



Optical extinction and scattering cross sections of plasmonic nanoparticle dimers in aqueous suspension

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Absolute extinction and scattering cross sections for gold nanoparticle dimers were determined experimentally using a chemometric approach involving singular-value decomposition of the extinction and scattering spectra of slowly aggregating gold nanospheres in aqueous suspension. Quantitative spectroscopic data on plasmonic nanoparticle assemblies in liquid suspension are rare, in particular for particles larger than 40 nm, and in this work we demonstrate how such data can be obtained directly from the aggregating suspension. Our method can analyse, non invasively, the evolution of several sub-populations of nanoparticle assemblies. It may be applied to other self-assembling nanoparticle systems with an evolving optical response. The colloidal systems studied here are based on 20, 50 and 80 nm gold nanospheres in aqueous solutions containing sodium lipoate. In these systems, the reversible dimerisation process can be controlled using pH and ionic strength, and this control is rationalised in terms of DLVO theory. The dimers were identified in suspension by their translational and rotational diffusion through scattering correlation spectroscopy. Moreover, their gigadalton molecular weight was measured using electrospray charge-detection mass spectrometry, demonstrating that mass spectrometry can be used to study nanoparticles assemblies of very high molecular mass. The extinction and scattering cross sections calculated in the discrete-dipole approximation (DDA) agree very well with those obtained experimentally using our approach.

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