Projection-slice four-wave-mixing spectroscopy using frequency combs

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Abstract: We demonstrate projection-slice spectroscopy using frequency combs that enables rapid measurements of couplings between sample's excited states. This information is not easily accessible with other one-dimensional methods and is critical for imaging and chemical sensing.

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Over the years, one-dimensional techniques such as Fourier Transform (FTIR) spectroscopy, fluorescence spectroscopy, coherent anti-stokes Raman spectroscopy (CARS), etc have proven to be very powerful both for fundamental science as well as for practical applications such as chemical sensing and imaging. When using these methods, chemical identification process is usually performed by measuring spectra (e.g. linear absorption or CARS) of a sample of interest and then comparing them to the databases of known spectra. However, these methods face challenges to distinguish between a three-level system with two resonant frequencies f_1 and f_2 and a mixture of two independent two-level systems with the same frequencies. Clearly one-dimensional spectra of these systems would look identical and this is a critical limitation for chemical sensing applications.

These limitations are typically overcome by using multidimensional methods e.g. multidimensional coherent spectroscopy (MDCS) [1] which, in the past two decades, has emerged as a very powerful technique for studying materials physical, chemical, and structural properties. There are different variations of MDCS but one of the most commonly used MDCS approaches uses a sequence of three pulses (A*-complex conjugate pulse, B, C, as shown in figure 1 (a)) incident on the sample of interest and generates a photon echo four-wave mixing (FWM) signal which is then heterodyne detected, using a local oscillator pulse, as a function of the delays between the excitation pulses. The recorded time domain interferogram (shown in figure 1 (b)) is then Fourier transformed with respect to the time delays between the incident pulses and over the time period during which the signal is emitted (t and τ) to generate a multidimensional coherent spectrum. This excitation scheme has many advantages and among them is the ability to detect the couplings between the excited states. This point is demonstrated in figure 1 (c) and (d) where cartoons of 2D spectra are plotted for a V - type and two independent two-level systems, respectively. On the 2D spectra, diagonal peaks (along the white dotted line) correspond to samples' resonances and off diagonal peaks determine the cross peaks (couplings) indicating that the resonances belong to the same system.

Clearly, MDCS provides critical information that is not easily accessible using 1D methods but the experimental setup either requires bulky and slow mechanical moving stages and complex phase cycling schemes or asynchronously scanning three lasers (Tri-Comb Spectroscopy [2]) and complex locking electronics [3]. They are not suitable for applications outside the lab.

Here we propose a simpler approach that enables rapid measurements of the main as well as the cross peaks without measuring the full multidimensional coherent spectra. Our approach is based on the projection-slice theorem which states that the one-dimensional Fourier transform of a slice in figure 1 (b) (along the white dotted line) is equivalent of the projections of figures 1 (c) and (d) on the diagonal line. The projections are shown in figure 1 (e) and (f). Figure 1 (e) clearly displays the cross-peak that can be used to differentiate a V - type system and two independent two-level systems. This approach was inspired by the dual-comb spectroscopy [4] detection technique that can allow the measurements along the line (figure 1 (b)) rapidly (simultaneously along t and τ) without using any mechanical moving parts.

A simplified schematic diagram of our experimental setup is shown in figure 1 (g). We used two home-built 540 MHz repetition rate frequency combs centered at 800 nm. The output of Comb 1 was split into two parts. One part was combined with Comb 2 and focused on the sample. The second part of Comb 1 was slightly delayed with respect to the first part and used as a local oscillator to interfere on a detector with signals generated from the sample. Our combs had slightly different repetition rates ($\sim$50 Hz) which allowed Comb 2 pulses to be scanned with respect to Comb 1 (increasing τ) and simultaneously photon echo pulses to be scanned with respect to LO pulses (decreasing t) as shown in figure 1 (h). This arraignment is equivalent of scanning along the line shown in figure 1 (b). We should note that in our experiment we used co-linear excitation geometry, which is desirable for imaging applications, and linear and nonlinear contributions were separated in the RF domain.

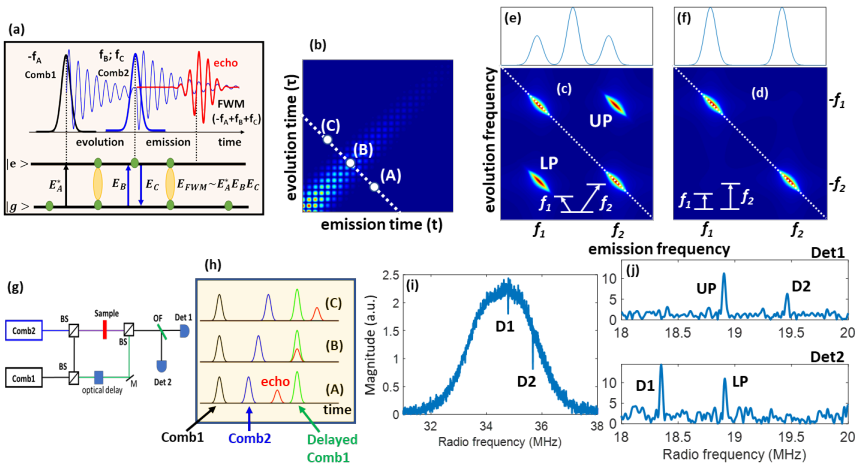

Fig. 1. (a) Photon-echo excitation scheme using two trains (blue and black) of pulses. (b) Cartoon showing the magnitude of a FWM signal as a function of evolution (τ) and emission (t) times. (c) 2D spectrum of a V-type system, (d) 2D spectrum of a two independent two-level systems. UP-upper cross peak, LP-lower cross peak. The evolution frequency is negative to reflect the negative phase evolution during the evolution period. (e) and (f) Projections of (c) and (d) on the diagonal lines. (g) Experimental setup, BS-beam splitter, M-mirror, OF-optical filter. (h) Acquisition along the slice in (b). (i) Linear dual-comb spectrum of rubidium D1 and D2 lines. (j) Fourier-transform of non-linear signals (slice) detected using Det1 (top) and Det2 (bottom).

For this proof-of-concept experiment, we used rubidium atoms as our sample that is a V type system with D1 and D2 absorption lines at 795 and 780 nm, respectively. The sample was loaded in a cell with 1 mm length and heated up to 130 degree C. We should note that the projection-slice acquisition approach described above, causes UP and LP peaks (shown in figure 1 (c)) for a V type system to be projected on top of each other. Resolving these peaks, in most cases, is not critical for imaging and sensing applications, however measuring them separately can be important and beneficial to probe fundamental processes (e.g. many-body effects [1]). For this reason, in our experiment we added an optical interference filter in front of Det 1 that passes 780 nm light. The reflected portion of the beam, which contains the signal at 795 nm (780 nm light is eliminated), was simultaneously detected on Det 2. In order to completely eliminate 780 nm light, we used an optical bandpass filter (centered at 795 nm) in front of Det 2 as well.

Results of the experiment are shown in figures 1 (i) and (j). Figure 1 (i) shows a typical linear absorption dual-comb spectrum with D1 and D2 absorption lines. On the other hand, in figure 1 (j) (top) and (bottom) we show 1D Fourier transforms of FWM echo signals detected using detector 1 and detector 2, respectively. The spectrum obtained using Det 1 shows projections of bottom diagonal and upper cross peaks (see figure 1 (c) for reference) whereas Det 2 shows projections of upper diagonal and lower cross peaks. The results clearly indicate that D1 and D2 lines belong to a V-type system which was not reflected on the linear absorption spectrum.

In conclusion, we have demonstrated projection-slice spectroscopy using frequency combs. We showed that this approach, in combination with linear spectra, provides critical information for sensing and imaging applications that is not easily accessible using traditional 1D methods. This approach doesn't use mechanical moving parts and with the development of micro-combs it can be used for many practical applications outside the lab.

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References

1. Steven T. Cundiff, and Shaul Mukamel, "Optical multidimensional coherent spectroscopy". Physics today **66**, 44-49 (2013).
2. Bachana Lomsadze, Brad. C. Smith and Steven Cundiff. "Tri-Comb Spectroscopy" Nature Photonics **12**, 676–680 (2018).
3. Bachana Lomsadze, and Steven Cundiff. "Frequency combs enable rapid and high resolution multidimensional coherent sepctroscopy" Science **357**, 1389-1391 (2017).
4. Ian Coddington, Nathan Newbury, and William Swann, "Dual Comb Spectroscopy". Optica **3**, 414-426 (2016).