LINEAR AND NONLINEAR MOLECULAR SPECTROSCOPY WITH LASER FREQUENCY COMBS

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The regular pulse train of a mode-locked femtosecond laser can give rise to a comb spectrum of millions of laser modes with a spacing precisely equal to the pulse repetition frequency. Laser frequency combs were conceived a decade ago as tools for the precision spectroscopy of atomic hydrogen. They are now becoming enabling tools for an increasing number of applications, including molecular spectroscopy.

Recent experiments of multi-heterodyne frequency comb Fourier transform spectroscopy (also called dual-comb spectroscopy) have demonstrated[a] that the precisely spaced spectral lines of a laser frequency comb can be harnessed for new techniques of linear absorption spectroscopy. The first proof-of-principle experiments have demonstrated a very exciting potential of dual-comb spectroscopy without moving parts for ultra-rapid and ultra-sensitive recording of complex broad spectral bandwidth molecular spectra. Compared to conventional Michelson-based Fourier transform spectroscopy, recording times could be shortened from seconds to microseconds, with intriguing prospects for spectroscopy of short lived transient species. The resolution improves proportionally to the measurement time. Therefore longer recordings allow high resolution spectroscopy of molecules with extreme precision, since the absolute frequency of each laser comb line can be known with the accuracy of an atomic clock.

Moreover, since laser frequency combs involve intense ultrashort laser pulses, nonlinear interactions can be harnessed. Broad spectral bandwidth ultra-rapid nonlinear molecular spectroscopy and imaging with two laser frequency combs is demonstrated[b] with coherent Raman effects and two-photon excitation. Real-time multiplex accessing of hyperspectral images may dramatically expand the range of applications of nonlinear microscopy.

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[a] B. Bernhardt et al., Nature Photonics 4, 55-57 (2010); A. Schliesser et al. Nature Photonics 6, 440-449 (2012); T. Ideguchi et al. arXiv:1201.4177 (2012)

[b] T. Ideguchi et al., Optics letters 37, 4498-4500 (2012); T. Ideguchi et al. arXiv:1302.2414 (2013)