Label-Free Nanoscopy with Infra Red: Breaking Away from the Diffraction Barrier

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Nanoscale and sub-diffraction resolution have been extensively demonstrated in far-field fluorescence microscopy, with reversible optical fluorescence transition (RESOLFT), non-linear structured illumination, and single molecule localization microscopies. However, these methods generally require the introduction of fluorescent markers in the samples and do not provide label-free local chemical information. Novel chemical imaging tools with high resolution are thus sought for. Sub-diffraction vibrational microscopies based on far-field Coherent Anti-Stokes Raman Spectroscopy have been proposed [1,2]. For these schemes, it is envisaged to control the spatial extension of the measured sample volume by suppressing the vibrational Raman coherence through an incoherent process involving the excitation of a neighboring vibration [1] or by periodically depleting the ground-state through Rabi oscillations [2].

Here we demonstrate a model framework for Infra Red Absorption Spectroscopy imaging with spatial resolution beyond the diffraction limit of Infra-Red radiation. This is achieved by confining the effective point-spread function (PSF) by generating a spatial contrast in the depletion of the ground-state of a vibrational mode of interest using a scheme of IR pumping and IR probing, which results in a PSF proportional to the local IR absorbance. The concept is illustrated by simulating PSF and images of patterned thin films of octadecyltrichlorosilane adsorbed on an IR transparent dielectric substrate (Figure).

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References:

Figure 1 (a) PSF for IR absorption nanoscopy for various pump energies. The IR absorption microscopy PSF is also shown (longest dash) for comparison. (b) Representation of an array of nine domains of octadecyltrichlorosilane (200×200 nm²). CH₂ stretch IR image (3.5 µm) computed with a pixel of 50×50 nm² and a NA of 0.85. (c) CH₂ stretch VD-IR image computed for a pump energy of 0.1 µJ. Scale bar is 0.5 µm in all images.