SCANLESS BACKGROUND-FREE 3D NANOMETRIC LOCALISATION OF PLASMONIC NANOPARTICLES USING OPTICAL VORTICES

George Zoriniants,[1] Francesco Masia,[2] Naya Giannakopoulou,[1] Wolfgang Langbein,[2] and Paola Borri,[1]

[1]Cardiff University School of Biosciences, Cardiff CF10 3AX, United Kingdom

[2]Cardiff University School of Physics and Astronomy, Cardiff CF24 3AA, United Kingdom

Single particle tracking using optical microscopy is a powerful technique with many applications in biology and material sciences. Despite significant advances, localising objects with nanometric position accuracy in a scattering environment remains challenging. Applied methods to achieve contrast are dominantly fluorescence based, with fundamental limits in the emitted photon fluxes arising from the excited-state lifetime as well as photobleaching. Furthermore, every localisation method reported to date requires signal acquisition from multiple spatial points, with consequent speed limitations. This work reports a conceptually new way of localising a single nano-object in a scattering environment with position accuracy at the nanoscale in three-dimensions at high speed, using the vectorial nature of tightly focused light (see Figure, top). The method exploits the strong optical absorption and scattering resonance of a single gold nanoparticle, which is detected in a nonlinear way using a sequence of short optical pulses generating resonant four-wave mixing (FWM). This provides a highly sensitive and specific particle detection which does not rely on fluorescence readout, is completely background-free even in highly scattering environments, and outperforms existing methods such as reflectometry, interferometric scattering microscopy, differential interference contrast, and photothermal imaging. By exploiting the optical vortex field pattern in the focus of a high numerical aperture objective, we demonstrate conceptually and experimentally, a position localization accuracy of better than **20nm in plane** and **1nm axially** (an impressive value for axial localisation), from scanless acquisition in the **1ms** time scale (Figure, bottom). The technique is also uniquely sensitive to the particle shape and orientation; hence this method paves the way towards a new form of single particle tracking, where not only the particle position, but also its asymmetry and orientation are detected, revealing unprecedented information about the particle complex dynamics (e.g. hindered rotation) while moving and interacting within a disordered 3D environment.

Figure **Top**: Field distribution in amplitude and phase in the focal plane of a 1.45 NA microscope objective, for an input field circularly polarised. A_+, A_-, and A_z are the co-circular, cross-circular and longitudinally polarised amplitudes, Φ_+, Φ_- and Φ_z the corresponding phases. **Bottom**: Time traces of the retrieved in plane (X) and axial (Z) particle position coordinates from the measured FWM amplitude and phase (symbols) compared with the coordinates recorded from the scanning piezoelectric sample stage (lines), for a single 25nm radius gold NP freely rotating while tightly caged in an agarose gel pocket.