## Supplementary Material:

## Optical Response of Quantum-Sized Ag and Au Clusters — Cage vs. Compact Structures and the Remarkable Insensitivity to Compression

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Figure S1: Absorption spectra of the cages and the pure compact clusters. The curves are the same as in Fig. 2 of the article, but here Au and Ag are shown on the same scale in order to highlight the qualitative differences between the quantum-sized clusters of the two materials.



Figure S2: Absorption spectra of the cages and the compact clusters. The results are the same as in Fig. 2 of the article, but here we plot the absorption cross section normalized to the number of atoms in each structure. Clearly, the absorption *per atom* is stronger in the silver cage structure than in the core-shell cluster  $Au_{55}Ag_{92}$ , although the spectra are otherwise very similar. Likewise, the  $Au_{92}$  shell shows a much stronger absorption per atom in the VIS region than both the compact cluster and the core-shell structure with an outermost Au shell.



Figure S3: Time-dependent dipole moments of the silver-based structures. After the perturbation at t = 0, the systems evolve freely. The evolution time to obtain the spectra is 25 fs, indicated by the dashed vertical line. The snapshots of the density difference with respect to the ground-state density shown in Fig. 4 of the article are taken at instants of high polarization as indicated by the crosses. (The inset shows a blow-up of the region in question.) For comparison, two snapshots of the compact Ag<sub>147</sub>, taken at times of zero overall polarization, indicated by the stars, are also taken. The snapshots are shown in Fig. S4.



Figure S4: Two snapshots of the time-dependent density of the compact  $Ag_{147}$  cluster, taken at times of zero overall dipole moment as indicated by the stars in Fig. S3. There is no strong uniform polarization (unlike at the times of maximum dipole moment shown in Fig. 4 of the article) but a finer pattern of minor modes that add up to zero.