Nanoscale

ARTICLE

Electronic Supplementary Information

Ultrafast transient optical loss dynamics in exciton-plasmon nano-assemblies

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Section 1. Locating the extinction maximum of NPs in GS_AuL

To locate the extinction of the NPs only in the hybrid system **GS_AuL**, we removed the QDs extinction by subtracting their properly scaled extinction from that of **GS_AuL**, see fig S1. To perform this operation, we choose a particular wavelength where there is little variation in the difference between the extinction of **GS** and **GS_AuL**. For our calculations we picked 470 nm, where the absorption curves of **GS** and **GS_AuL** are nearly parallel, i.e. they differ by a constant. This means that there is a very small contribution from the NPs to the total extinction in **GS_AuL**. We then subtract the optical density of **AuL** at this location from that of **GS_AuL**, and divide the result by the optical density of **GS**. By doing this we receive a scale factor that we use to scale down the extinction of **GS** so that it can be directly subtracted from **GS_AuL**. After performing the subtraction, we find that the new peak is located at ≈ 590 nm approximately 40 nm away from the measured extinction of the hybrid system.

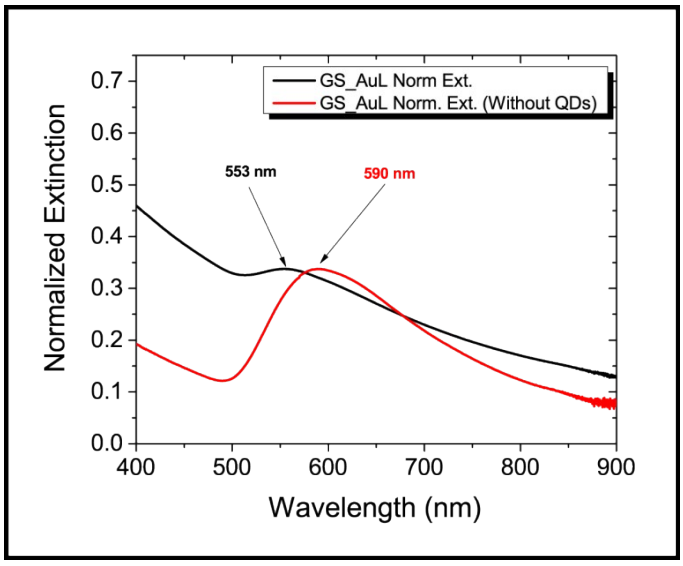

Figure S1: Normalized extinction of **GS_AuL** before and after excluding the extinction of the incorporated QDs. The extinction of Au NPs in **GS_AuL** is found to be located at ≈ 590 nm.

Section 2. UTAS results for AuH

As we mentioned earlier, **AuH** is the same as **AuL** but with higher NP concentration. Figure S2a shows that **AuH** has essentially the same transient absorption behavior exhibited by **AuL** where we see a bleach that blue shifts over time and two uneven absorption wings where the lower energy absorption wing is below the noise level. However, when we compare **AuL** and **AuH** (see Figure S2b) we see that the magnitude of the transient absorption/bleach is significantly higher for **AuH** than for **AuL** which is expected given that it has higher NP concentration.

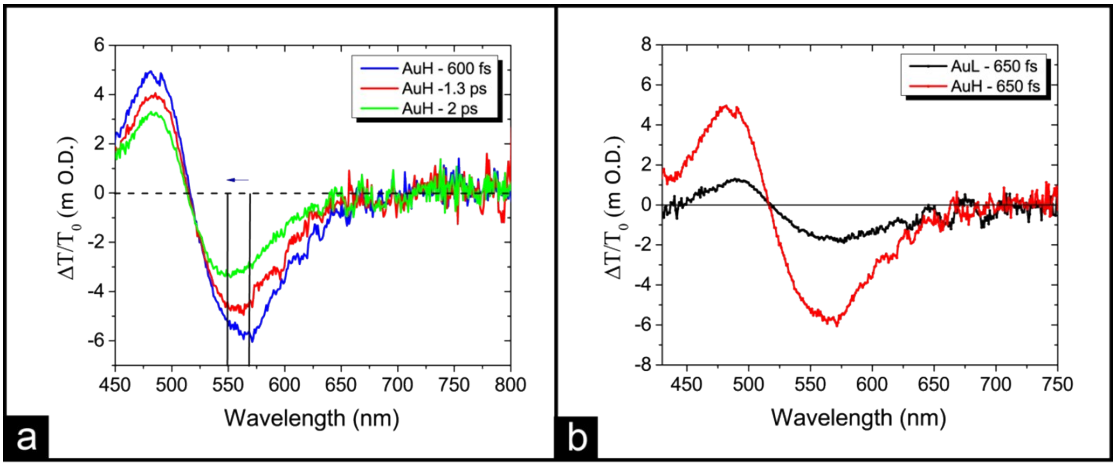

Figure S2: (a) Shows the TA results for **AuH** for three delay times; 600 fs, 1.3 ps, and 2 ps. Just like **AuL** the transient absorption signal is a bleach with two uneven absorption wings at higher and lower energies with respect to the bleach. The bleach maxima also blue shifts as the delay time increases. Compared to **AuL** we see in (b) that the TA signal is larger in magnitude.

Section 3. UTAS results for GS_AuH

GS_AuH has the same gain concentration as **GS_AuL** but with higher NP concentration. Normalized extinction cross section of GS_AuH is shown in Figure S3a. Generally, it exhibits the same behavior as **GS_AuL** (Figure S3b). When we compare **GS_AuH** to **AuH**, we also see loss mitigation signature very clearly (Figure S3c). By comparing the results of **GS_AuH** for 800 nm vs. 400 nm, we can see that without exciting the gain, loss mitigation does not take place (Figure S3d). This is similar to the results of **GS_AuL** which is presented in Figure 3b in the manuscript. Moreover, **GS_AuH** shows a stronger signal compared to **GS_AuL** consistent with the fact that it has higher NP concentration. These results provide extra evidence on the existence of loss mitigation in the hybrid system. Finally, it is clear in Figure S3e that the lower energy absorption wing exists and appears, as it rises above the noise level as we discussed in the manuscript.

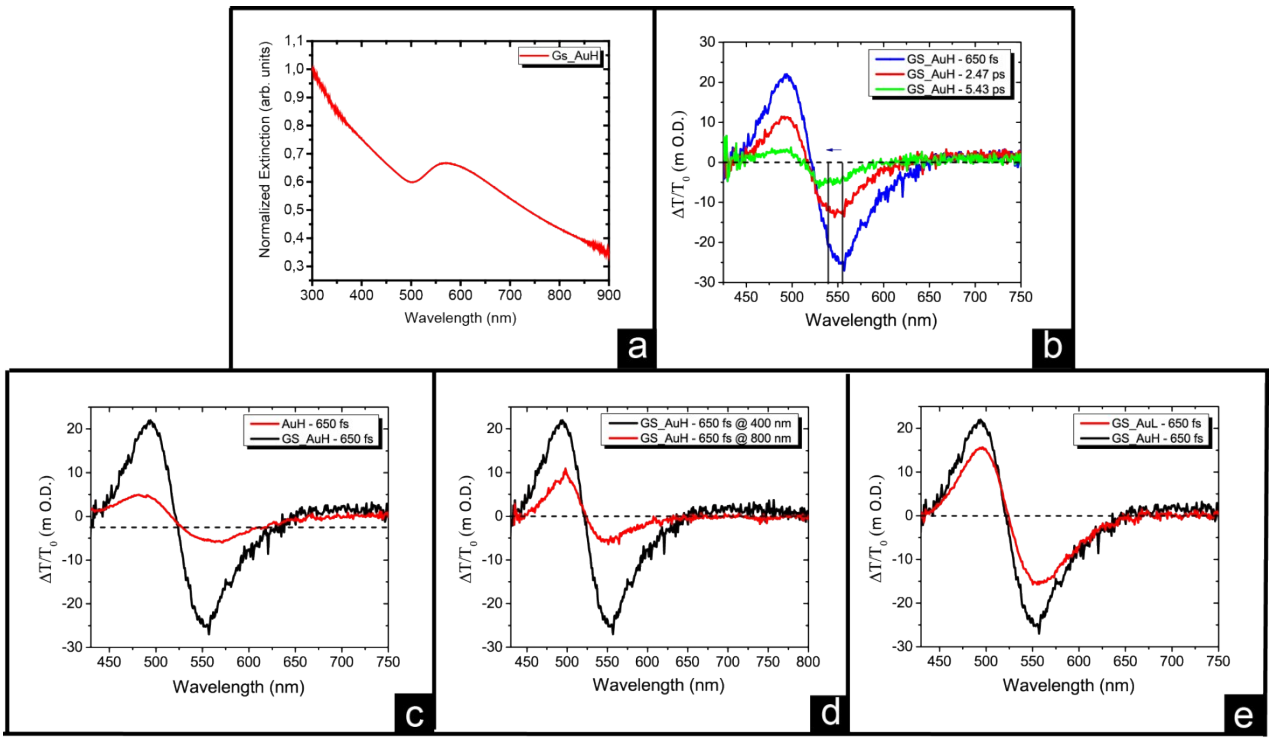

Figure S3: (a) Normalized extinction cross section of GS_AuH hybrid system. (b) The TA of **GS_AuH** for three delay times; 650 fs, 2.47 ps, and 5.43 ps shows similar behavior to that of GS-AuL. Compared to **AuH**, that has the same NP concentration, we see in (c) that the TA spectrum is significantly larger in magnitude due to loss mitigation. This effect does not take place upon pumping **GS_AuH** with 800 nm as shown in (d). (e) shows a comparison between the UTAS results for **GS_AuL** and **GS_AuH**.

Section 4. Maximum bleach wavelength vs. pump energy:

Figure S4a shows the normalized transient bleach for **AuL** for pump energies 1 ?j, 2 ?j, and 6 ?j for a given delay time. There is no shift as a function of pump power for **AuL** unlike the case for **GS_AuL** shown in Figure S4b.

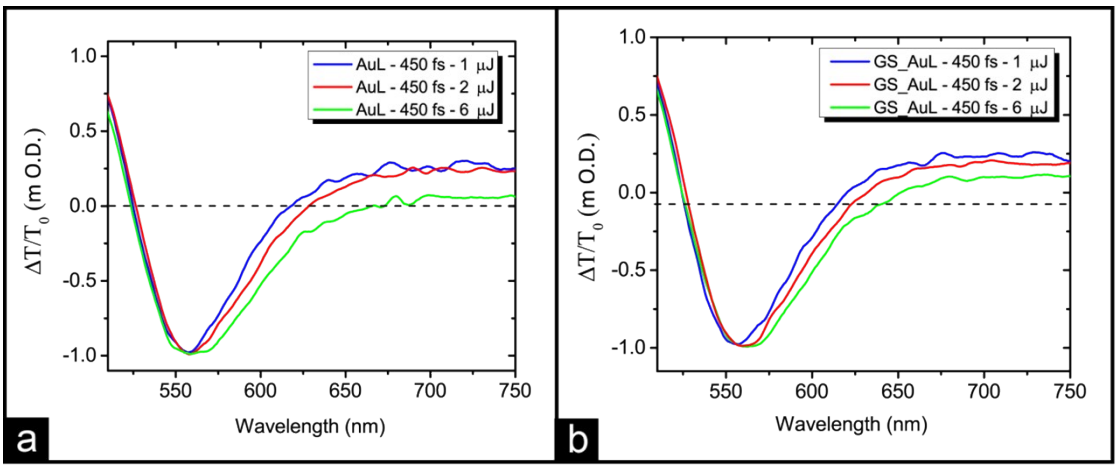

Figure S4: (a) Normalized TA of **AuL** for three pump powers showing no shift of the bleach maxima. On the other hand, in (b) the normalized differential absorption of **GS_AuL** shows a red-shift to its bleach maxima as we increase the pump power towards the emission maximum of the gain.