Quantitative Extinction and Coherent Nonlinear Four-Wave Mixing Microspectroscopy of Single Gold Nanoparticles

L. Payne, G. Zoriniants, F. Masia, W. Langbein and P. Borri

Cardiff University School of Biosciences, Museum Avenue, Cardiff CF10 3AX, UK; borrip@cf.ac.uk

Cardiff University School of Physics and Astronomy, The Parade, Cardiff CF24 3AA

Quantifying and eventually controlling the optical properties of small (<50nm) metallic nanoparticles (NPs) at the single particle level is key to many recently proposed applications ranging from sub-wavelength optical devices, catalysis and photovoltaics to biomedical sensing and optical “nanoscopy” including correlative light/electron microscopy.

Here, firstly we show a simple, rapid, and quantitative wide-field technique to measure the optical extinction $\sigma_{\text{ext}}$ and scattering $\sigma_{\text{sca}}$ cross-section of single Au NPs using wide-field microscopy which enables the simultaneous acquisition of hundreds of NPs for statistical analysis [1] (see Fig. 1a). We will report on improving the sensitivity limit down to 10nm$^2$, allowing to measure single 5nm Au NPs. Furthermore, we have investigated the nonlinear four-wave mixing (FWM) response at the surface plasmon resonance (SPR) in these NPs with a high-resolution multiphoton microscope coupled to a phase-sensitive heterodyne detection scheme with which we resolve single NPs and detect the **ultrfast changes of the real and imaginary part of their dielectric function** [2] (see Fig. 1b). Beyond fundamental interest, this phase-resolved single particle detection provides an intrinsic ratiometric readout which has the potential to bring unprecedented sensitivity in SPR-based sensing applications, for example using NP dimers as “plasmon rulers”. Work is on-going to fabricate Au NP dimers with controlled interparticle distances and results will be presented.

**Fig. 1** a): Optical extinction image (grayscale 0-1%), and cross section $\sigma_{\text{ext}}$ of single Au NPs using the wide field method in [1]. b): Transient changes of the real and imaginary parts of the dielectric function of a single 20-nm Au NP. Dashed lines are corresponding calculations based on the electron temperature cooling model in [2].