Supplementary Information

Surface Plasmon Damping Quantified with an Electron Nanoprobe

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Calculating the Q-factor

Several methods are available to relate the Q-factor to the resonance's angular frequency and damping parameter. The partial, 2nd order differential equation of a driven, damped harmonic oscillation can for example be solved using Green's functions techniques,[S1] or by considering the mean stored energy in the oscillation.[S2] Based on the latter, it can be insightful to consider the oscillation at maximum kinetic energy, i.e. zero potential energy. By taking the first derivative of Eq. (2), it can easily be seen that the quality factor, defined as the energy stored in an oscillator, divided by the energy dissipated per radian, can be described as

$$Q = \frac{E}{\frac{1}{\omega_1}\frac{dE}{dt}} = \frac{\frac{1}{2} m e^{-2\gamma t} A^2 \omega_1^2}{\frac{1}{\omega_1}\gamma m e^{-2\gamma t} A^2 \omega_1^2} = \frac{\omega_1}{2\gamma} \approx \frac{\omega_p}{2\gamma}. \quad \text{(S1)}$$

Multiplying ω_p and γ with the reduced Planck constant, $\hbar$, allows the direct calculation of the quality factor from the plasmon energy and peak width:

$$Q \approx \frac{E_p}{\Gamma}. \quad \text{(S2)}$$

Spectral broadening effect

Figure S1 shows the effect of peak broadening due to the non-zero instrument function. The Q-factor is plotted *vs*. the plasmon resonance energy, from the same set of experimental data as Fig. 3. Here, the raw, unprocessed data is compared with the processed data where Eq. (5) was used to remove the effect of spectral broadening.

The Γ_G was measured to be varying between 50 and 70 meV, depending on the electron monochromator set-up used for each given experiment. The grey diamonds show the distribution of Q, as calculated directly from the raw data. The correct Lorentzian widths Γ_L were obtained by applying equation (5), after which equation (3) was used to calculate Q for the purple circles. It can be seen that without removing the broadening effect of the finite EELS energy resolution, the quality factor would be underestimated—particularly at low energies—by about 10 to 30% in our case. This broadening effect would be more severe for EELS spectra with lower monochromaticity. The presented results in this work always show Q and T_2 without instrumental broadening.

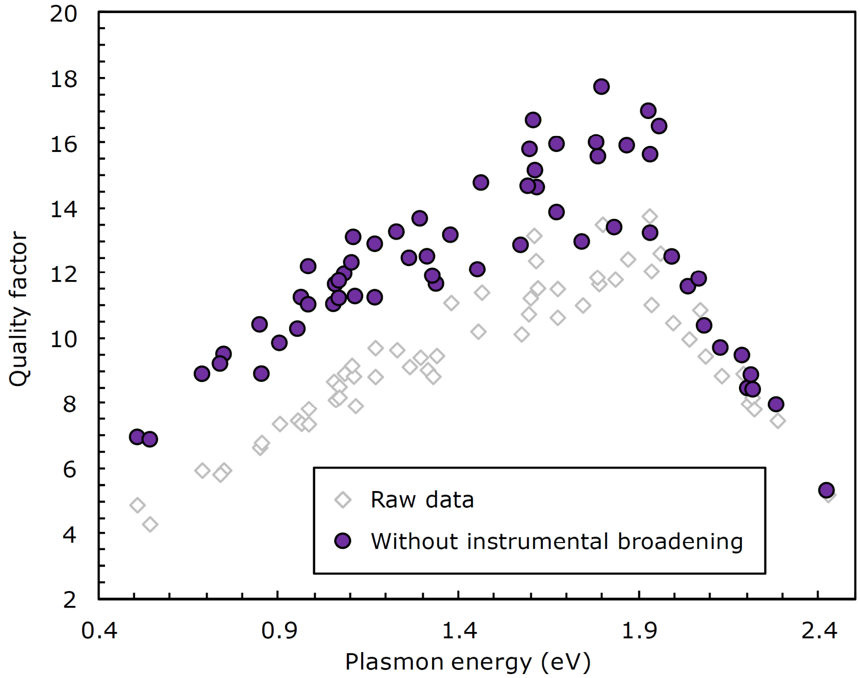

Figure S1 | The plasmon quality factor as a function of resonance energy, measured from individual gold nanoparticles. The grey diamonds show the Q-factor calculated with Eq. (3) from the raw, monochromated EELS data. The purple circles plot the Q-factor after removal of the instrumental broadening, using Eq. (5).

EELS *vs*. Optical spectra

Figure S2 below shows experimental results comparing EELS spectra from individual gold nanocrosses with the optical absorption spectrum from a suspension of the same particles. The EELS spectra were taken from the tips of the nanocrosses, to ensure that the main plasmon mode would be optically excitable. It can be seen that plasmon resonances of individual particles take place in narrow energy intervals. However, the overall optical response of the nanocross suspension shows broadband absorption. This is the result of the summation of many absorption spectra of differently-sized individual particles, averaging the overall response to a broad absorption band. A similar effect was observed earlier for nanoporous gold.[S3]

The somewhat suppressed intensity around 2.4 eV for the EELS spectra is the result of the ‘transverse’ plasmon resonances,[S4] which take place over the whole particle surface, and therefore absorb light relatively strongly. The EELS measurements on the other hand, were only taken at the tips of the gold nanocrosses, explaining the relative dominance of the 'longitudinal' plasmon modes at lower energy.[S4]

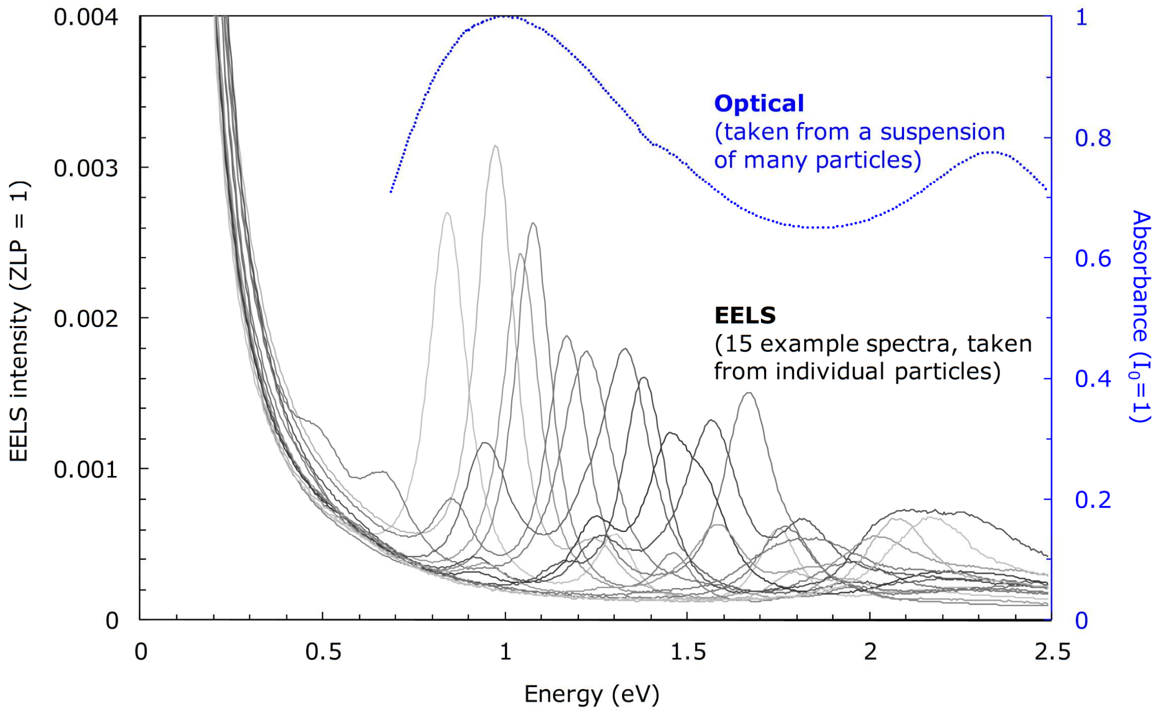

Figure S2 | Light absorption *vs.* EELS. The blue dotted line shows the experimental, normalized light absorption spectrum of a suspension of gold nanocrosses, plotted to the right vertical axis. From the same particle suspension, a TEM sample was made and example EELS spectra were taken from 15 individual particles, plotted here in grey to the left vertical axis. The EELS spectra show the raw data, including the 'zero-loss peak' background signal that is most prominent at low energies. All EELS spectra were normalized at the zero-loss peak maximum.

REFERENCES

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