NONLINEAR DUAL-COMB SPECTROSCOPY WITH TWO-PHOTON EXCITATION

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Dual frequency comb spectroscopy has proven to be a powerful method for acquiring broadband, high resolution spectra with measurement times that are much shorter than in traditional moving-mirror Fourier transform spectroscopy. Because the measurements are carried out with femtosecond lasers, this technique has great potential for decreasing the measurement times and improving the signal-to-noise ratio of nonlinear spectroscopic measurements, such as two-photon excitation or Raman processes. In the case of two-photon excitation, an entire spectrum can be obtained at a given signal level using dual-comb spectroscopy in the same time that a measurement of a single transition frequency would be obtained with a continuous laser of the same average power.

In this presentation, I will show the latest results in extending the dual-comb technique to two-photon excitation spectroscopy, with measurements on gas-phase rubidium and liquid-phase dye samples. In our realization of dual-comb spectroscopy, two frequency combs with slightly different repetition rates are combined on a beam splitter and directed into a sample, and we measure the intensity of the resulting fluorescence as a function of time. Because of the different repetition rates, the time delay between a pulse from the first comb and the next pulse from the second comb changes linearly with time, simulating the action of the moving mirror in a traditional Michelson interferometer. The Fourier transform of the measured time-domain interferogram produces a radio-frequency spectrum that can be directly converted to a broadband optical spectrum through a linear scaling of the frequency. To achieve the highest possible resolution, it is necessary to compensate the residual relative fluctuations of the repetition rate and the carrier-envelope offset frequency of the frequency combs. Measuring RF beatnotes of each comb with two CW lasers provides two error signals that can be used to correct the recorded interferograms. This correction is applied differently for one-photon and two-photon spectra, providing a method of distinguishing the two.