

1 Supplementary Information (SI)

2 **Table S1: Number of particles measured with ns-LAAPTOF with different pulse energies at focus**
 3 **positions F1 and F2.**

| Particle types | Diameter (d_p) nm | Number of spectra obtained for three different laser energies | | | | | |
|---------------------------------|-----------------------|---|-------|-------|-------|-------|-------|
| | | 0.8 mJ | | 4 mJ | | 8 mJ | |
| | | at F1 | at F2 | at F1 | at F2 | at F1 | at F2 |
| PSL500 | 500 | 500 | 500 | 500 | 500 | 500 | 500 |
| PSL800 | 800 | 500 | 500 | 500 | 500 | 500 | 500 |
| PSL1000 | 1000 | 500 | 500 | 500 | 500 | 500 | 500 |
| Silica | 1100 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 |
| NaCl | 400 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 |
| NH ₄ NO ₃ | 400 | 500 | 500 | 500 | 500 | 500 | 500 |
| Au-Ag | 600 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 |
| Au-SiO ₂ | 400 | 1000 | 1000 | 1000 | 1000 | 1000 | 1000 |
| Au-PAH | 400 | 500 | 1000 | 500 | 1000 | 500 | 1000 |

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5 **Table S2: Number of particles measured with fs-LAAPTOF with different pulse energies at focus**
 6 **positions F1 and F2.**

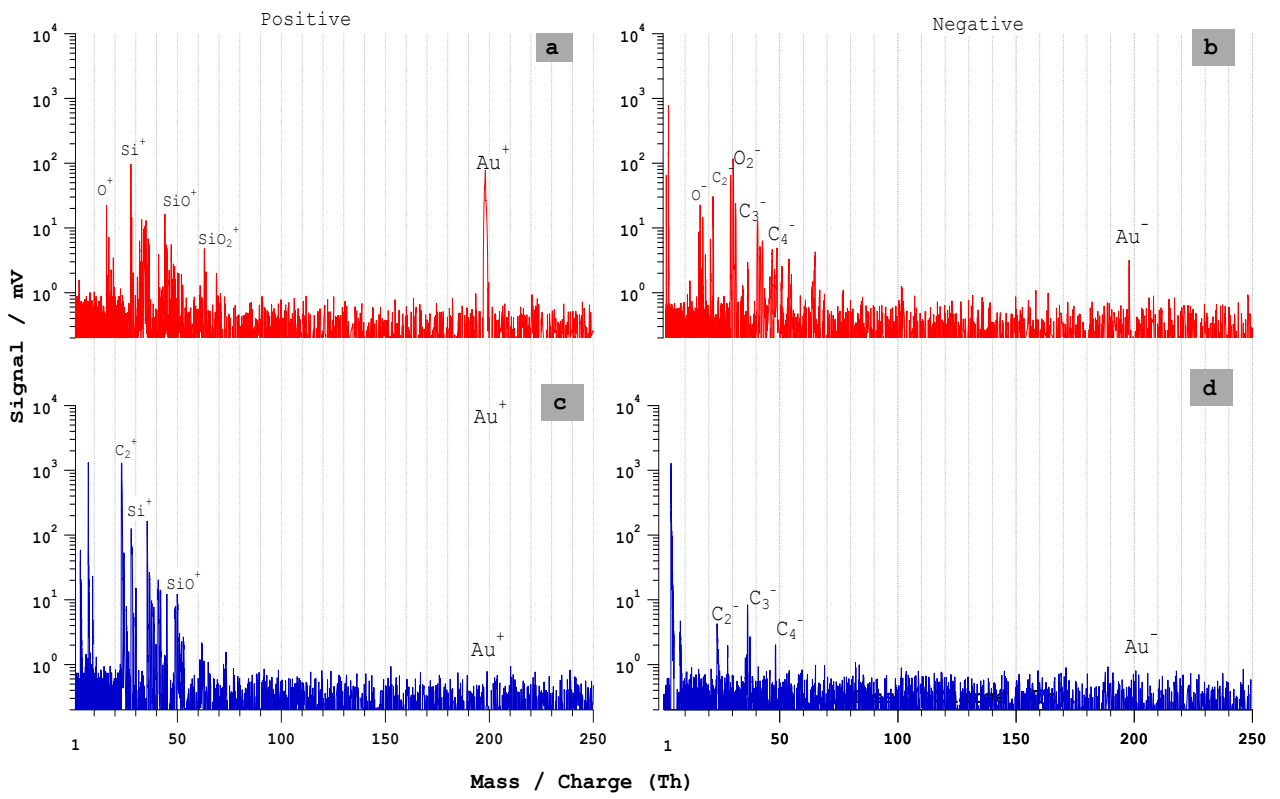
| Particle types | Number of spectra obtained at 266 nm | | Number of spectra obtained at 800 nm for four different laser energies | | | | | | | | | |
|---------------------------------|--------------------------------------|------|--|-------|--------|-------|--------|-------|--------|-------|--------|-------|
| | | | 0.2 mJ | | 0.3 mJ | | 1.7 mJ | | 3.2 mJ | | 3.5 mJ | |
| | | | at F1 | at F2 | at F1 | at F2 | at F1 | at F2 | at F1 | at F2 | at F1 | at F2 |
| PSL500 | 500 | 750 | 10 | 50 | 500 | 500 | 1000 | 260 | 160 | 320 | | |
| PSL800 | 500 | NA | 10 | 30 | 1000 | 200 | 1000 | 600 | 120 | 500 | | |
| PSL1000 | 500 | NA | 10 | 20 | 300 | 500 | 130 | 310 | 1500 | 310 | | |
| Silica | 1000 | 450 | 300 | 120 | 1000 | 270 | 1000 | 430 | 1000 | 560 | | |
| NaCl | 1000 | 500 | 1000 | 1000 | 1000 | 1000 | 80 | 1000 | 500 | 400 | | |
| NH ₄ NO ₃ | 600 | 1000 | 60 | 50 | 500 | 380 | 1000 | 350 | 500 | 500 | | |
| Au-Ag | 1300 | 1000 | 140 | 10 | 1000 | 500 | 1500 | 500 | 500 | 250 | | |
| Au-Silica | 600 | 300 | 1000 | 10 | 500 | 200 | 650 | 580 | 350 | 240 | | |
| Au-PAH | 300 | 1000 | 20 | 10 | 1000 | 500 | 500 | 500 | 400 | 600 | | |

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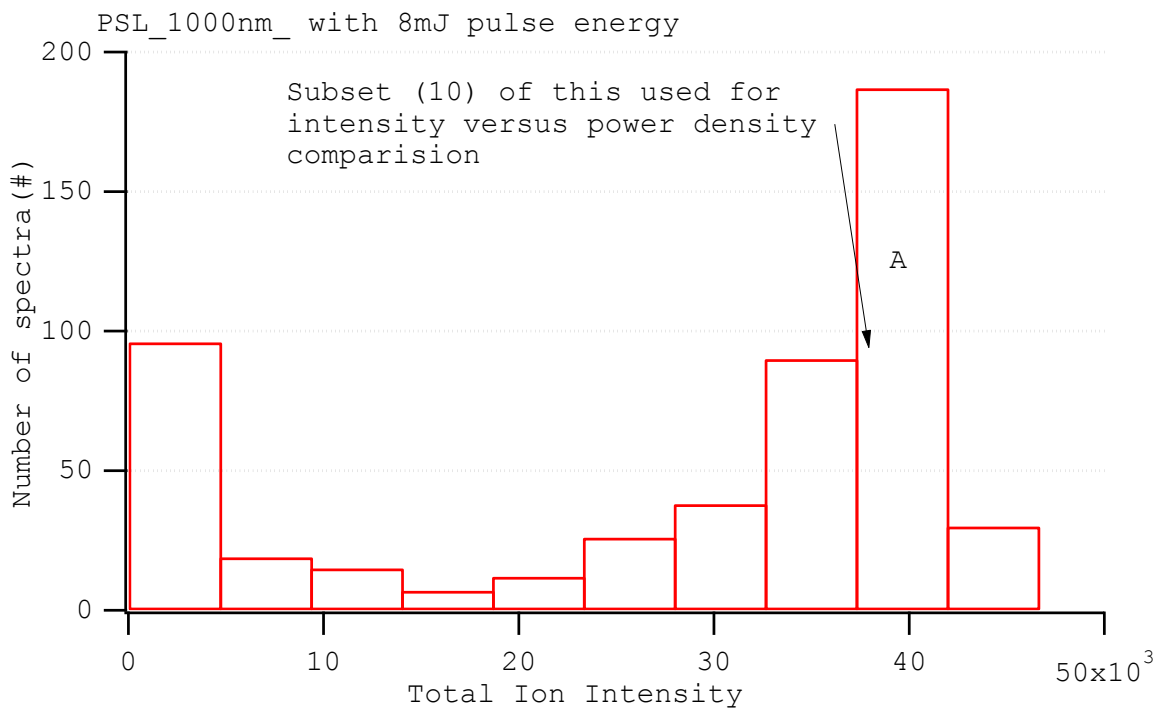
10 Gold-SiO₂ core-shell particles



11 **Figure S1: Mass spectra of gold-SiO₂ core-shell particles (a) & (b) with nanosecond laser, (c) & (d) with**
12 **femtosecond laser.**

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14 **Variability of the total ion intensity for constant conditions collecting 500 mass spectra.**



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16 **Figure S2: Distribution of the number of spectra obtained for 1000 nm PSL particles with the ns-laser at 8**
17 **mJ energy for different total ion (both positive and negative) intensity. The best 10 spectra were selected**
18 **from section A shown in the plot.**

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