Phase-Modulated Two-Photon Interferometry and Spectroscopy

J. Lavoie[1,2], Tiemo Landes[1,2], Amr Tamimi[1,3], Brian J. Smith[1,2], Andrew H. Marcus[1,3], and Michael G. Raymer[1,2]

[1] Oregon Center for Optical, Molecular & Quantum Science, University of Oregon, Eugene, OR 97403, USA

[2] Department of Physics, University of Oregon, Eugene, OR 97403, USA

[3] Dept of Chemistry and Biochemistry, University of Oregon, Eugene, OR 97403, USA

Abstract

We introduce optical lock-in detection in photon counting to enhance the extraction of information in "quantum-light spectroscopy," based on using time-frequency entangled photon pairs to probe a molecular sample. The sample is placed inside a Mach-Zehnder interferometer, into which photons are injected, and detected in coincidence at the interferometer output. Clear signatures of two-photon interference (both Hong-Ou-Mandel and 'N00N') are observed and separated into distinct information channels. In addition to achieving an increased signal-to-noise ratio (SNR) at low count rates, the experiment uses a "down-sampling" technique that increases the efficiency of data collection by an order of magnitude.

Time-frequency entangled photon pairs (EPP) are a promising resource for enhancing nonlinear spectroscopy and metrology. Such bi-photon states can be created tightly correlated in time while also being anti-correlated in frequency, such that the sum of the photon energies is sharply defined. The short correlation time (~20 fs) allows EPP to play a role similar to that of ultrashort laser pulses in multidimensional spectroscopy [1]. At the same time, the entanglement—analogous to the EPR state—enables high spectral resolution of two-photon molecular absorption features. Earlier we proposed entangled-photon-pair two-dimensional fluorescence spectroscopy (EPP-2DSF) [2].

We introduce optical lock-in detection to increase SNR in EPP coincidence detection, leading toward implementation of the EPP-2DFS method. The method presented here uses a Mach-Zehnder interferometer (MZI) with a linear-response sample in one internal path, allowing both amplitude and phase information to be acquired simultaneously. EPP enter one MZI port and are detected in coincidence at a single output port as a function of interferometer delay. Our scheme uses a "down-sampling" technique, analogous to the methods used in the "classical" 2DFS scheme developed by one of us [3]. By introducing a relative phase modulation between MZI arms, injecting a reference laser beam into the MZI, and locking the detection to multiples of the difference-modulation frequency f, we increase SNR and reduce the number of samples needed.

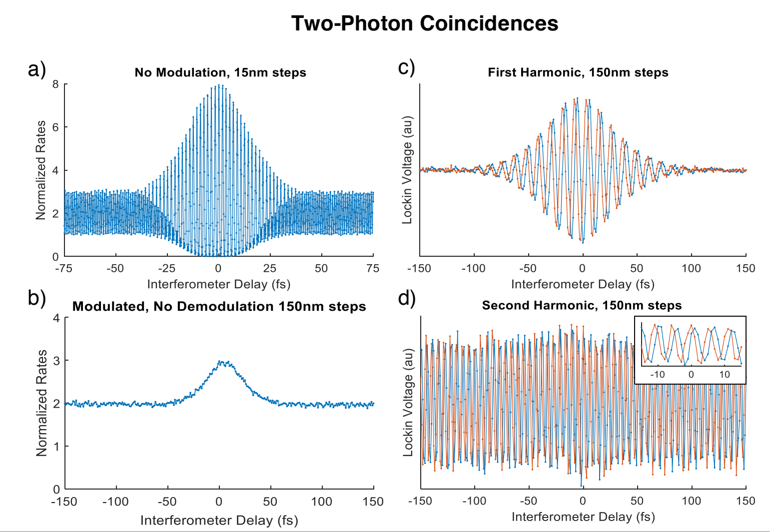

Fig. 1. Coincidence rate vs. MZI delay, with no internal sample.
(a) Fully-sampled, with 15 nm delay steps. (b-d) Down-sampled coincidence rate using 150 nm delay steps, lock-in detected relative to the difference frequency modulation f between the signal photon carrier frequency and a reference laser detuned from the signal carrier. (b) at zero modulation frequency, Hong-Ou-Mandel peak;
(c) at the modulation frequency f, one-photon interference;
(d) at twice the modulation frequency $2f$ - NOON-state interference.

[1] K. E. Dorfman, F. Schlawin, and S. Mukamel, *Nonlinear optical signals and spectroscopy with quantum light*, Rev. Mod. Phys. **88**, 045008-1-67 (2016).

[2] M. G. Raymer, A. H. Marcus, J. R. Widom, D. L. P. Vitullo, *Entangled Photon-Pair Two Dimensional Fluorescence Spectroscopy (EPP-2DFS)*, J. Phys. Chem. B, **117**, 15559-15575 (2013).

[3] P. F. Tekavec, G. A. Lott, and A. H. Marcus, *Fluorescence-detected two-dimensional electronic coherence spectroscopy by acousto-optic phase modulation*, J. Chem. Phys., **127**, 214307 (2007).