Femtosecond Fieldoscopy a New Technique for High Resolution Spectroscopy and Imaging

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Vibrational spectroscopy allows for non-perturbative, label-free identification of complex molecular composition and offers intrinsic chemical selectivity due to the specific vibrational frequency of different molecules. Among various label-free imaging techniques, stimulated Raman scattering (SRS) stands out, due to its low water absorption cross-section, high penetration depth, high spatial resolution, and linear proportionality with the molecular concentration in the sample under scrutiny. In SRS, two excitation pulses known as pump and stokes excite molecular vibrations coherently, leading to energy loss at pump frequency, and energy gain at Stokes frequency. For complex samples consisting of an ensemble of molecules at different resonance frequencies, broadband, femtosecond stokes, or pump pulses can simultaneously excite the entire molecular fingerprint. Broadband detection requires multichannel lock-in amplifiers or interferometric reference measurements [1]. In this work, we present a new approach for sensitive broadband detection of SRS signals, by taking advantage of the recent progress in ambient air, near-petahertz field-resolved detection techniques [2]. Here, broadband stokes pulses are employed to excite the Raman resonances in a liquid sample impulsively, while the presence of narrowband Raman pump pulses ensures the detection selectivity. Afterward, a near-single cycle probe pulse is employed to resolve the electric field of the stokes gain with attosecond precision [3]. Modulating the Raman pump at half of the laser repetition rates allows for background-free, broadband field-resolved detection of the Raman gain, obviating any need for reference measurements. This approach allows for the simultaneous detection of the entire SRS fingerprint of a sample, acquiring information on both phase and amplitude. Moreover, we present the potential of the technique for high-resolution label-free microscopy.

References

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