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Ultra-broadband two beam CARS using femtosecond laser pulses☆

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ABSTRACT

Femtosecond (fs)-CARS is a promising approach for gas spectroscopy under high pressure and temperature conditions, as it allows probing molecular states on a time scale which is significantly shorter than the typical decay time induced by interfering collisions. Usually, fs-CARS is performed in a three beam setup, which requires maintaining spatial and temporal overlap of the pulses at the focal point. This is a challenging task, especially in harsh environments such as in a combustion chamber. In this study, we present an alternative approach, which uses two beams in a collinear configuration. An ultra-broadband, sub 7 fs laser pulse acts as pump and Stokes pulse, and a ~500 fs pulse is used for probing. We show that this configuration is suitable for measuring the gas temperature and concentration. Furthermore, possible single shot measurements of the gas temperature are evaluated.

1. Introduction

In recent years, femtosecond coherent Anti-Stokes Raman scattering (fs-CARS) has been intensively investigated in various configurations, as it is a promising approach for applications in gas spectroscopy [1]. A major advantage over CARS techniques using longer pulse durations is the possibility to extract gas temperatures at high pressure [2]. Usually, the Stokes pulse is provided by a Ti:sapphire laser, and the pump and probe pulses are provided by an optical parametric amplifier (OPA) in a BOXCARS configuration [3]. This configuration makes it difficult, however, to maintain the spatial and temporal overlap of the three beams in harsh environments such as in a combustion chamber. Moreover, investigations are limited to the one gas species, for which the corresponding Raman shift is provided by the OPA.

Using ultra-broadband, sub 7 fs pulses could overcome these drawbacks, as their large spectrum may provide the pump, Stokes and even the probe pulse within the same laser pulse. Roy et al. [4,5] used a single-beam configuration to investigate gaseous N_2 and CO_2. This configuration requires a spatial light modulator (SLM), which rotates the polarization of a small part of the pulse spectrum to realize the 'probe pulse'. However, the maximum pulse energy of the sub 7 fs pulse is limited by the damage threshold of the SLM. Moreover, the setup was used to detect only either N_2 or CO_2 at the same time so far, hence, the multiplex capability of the ultra-broadband pump/Stokes-pulse remained unexploited.

Bohlin et al. demonstrated the use of an ultra-broadband pump/Stokes and a probe pulse for rotational CARS [6]. Moreover, they *simultaneously* detected multiple species in an open gas flow in a two-beam configuration, using a sub 7 fs pulse for the pump/Stokes pulse and a 90 ps pulse for the probe [7]. Although this method generates spectra with high resolution, it does not benefit from essential advantages of ultra-broadband fs-CARS, too. For example, a common technique to determine the gas temperature from a fs-CARS experiment is to measure the decay of the Raman coherence within the first ps after excitation [8]. This is especially advantageous for high pressure applications, as it was shown for various N_2 mixtures that the decay does not depend on pressure during the first 1–3 ps up to 50 bar [2]. In neat N_2, Knopp et al. demonstrated that disturbing line mixing effects begin at probe pulse delays of ~25 ps for a pressure of 5 bar [9]. Thus, a probe pulse duration of 90 ps is too long for pressures considerably above ~1 bar.

Furthermore, the multiplex approach of using a sub 7 fs pulse could allow determining both gas temperatures and concentrations from a single spectrum. This capability was shown in a previous study in a hydrocarbon flame using a two-beam configuration with a 70 ps probe pulse [10]. For high pressure applications and, hence, probe pulse durations on a fs-time scale

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with a considerably larger spectral width, the multiplex approach could be combined with a technique that uses a chirped probe pulse [11]. It encodes the temporal probe pulse delay into the spectrum of the CARS pulse. Thus, the scan of the probe pulse delay is avoided, and the gas temperature can be extracted from a single spectrum. Moreover, the same spectrum could be used for concentration measurements, if the Raman cross sections of the molecules in question are known.

The present study investigates the possibility to use a probe pulse with high repetition rate (30 kHz) and a pulse duration below 1 ps. Thereby, we focused our research on the following questions: (i) Does this configuration allow measuring the decay of the Raman coherence as with three-beam fs-CARS? (ii) Is this technique also suitable to determine gas temperatures and concentrations from a single spectrum? (iii) Are the pulse parameters suitable for transmission through the optical ports of a gas cell?

2. Material and methods

The experimental setup is shown in Fig. 1. Sub 7 fs pulses are generated by a carrier envelope phase (CEP-)stabilized, optical parametric chirped pulse amplification (OPCPA) laser, which is a modified system presented by Rothhardt et al. [12]. For the present study, only the first stage of that system was used. It provided a spectrum ranging from ~700 nm up to 1200 nm (Fig. 2) with maximum pulse energies of 8 μJ at a repetition rate of 30 kHz. The stability of the pulse energy was ±5% (peak-to-peak) on a time scale of ~1 s. On longer time scales, peak-to-peak deviations of ±10% were observed, possibly due to the cooling cycle of the pump diodes. The pulses were compressed by means of broadband, dispersive mirrors. Fine tuning was performed with a glass wedge, and a spatial light modulator (SLM) was used for compensating higher order chirps. Using a SPIDER device (venteon SPIDER, Laser Quantum Ltd., Stockport, UK) and equivalent glass blocks of different size, it was verified that this configuration is capable of compensating the chirp induced by the subsequent CARS setup. The spectral phase of a compressed pulse is shown in Fig. 2 (red line). The corresponding pulse duration is $t_{pump/Stokes} = 5.2$ fs, which is close to the Fourier limit ($t_{TFL} = 5.1$ fs).

The pump pulse from a fiber chirped pulse amplification system (FCPA), which was originally used for the second OPA stage in Ref. [12], served here as a probe for fs-CARS. The FCPA provides pulses

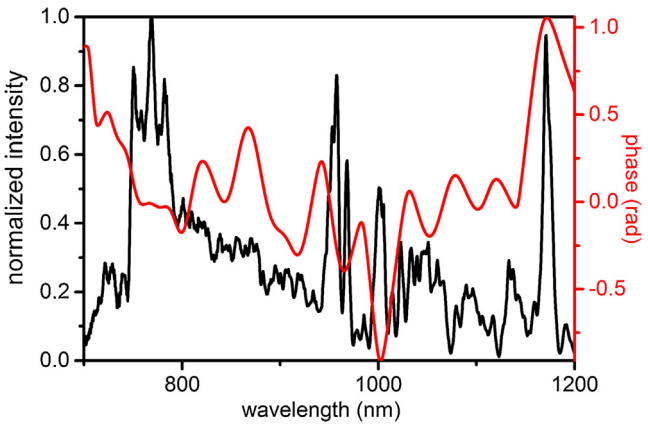

Fig. 2. Spectrum and phase (red) of the compressed pump/Stokes pulse. (For interpretation of the references to colour in this figure legend and text, the reader is referred to the web version of this article.)

at 30 kHz, a central wavelength of ~1030 nm and a pulse duration of 470 nm, which was measured using an intensity autocorrelator (pulseCheck 50, A.P.E. Angewandte Physik & Elektronik GmbH, Berlin, Germany). Subsequent frequency doubling (SHG) in a 1 mm BBO crystal generated pulses with energies up to 160 μJ at a central wavelength of 514.88 nm. For the given crystal length and pump power (~100 GW/cm^2), the influence of SHG on the pulse duration should be negligible [13]. The temporal delay was adjusted by a motorized translation stage with a minimum step size of 330 fs. For CARS, a collinear beam geometry was chosen, using a dichroic mirror to combine the pump/Stokes and the probe pulse. Both beams were p-polarized and focused by a 90° off-axis parabolic silver mirror with a focal length of f ~ 150 mm (MPD269-P01, Thorlabs Inc., Newton, NJ, USA).

The experiments were performed in air and in a high temperature, high pressure gas cell (GS05850, Specac Limited, Orpington, UK). The cell is specified for pressures up to 69 bar (1000 psi) and has a volume of 80 ml. The cell consists of a body with two opposite sapphire windows, which have a clear aperture of ~21 mm and a thickness of 3 mm. The group velocity dispersion (λ = 800 nm) for sapphire is ~58 fs^2/mm [14] compared to 45 fs^2/mm for BK7 [15], for example. Inside of the cell, there is a heater

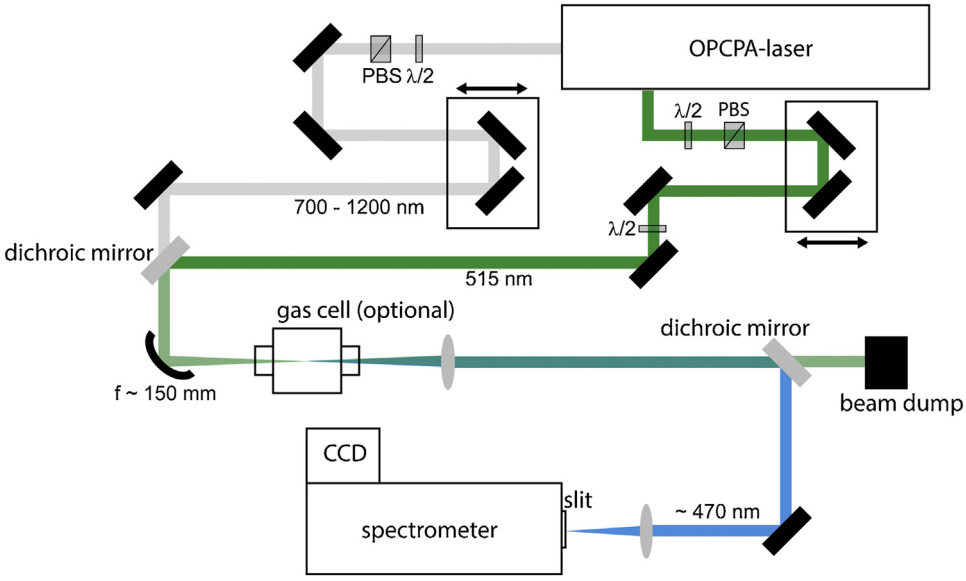

Fig. 1. Sketch of the experimental setup for ultra-broadband two beam CARS.