High-speed simultaneous Raman-fluorescence spectrometer

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Raman scattering and fluorescence spectroscopies measure phenomena that are often well-separated from each other energetically, such as molecular vibration for the former and electronic excitation for the latter. Despite this, the inelastic nature of Raman scattering allows both modalities to be driven by the same light source, encouraging the possibility of simultaneous measurement. In practice, however, simultaneous measurement is challenging due to fluorescence signals overwhelming those of Raman scattering by many orders of magnitude. In this talk we report our experimental demonstration of a Raman-fluorescence spectrometer which employs coherent Fourier-transform anti-Stokes Raman scattering (FT-CARS)^{1,2} and Fourier-transform two-photon excitation (FT-TPE)^{3,4} to perform simultaneous measurement in the time domain at an unprecedented spectrum acquisition rate of >20,000 scans/sec. We find time domain acquisition well suited to signal separation given the different coherence lifetimes of FT-CARS and FT-TPE. Potential applications of the spectrometer are numerous, ranging from cytometry to microscopy in various settings.



Figure: Schematic of setup and experimental results from Coumarin 314 in chloroform. (a) Simultaneously acquired time domain interferograms for two-photon fluorescence (top trace, signal around zero delay) and coherent anti-Stokes Raman scattering (bottom trace, signal from 200 to 1400 fs). (b) Two-photon fluorescence excitation of Coumarin 314 and (c) Raman scattering spectra of chloroform, recovered by Fourier transform of the time domain data.

References

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