}, indicating that Pd has better adhesion on the substrate (Fig. S2d, 10 pulses).

The influence of the interface material can also be observed under other laser treatment conditions. Almost every triangular island on Pd/Ag/Glass becomes spherical and randomly distributed after applying five 100 mJ cm^{-2} pulses (Fig. S2b, 5 pulses), while many triangular islands on Ag/Pd/Glass still remain intact under this condition (Fig. S2e, 5 pulses). This suggests that the wettability of the interface material influences the morphology of the nanoparticles. When using a material with better wettability, a higher fluence or number of pulses is required to produce spheres. However, when the applied energy exceeds a certain threshold, e.g., 200 mJ cm^{-2}, the particles are ablated, and the lattice arrangement is disrupted (Fig. S2c and S2f, one pulse with 200 mJ cm^{-2}).

S3: The coupling feature of SLR in Pd system

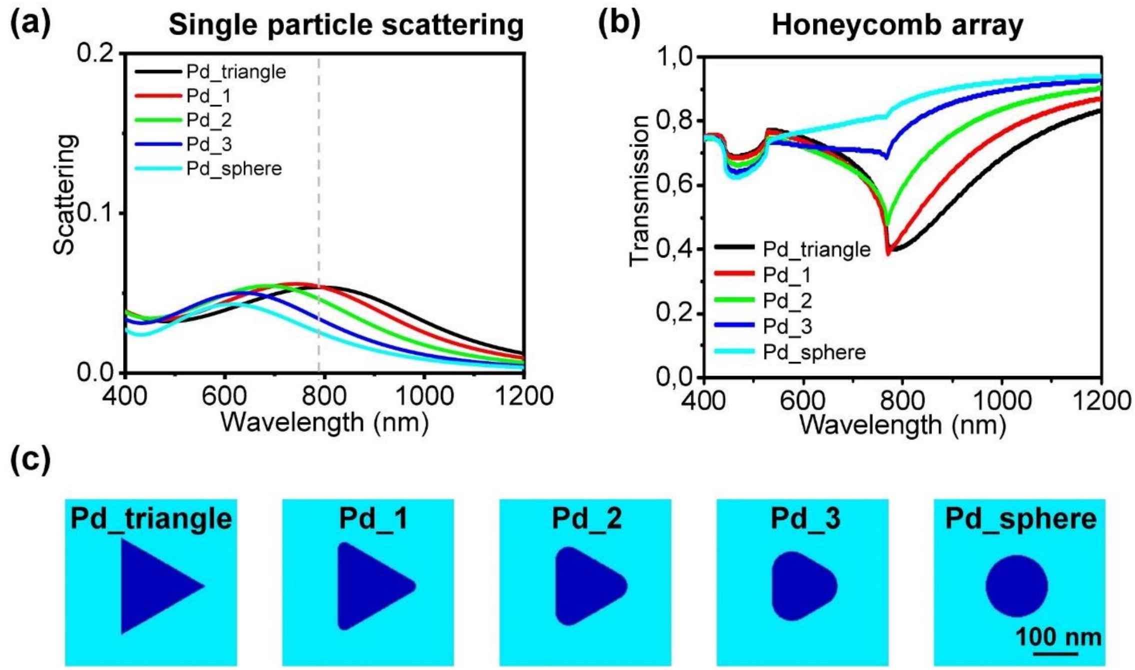

Fig. S3 (a) Simulated single particle scattering spectra, where the gray dashed line indicates the theoretical RA resonance and (b) simulated transmittance spectra of the honeycomb Pd array with various particle morphologies. (c) Schematics of the particle units employed in the simulation. The scale bar is 100 nm and applied to all images in Fig. S3c.

To understand how particle morphology affects the characteristics of surface lattice resonances (SLRs), we simulated the scattering spectra of individual particles with different shapes and the transmission spectra of their arrays. The height of each particle was calculated with reference to the volume of the pristine sample (Pd_triangle). As we know, SLRs arise from the coupling of Rayleigh anomaly (RA) and localized surface plasmon resonance (LSPR). The theoretical RA is located at 791 nm, as indicated by the gray dashed line in Fig. S3a. Since the LSPR of the Pd triangle is close to the RA, it results in a strong coupling, which manifests as a shallow dip in the transmission spectrum of the array. In contrast, the LSPR and RA of the Pd sphere are beyond the coupling range, resulting in no significant resonance feature in the transmission spectrum.

S4: Particle analysis

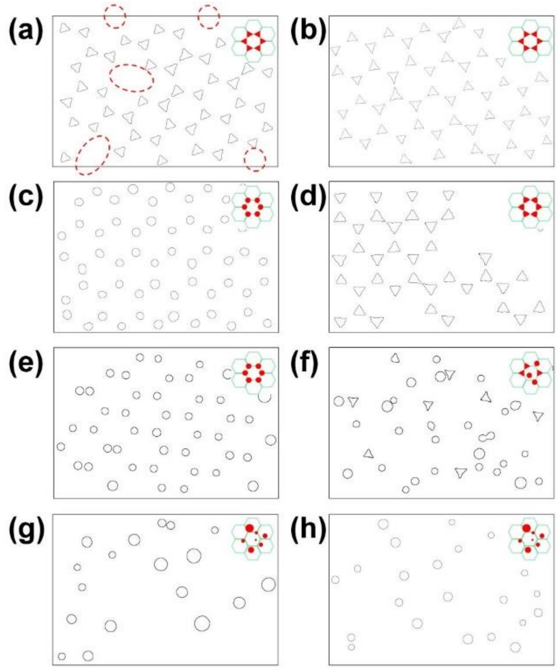

Fig. S4 The outline of the analyzed area. (a) Ag (b) Pd particles before annealing, (c) Ag (d) Pd particles after annealing, (e) Ag (f) Pd samples after laser treatment with a single 100 mJ cm^{-2} pulse, (g) Ag particles treated with a single 200 mJ cm^{-2} pulse, and (h) Pd particles treated with five 100 mJ cm^{-2} pulses.

Our particle analysis is performed using ImageJ, which is a free software designed for image processing. We utilized secondary electron images with sharp particle edges for our analysis. The steps we followed for particle analysis are outlined in the user guide provided by ImageJ. (https://imagej.nih.gov/ij/docs/guide/146-30.html#toc-Subsection-30.2).

In Fig. S4, we display the contours of the analyzed particles. To calculate the size distribution and circularity of the particles, we excluded particles that were cut by the image boundary or stuck together. For example, in Fig. S4a, the excluded particles are marked in red circles.

S5: Ag and Pd honeycomb-packed array treated by 3 pulses with different fluences

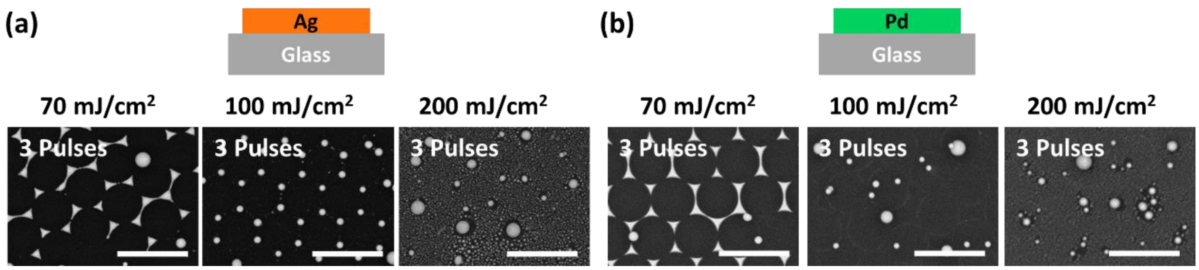

Fig. S5 The SEM images of (a) Ag and (b) Pd honeycomb-packed array treated by 3 pulses with 70, 100, 200 mJ cm^{-2}. The scale bars in all the images are 1 μm.

S6: Measured transmittance spectrum

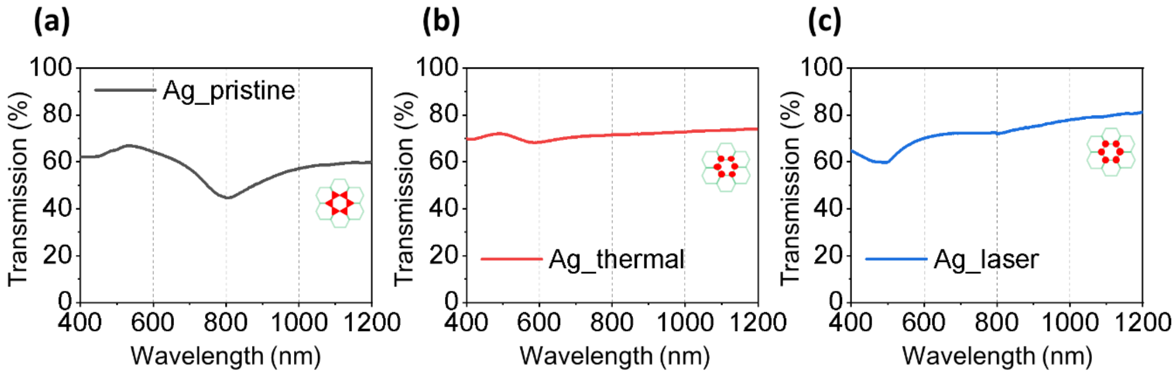

Fig. S6 Measured transmittance spectrum of the (a) pristine (black), (b) thermal-annealed (red), and (c) laser-treated (blue) honeycomb-packed Ag array. The schematics on the right side of each spectrum indicate the morphology and spatial arrangement of the corresponding sample.