Scaling-Up Nano-Plasmon Catalysis: The Role of Heat Dissipation

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DAMAGE CAUSED BY IRRADIATION

We confirmed by SEM (Figure S1) that the degradation of the Raman spectra presented in Figure 3 of the manuscript were caused by damages to the nanoparticle substrate. As the high total power of the excitation laser beam could not dissipate quickly enough, a partial melting of the particles occurred.

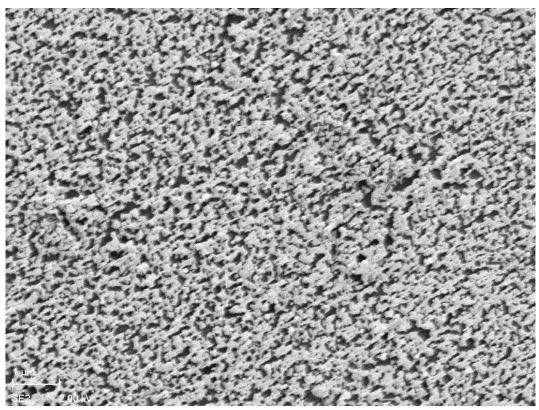

FIG. S1. **SEM 175 µm spot:** 8 µm × 5.5 µm SEM image of from the center of the irradiated spot with a diameter of 175 µm. The damage due to the melting of the nanotriangles is clearly visible.

X-RAY REFLECTIVITY OF GOLD NANO TRIANGLES

X-ray reflectivity (XRR) is a technique that allows the surface-sensitive characterization of thin layers by interference of specular reflections of the bottom and top interfaces of the layer that give rise to characteristic minima and maxima as function of the energy transfer, $Q = 4/\lambda \ \sin\vartheta$, with ϑ being the incidence angle of the X-rays with wavelength λ onto the sample. We measured a monolayer of gold nano triangles (NTs) on a Silicon substrate at the XPP-KMC-3 endstation at the synchrotron BESSY II. The incidence angle of the X-rays on the sample was scanned between 0 and 3° incidence angle, which corresponds to $Q = 0$ to $4\,\mathrm{nm}^{-1}$ in reciprocal space. The corresponding dataset shown in the inset of Fig. 1a) contains a dominant oscillation frequency that corresponds to a wavelength of $\lambda = 0.9\,\mathrm{nm}^{-1}$ determined by a Fourier-transformation. This wavelength corresponds to a layer-thickness of $7 \pm 2\,\mathrm{nm}$. The absence of higher frequencies in the signal proofs that our sample contains a negligible part of stacked gold-nano-triangles over the X-ray spot size of approximately $0.5 \times 10\,\mathrm{mm}^2$.

X-RAY DIFFRACTION (XRD)

We measured at the XPP-KMC-3 endstation at the synchrotron source BESSY II the 222 Bragg reflection of the [111] oriented gold NTs at the X-ray energy of 8 keV. We selected the 2nd order Bragg reflection to minimize the X-ray footprint. The sample was mounted on Peltier-cooled sample mount, which results in a stable substrate temperature of 15°C that was attached with silver paint to the Peltier cooler. In Figure S2a) and b) we show the Bragg peaks of the gold NTs on Si and glass substrates for different Laser intensities, respectively. For every measured laser intensity, a new spot on the sample was selected and measured before illumination (black thick curves) and during illumination. For every measurement we waited 60 s for thermal equilibration between gold NTs.

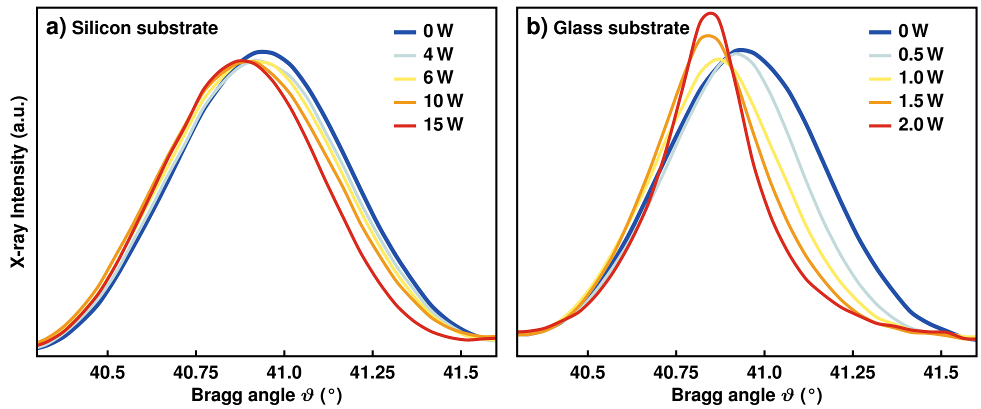

FIG. S2. **XRD results:** Bragg peak shift of gold NTs on a) a silicon substrate and b) glass substrate for different intensities. The Bragg peak shift to smaller angles due to heating of the gold NTs. On the glass substrate a sharpened peak is observed for high intensities.

We note a peak sharpening at high intensities. For very thin layer we can neglect typical Bragg broadening effects and the width of the Bragg peak is approximately given by the thickness. In our case the peak width is slightly smaller as theoretical values for 7 nm layers, which indicates a small amount of stacked gold NTs (5-10%). After illumination of the sample with glass substrate with high intensities we observe a sharpened Bragg peak that is related to thicker nano particles. This indicates a particle coalescence of the gold NTs.

DETERMINATION OF THE LINEAR EXPANSION COEFFICIENT

The temperature-dependent shift of the Bragg peak allows us to calculate the linear expansion coefficient for gold NTs along the [111] direction. It is measured with a closed-cycle cryostat setup available at the XPP-KMC-3 endstation at the synchrotron source BESSY II that allows the variation of the sample temperature from 20 to 350 K. From the Bragg Peak position the out-of-plane lattice constant in [111] direction is extracted and shown in Figure S3. A linear fit yields the linear expansion coefficient of $\alpha = 1.49 \cdot 10^{-5}$ 1/K.

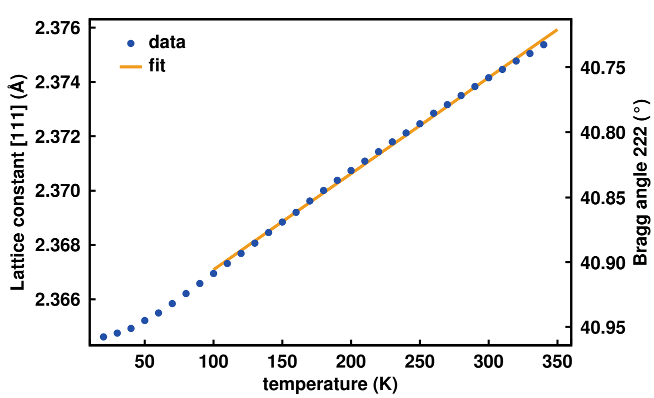

FIG. S3. **The Temperature-dependent lattice constant** is measured from 20 to 350 K and fitted to extract the linear expansion coefficient α.