

Ultrafast Phototransient Holographic Imaging and Spectroscopy

by

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Abstract

Transient absorption and photothermal microscopy are widely used for studying processes such as carrier diffusion or the optical response of nanoscopic objects^[1-2]. Beyond spectroscopy, these techniques allow label-free detection down to the single molecule limit^[3]. Unfortunately, essentially all experimental implementations require raster-scanning the sample as they crucially rely on lock-in detection. As a result, these techniques are ill-suited for observing large-areas or dynamically evolving systems such as biological or clinical samples.

I will discuss our solution to this problem: ultrafast holographic transient (UHT) microscopy^[4,5], a new methodology that eliminates all major limitations of photothermal imaging. We implemented a lock-in like widefield imaging camera that allows signal demodulation at arbitrary fast frequencies, irrespective of the camera's exposure time: essentially an all-optical lock-in amplifier with $>10^6$ detectors (pixels).

I will present a detailed description of the experimental implementation followed by several proof-ofconcept experiments explaining both fundamental holographic concepts^[6,7] as well as the implementation of UHT.

I will show ultrafast spectrotemporally resolved measurements on gold nanoparticles where we simultaneously captured the temporal response of >100 particles with single particle sensitivity. I will then extend these concepts towards freely diffusing particles, within large volumes-of-observation, where we performed time-resolved spectroscopic measurements while, simultaneously, 3D tracking all particles. Finally, I will discuss our efforts to combine UHT microscopy with quantitative live-cell 3D imaging.

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- [3] A. Gaiduk, M. Yorulmaz, P.V. Ruijgrok and M.Orrit; Science 330, 353–356 (2010)
- [4] M. Liebel, F.V.A. Camargo, G. Cerullo and N.F. van Hulst; Nano Lett. 21, 1666–1671 (2021)
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