Supporting Information for the paper

Electron and Lattice Heating Contributions to the Transient Optical Response of a Single Plasmonic Nano-Object

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Contents of the Supporting Information

1) Optical measurements and analyses performed on a single gold ND supported on a sapphire substrate (referred to as ND2 in this document) distinct from that considered throughout the main text (figures S1 to S5).

2) Experimental investigation of pump fluence effect on the transient optical response of single gold NDs (Figure S6).

3) Dependence of electron heating-induced gold dielectric function changes $\Delta \epsilon_e$ on electronic temperature rise ΔT_e (Figure S7).

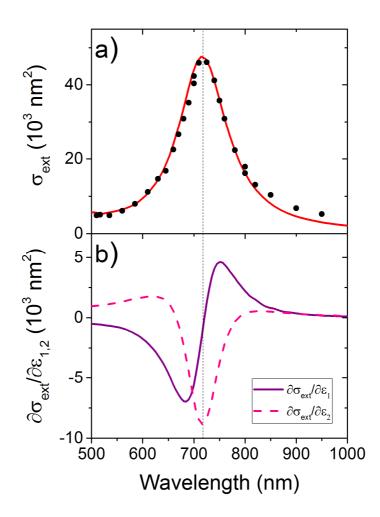


Figure S1. Linear optical properties of ND2. (a) SMS-measured absolute extinction spectrum of ND2 (black dots). It is well reproduced by FEM calculations performed with *D*=101 nm, *h*=21 nm ND diameter and thickness (red line). (b) $\partial \sigma_{ext}/\partial \epsilon_1$ (purple) and $\partial \sigma_{ext}/\partial \epsilon_2$ (pink) derivatives deduced from the FEM calculations. The vertical dashed line indicates the SPR central wavelength λ_{SPR} =718 nm.

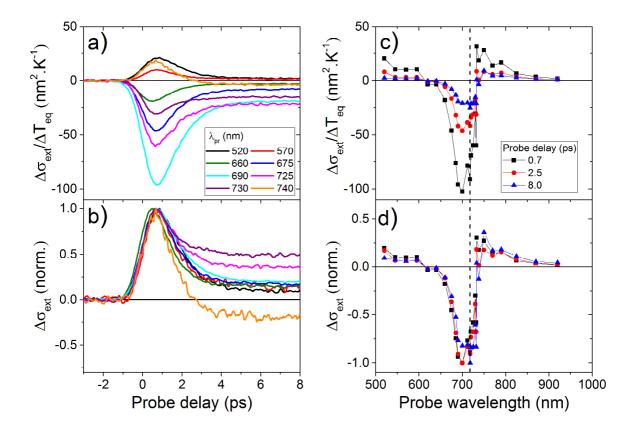


Figure S2. Experimental time-resolved signals. (a) Examples of time-resolved signals measured on ND2 using different probe wavelengths (indicated in the legend), divided by the estimated equilibrium ND temperature rise ΔT_{eq} . (b) Same signals as in (a), normalized to unitary maximal value. (c) Transient extinction spectra deduced from time-resolved signals at probe delays of 0.7, 2.5 and 8.0 ps. (d) Same spectra as in (c), normalized to -1 near 700 nm. The vertical dashed line in panels (c) and (d) indicates the central position of the SPR of ND2.

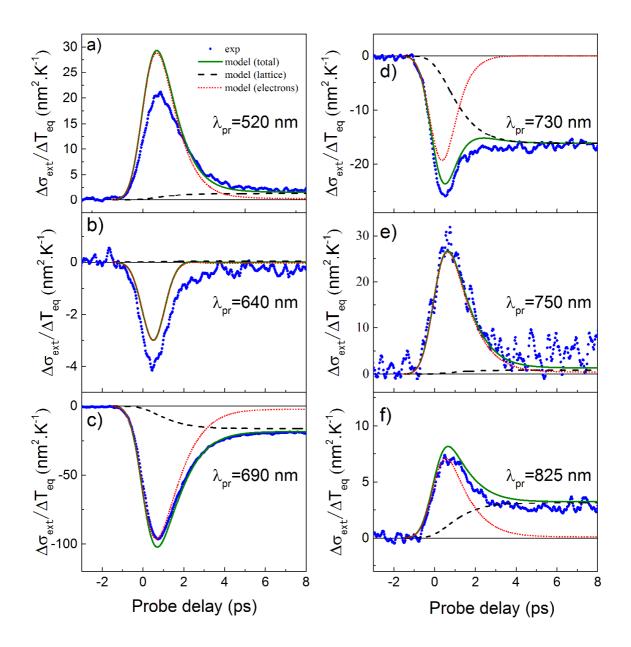


Figure S3. Measured and simulated time-resolved signals at specific wavelengths. The transient extinction changes measured on ND2 using various probe wavelengths λ_{pr} (indicated on each panel) are shown by blue dots. The transient extinction changes simulated using the complete model described in the main text (with ΔT_{eq} =9.5 K, the average equilibrium temperature rise of ND2 in these experiments) are shown by green lines (λ_{pr} values of 525, 640, 690, 725, 765 and 835 nm were used for the simulations shown in panels a-f). These simulated signals were obtained by summing the contributions of electron and lattice heating, which are shown as dotted red and dashed black lines, respectively.

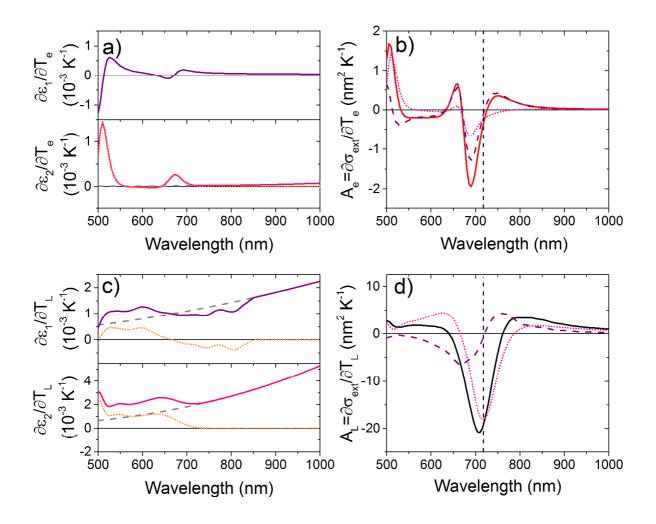


Figure S4. Dependence of the gold dielectric function and ND2 extinction cross-section on electronic and lattice temperatures. (a) $\partial \epsilon_1 / \partial T_e$ (top, purple line) and $\partial \epsilon_2 / \partial T_e$ (bottom, pink line) derivatives (same as Figure 4a). (b) Deduced $A_e = \partial \sigma_{ext} / \partial T_e$ derivative. The contributions of ϵ_1 and ϵ_2 changes to A_e are shown as dashed blue and dotted green lines, respectively. (c) $\partial \epsilon_1 / \partial T_L$ (top, purple line) and $\partial \epsilon_2 / \partial T_L$ (bottom, pink line) derivatives (same as Figure 4c). (b) Deduced $\partial \sigma_{ext} / \partial T_L$ derivative (same color codes as in (b)). The vertical dashed line in panels (c) and (d) indicates the central position of the SPR of ND2.

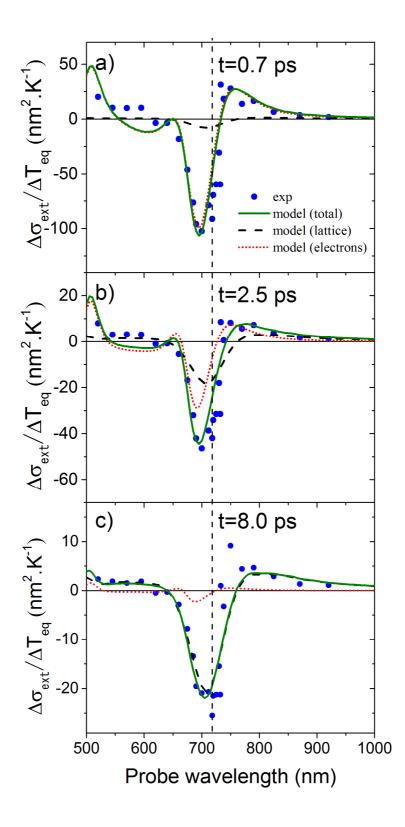


Figure S5. Measured and simulated transient extinction spectra at specific delays. The transient extinction spectra measured on ND2 for pump-probe delays of (a) 0.7 ps, (b) 2.5 ps and (c) 8.0 ps are shown by blue dots. In each case, the transient extinction spectra simulated using the complete model described in the main text are shown by green lines, with the contributions of electron and lattice heating shown as dotted red and dashed black lines, respectively. The vertical dashed line indicates the central position of ND2 SPR.

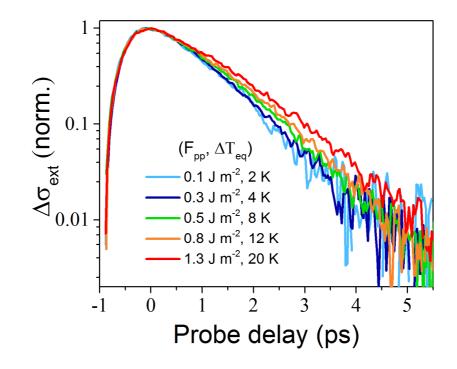


Figure S6. Effect of pump fluence on time- resolved signals. The time-resolved signals shown in this figure were measured on a single Au ND (with dimensions close to those of ND2) using pump and probe wavelengths of λ_{pp} =410 nm and λ_{pr} =700 nm and various pump fluences F_{pp} (each corresponding to a distinct equilibrium temperature rise ΔT_{eq}), indicated in the caption. All signals were normalized to have a common maximal value of 1. The increase of the decay time at high fluences is expected and ascribed to an increased electron-phonon energy transfer time due to the T_e-dependence of the electronic heat capacity. In contrast, the rise time of the signals, which for the λ_{pr} used here approximately reflects the progressive injection of energy in the ND by the pump pulse (Figure 1b), is almost insensitive to pump fluence.

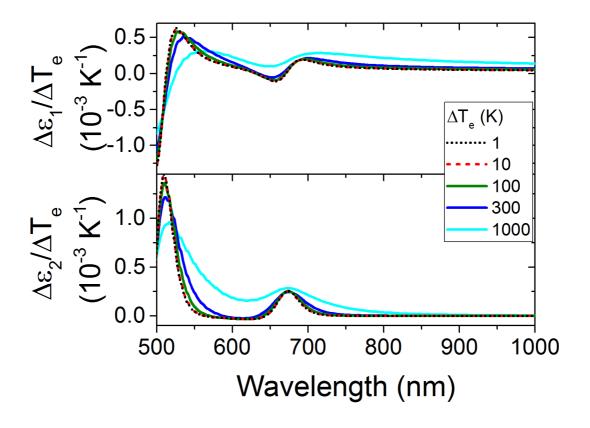


Figure S7. Changes of gold complex dielectric function induced by electronic heating. The real (top) and imaginary (bottom) components of $\Delta\epsilon$ computed for $\Delta T_L=0$ and ΔT_e values indicated in the inset are shown after division by ΔT_e . This representation allows to appreciate the linear $\Delta\epsilon$ (ΔT_e) variation in the low ΔT_e regime (in which $\Delta\epsilon_{1,2}/\Delta T_e$ plots correspond to those of the $\partial\epsilon_{1,2}/\partial T_e$ derivatives shown in Figures 5 and S4) and the non-linear one when ΔT_e reaches hundreds of K.