Vibrational Interferometry Enables Single-Scan Acquisition of all $\chi^{(3)}$ Multi-Dimensional Coherent Spectral Maps

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Abstract: We demonstrate a new method for multidimensional coherent spectroscopy of nanostructures. We use a heterodyne technique implemented with a confocal microscope to record the amplitude and phase of all degenerate third-order wave-mixing processes.
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1. Introduction

Multidimensional Coherent Spectroscopy (MDCS) is a powerful method for probing the optical properties of physical systems by correlating the absorption, emission, and mixing frequencies of a nonlinear four-wave mixing (FWM) signal. MDCS spectra have been demonstrated to unambiguously measure coherent vs. incoherent coupling as well as homogenous and inhomogeneous linewidths in a variety of systems. Conventional methods for MDCS rely on wave-vector phase matching—a technique that fails for emission from point sources and is not compatible with nano-emitters integrated into waveguide geometries. Alternative methods appropriate for individual nano-objects such as quantum dots, single molecules, or carbon nanotubes rely on phase cycling to detect "action" signals as fluorescence, photoelectrons, or photocurrent. These methods suffer from poor collection efficiency and have limited ability to amplify the signal, since heterodyne detection is not possible. Furthermore, these techniques can only measure FWM signals across a ~10 ps timescale, or they can lack a suitable optical reference to properly process the signals [1-6].

2. Methods

Here, we present a new collinear method, appropriate for long-lived dipole radiation in the far field, that circumvents these limitations through vibrational-interferometry of the inter-pulse delays using a far-detuned continuous-wave reference laser. Our approach uses a pulse-to-pulse phase-cycling scheme with heterodyne detection insuring shot noise-limited signal-noise Unlike conventional methods, we record *all* degenerate wave-mixing processes in a *single* measurement, *i.e.,* zero-quantum and one-quantum FWM signals (both rephasing and non-rephasing pathways), the two-quantum FWM signal, as well as time-resolved linear absorption. Simultaneous measurement of these signals enables complete characterization of the $\chi^{(3)}$ resonant nonlinear optical response of nano-systems with instrument limited ~ 8 orders of temporal dynamic range, ~4 μeV resolution/ 21eV bandwidth, signals as low as 1 photon/pulse, and ~10's of attoseconds precision.

To achieve pulse-to-pulse phase cycling, we drive an acousto-optical modulator (AOM) in each arm of a nested Mach-Zehnder interferometer at radio frequencies that differ in the MHz range (Fig. 1A). The AOMs provide a unique

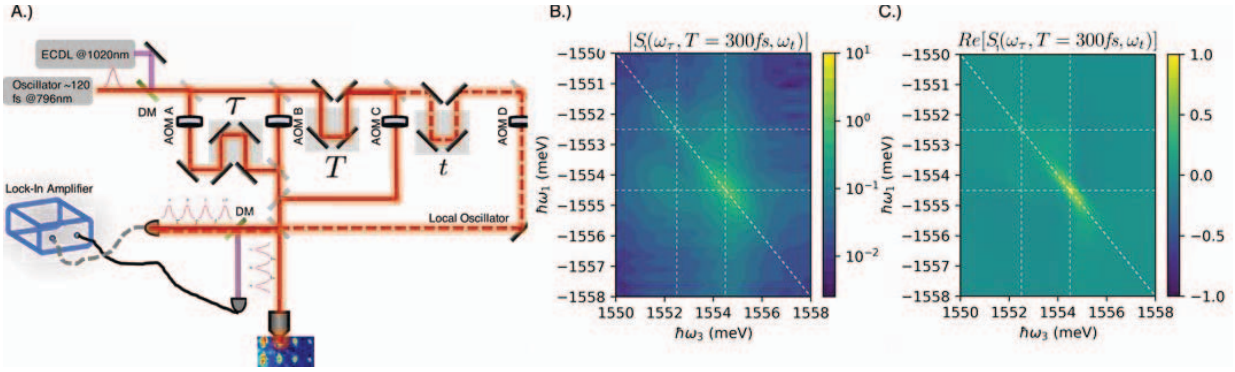

Figure 1 A.) Experimental apparatus showing two Mach-Zehnder interferometers nested within a larger interferometer. Each arm of the interferometer has an AOM driven at a unique radio frequency. At the interferometer entrance/output, both a Ti:Sapph. oscillator (at 796 nm) and a CW laser (at 1020 nm) are combined/split on a dichroic beamsplitter (DM). B.) Measured amplitude of a rephasing pulse sequence showing three orders of magnitude signal-to-noise. C.) Real part of rephasing pulse sequence demonstrating the recovery of the four-wave mixing phase.

carrier-envelope offset for each arm of the interferometer. Experimentally this manifests as a pulse-to-pulse phase accumulation between interferometer arms that evolves at the "beat" frequency of the different AOM drive frequencies. Conveniently, linear signals are recorded at the beat of each pair of interferometer arms, while nonlinear FWM signals are recorded at the higher-order beats involving all four arms of the interferometer. Three pulses (ABC) are recombined and sent through single-mode fiber before focusing through the microscope. A signal is collected in the reflected direction and heterodyne-mixed with a local oscillator through a single-mode photodetector

Unlike conventional methods of achieving phase stability, our interferometer has no common optical path, no requirements for actively stabilizing the interferometer path length, and does not need an external reference for demodulation using a lock-in amplifier. Instead, we time-resolve the interferometer vibrations (which will appear as phase modulation on our signals) by sampling with a high-speed digitizer at ~28 kHz. All three of the degenerate FWM signals, a time-resolved linear absorption signal (C-LO), and an autocorrelation signal between arms (A-B) are recorded synchronously at different radio frequencies. Simultaneously, we send a strongly red-detuned (1020 nm) reference laser through the interferometers and detect modulation beats on a separate detector. The reference laser samples the interferometer vibrations and position as a delay stage is scanned continuously. Because the reference beat note is phase sensitive, modulation caused by the interferometer vibrations manifests as relative Doppler shifts in the interferometer arms. The strongly red-detuned reference laser (relative to the oscillator at 796 nm) is an improvement over previous schemes that require the use of a degenerate optical reference because it avoids background population due to reference laser excitation, does not rely on stage encoders for interferometer position, and does not need to be tuned if the oscillator wavelength is adjusted. The effect of post-processing is to create a super-heterodyne optical receiver with feed-forward carrier phase recovery. Thus, the FWM signals cans be shifted to an arbitrary rotating optical frame using this reference. This experiment is appropriate for measuring long-lived coherences and recombination lifetimes limited only by the stage length.

3. Results

To demonstrate our technique, we resonantly excite a four-quantum-well sample with 100 fs pulses (at 796 nm). Our sample consists of four 8-nm AlGaAs/GaAs quantum wells grown on top of a semiconductor Bragg mirror. All three-degenerate wave-mixing signals (rephasing, non-rephasing, and two-quantum) are recorded simultaneously at their respective demodulation frequencies. The amplitude (Fig.1B) of a typical rephasing pulse sequence demonstrates three orders of magnitude of SNR with an excitation power of 13 μW (spot size ~9 μm). The spectrum has two diagonal peaks corresponding to a heavy-hole exciton and a weak trion which are coupled as indicated by the presence of off-diagonal cross-peaks. We attribute the presence of the weak trion to unintentional background doping in the growth process. The real part of the of the rephasing FWM signal (Fig. 1C) is purely absorptive, demonstrating the excellent ability to recover the carrier phase of the FWM signal.

4. Conclusion

In summary, we have demonstrated a technique that measures all four-wave mixing quantum pathways simultaneously in a *single* measurement. This experiment does not rely on wave-vector matching in the far field, can operate with diffraction-limited spatial resolution (*i.e.*, high numerical aperture objectives), and leverages heterodyne detection making it uniquely suited to optical experiments on single and few quantum emitters. Further, by using an optical reference laser recorded with better than 60-70 dB of signal-to-noise, our achievable phase stability and timing resolution is better than 20 attoseconds.

5. References

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