



Dispersive Fourier transformation for megahertz detection of coherent Stokes and anti-Stokes Raman spectra



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ABSTRACT

In many fields of study, from coherent Raman microscopy on living cells to time-resolved coherent Raman spectroscopy of gas-phase turbulence and combustion reaction dynamics, the need for the capability to time-resolve fast dynamical and nonrepetitive processes has led to the continued development of high-speed coherent Raman methods and new high-repetition rate laser sources, such as pulse-burst laser systems. However, much less emphasis has been placed on our ability to detect shot to shot coherent Raman spectra at equivalently high scan rates, across the kilohertz to megahertz regime. This is beyond the capability of modern scientific charge coupled device (CCD) cameras, for instance, as would be employed with a Czerny-Turner type spectrograph. As an alternative detection strategy with megahertz spectral detection rate, we demonstrate dispersive Fourier transformation detection of pulsed (~90 ps) coherent Raman signals in the time-domain. Instead of reading the frequency domain signal out using a spectrometer and CCD, the signal is transformed into a time-domain waveform through dispersive Fourier transformation in a long single-mode fiber and read-out with a fast sampling photodiode and oscilloscope. Molecular O- and S-branch rotational sideband spectra from both N₂ and H₂ were acquired employing this scheme, and the waveform is fitted to show highly quantitative agreement with a molecular model. The total detection time for the rotational spectrum was 20 ns, indicating an upper limit to the detection frequency of ~50 MHz, significantly faster than any other reported spectrally-resolved coherent anti-Stokes Raman detection strategy to date.

1. Introduction

Coherent anti-Stokes Raman spectroscopy (CARS) is today employed in a variety of research applications, such as cell biology [1], combustion diagnostics [2], and the standoff detection of explosives [3]. It is the combination of excellent chemical selectivity, i.e. the ability to probe distinct quantum states of individual molecular species, and the coherent properties of the generated signal beam, that make this technique very powerful. For many applications, rapidly changing systems, with characteristic time-scales ranging from μ s to ms require that measurements are performed nearly instantaneously, ideally within a single shot of a short-pulsed laser. This is a unique strength of Raman-based spectroscopies, as it is the time-duration of the probe pulse which ultimately limits the temporal resolution of the technique. Thus, users of coherent Raman spectroscopies have long been able to “freeze” dynamics of interest by recording data from a single laser shot. Statistics can then be built up from single realizations of the measured physical scalar, such as the instantaneous molecular temperature or concentration, as evaluated from the measured molecular spectrum.

However, to follow time-correlated dynamics of a system which is evolving on the time scale of μ s, the spectral detection strategy must also be capable of a very high refresh rate, in the megahertz regime [4]. Cutting-edge CMOS camera technologies are now capable of 1 MHz detection frame rates when operated in a cropped array mode with ~128 active pixels across the chip for spectral detection. Such systems are also limited to 12-bit depth, while the squared dependence on molecular density for coherent Raman spectroscopy often requires 14- or 16-bit depth for sufficient dynamic range. Line-scan CCD cameras have been demonstrated for continuous spectral measurements at 100-kHz, for example, in transient absorption spectroscopy experiments [5]. Faster CCD detection rates may be possible with multi-frame CCD options [4,6], but such technologies are limited by the physical framing built into the camera to ~10's of frames and also suffer from blooming effects as the charge is transferred through the storage array, reducing the quantitative accuracy of stored data.

Another option, the topic of the current work, is to consider frequency-resolved spectral detection performed in the time-domain by the technique of dispersive Fourier transformation (DFT) [7]. The

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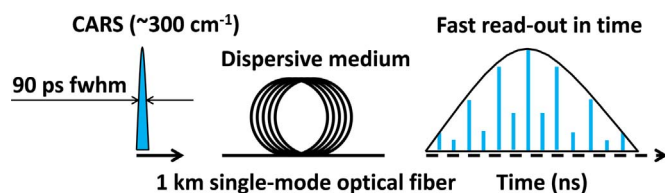


Fig. 1. Illustrated principle of spectral detection performed in the time-domain. A pulsed broadband spectrum is sent through a dispersive medium, consisting of a long single-mode fiber, where the discretized wavelengths get separated with group velocity dispersion and classified based on their time of arrival. At the output the spectral components are detected with a fast detector sampling device.

basic idea, depicted in Fig. 1, is to send the pulsed broadband CARS signal through a long fiber where the intrinsic group velocity dispersion (GVD) of the medium acts to spectrally separate the different wavelengths of the signal. The discretized spectral components are then registered with a fast single-pixel detector sampling read-out and get classified based on their arrival time, in the same manner as a classic time-of-flight instrument. This detection scheme has been proposed in connection to fiber dispersion measurements [8], and variants of the same scheme have been explored in optical sensing applications [9–11], where molecular absorption spectra resulting from supercontinuum wavelength sweeps are mapped as waveforms in the time-domain. Instead, in the current work, it is the fact that the signal pulse in a CARS experiment will mimic the time-behavior of the probe pulse, i.e., a short or ultra-short pulse of light which contains the frequency-resolved coherent Raman spectrum, which allows the use of DFT to frequency-resolve the signal spectrum via chromatic dispersion in time with fast detection.

2. Experimental

The generation of the coherent Raman signal was performed with time synchronized femtosecond (fs) and picosecond (ps) laser systems configured in a hybrid fs pump ps probe setup [12,13], displayed in Fig. 2. The fs laser system consisted of an oscillator (KM Labs, Halcyon) synced to an external 100 MHz radio frequency (RF) source acting as a master clock. The oscillator pumped a fs regenerative amplifier (KM Labs, Wyvern 1000), producing output pulses of 45 fs full-width at half maximum (fwhm) centered at a wavelength of ~800 nm, and operated at about 3 mJ/pulse with a rate of 1 kHz.

The ps laser system consisted of a 20 Hz regenerative amplified mode-locked Nd:YAG laser, with a seed laser phase-locked to the same

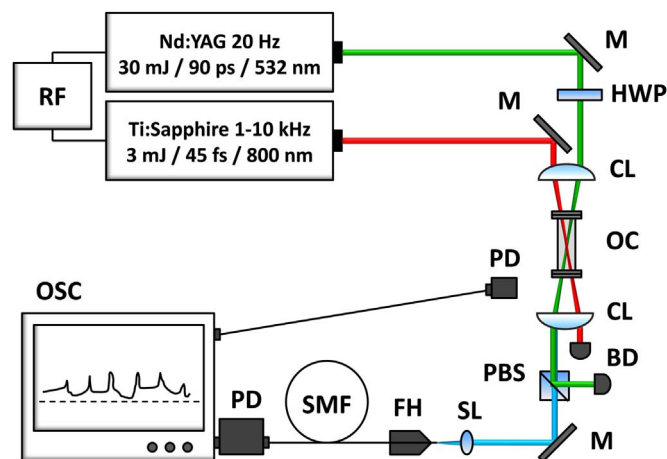


Fig. 2. Experimental setup for generating coherent rotational Raman signals and time-domain fiber dispersion detection strategy. M – mirror, HWP – half wave plate, CL – cylindrical lens, OC – optical cell, BD – beam dump, PBS – polarizing beam splitter cube, SL – spherical lens, FH – fiber holder, SMF – single-mode fiber, PD – photo diode, OSC – oscilloscope, RF – 100 MHz radio frequency master clock.

external 100 MHz RF source, allowing for precise electronic timing between the fs and ps pulses at the experiment with sub-ps jitter. The output of the frequency doubled Nd:YAG at 532 nm was ~30 mJ/pulse with a pulse duration of approximately 90 ps fwhm. The laser beams were setup in a crossed two-beam configuration [14], with small enough incident angle [15] to achieve near perfect phase-matching condition for all the involved transitions ($< 360 \text{ cm}^{-1}$) of this study. An optical cell was placed at the crossing in order to generate signals from pure gas-mixtures of two separate molecules, N_2 and H_2 . The coherent Raman scattered signals of both the species, containing rotational sidebands from Stokes and anti-Stokes shifted light, respectively, were isolated from the probe beam using a polarization gating technique [14]. The signal beam was injected via a $f=25 \text{ mm}$ focusing lens into a 1 km long single-mode (SM) optical fiber (Nufern, 460-HP) placed on a high finesse x-y-z pedestal stage. While the dispersion length for fs pulses could have allowed a shorter fiber to be used, ps length pulses are required in hybrid fs/ps CARS in order to frequency resolve rotational coherences [12,16]. An FC-connector mounted at the end of the fiber channeled the dispersed light into a high-speed unamplified photodiode detector (Picometrix, D-15-FC), having an impulse response of 15 ps full duration at half maximum (fdhm). The photodiode output was finally digitalized using a 6 GHz bandwidth digital storage oscilloscope (Tektronix, TDS6604) with a 20 GS/s sample rate for single-shot acquisition of the signal waveform. The recordings were started at a fixed delay, triggered from the output of a photodiode (EOT ET-3500) placed near the signal generation plane.

3. Results and discussion

The transformation of the frequency-resolved CARS signal pulse into a time-domain waveform through the fiber is given by $\Delta\tau = |D|z\Delta\lambda$, where D is the GVD of the dispersive element, which was measured to be $D=496 \text{ ps nm}^{-1} \text{ km}^{-1}$, $\Delta\lambda$ is the bandwidth of the signal pulse and z is the propagation distance in the fiber. The maximum spectral resolution of the current DFT instrument is thus limited by the detection bandwidth of our 6 GHz oscilloscope, as both the total fiber dispersion and fast photodiode could provide resolution approximately one order of magnitude greater than could be detected on the oscilloscope. The spectral resolution, measured as the FWHM of the Rayleigh line, was $\sim 8 \text{ cm}^{-1}$, limited by the bandwidth of our current oscilloscope, and could thus be improved to $\sim 0.8 \text{ cm}^{-1}$ if the limiting factor was instead the fast photodiode and DFT fiber. However, as shown below, the acquired resolution was sufficient for quantitative fitting of the molecular spectrum to evaluate the N_2 rotational temperature. While the achieved resolution is below that possible with a grating and camera based spectrometer, the detection time is far faster, as is common to DFT spectroscopy based detection schemes.

Fig. 3 displays the results from the sampling of coherent rotational Raman signals recorded in pure gas-phase samples of N_2 (a) and H_2 (b). The acquisitions shown were slightly averaged from 10 instantaneous measurements, to reduce contributions from the detector noise. The detected waveforms show distinct dispersed spectral components, from both the O ($\Delta J = -2$) and S ($\Delta J = 2$) branch rotational sidebands, where J denotes the rotational quantum number of the molecules. In the context of a non-linear technique, the O and S branches correspond to redshifted (Stokes) and blueshifted (anti-Stokes) light, respectively, relative to the fixed wavelength of the probe beam. Therefore, in Fig. 3, with positive dispersion of the optical fiber, the coherent Stokes Raman scattered light (CSRS) arrives before the CARS light, and accordingly have been assigned negative times relative the zero-shift probe beam, detected at time zero. Note, that the remaining probe light has been transmitted intentionally through the system to be used as a point of reference. Otherwise, the polarization gating of the two-beam rotational CARS technique can easily achieve a $1:10^6$ suppression of the probe beam. In the insert of Fig. 3a rotational CARS spectrum is shown recorded in a mixture with 50% N_2 and 50% H_2 as a result from a

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