Coherence lifetime imaging of nitrogen during a laser-induced air spark with FRAME-enhanced coherent anti-Stokes Raman spectroscopy

Vassily Kornienko[1], Yupan Bao[1], Simon Ek[1], Ali Hosseinnia[1], Meena Raveesh[1], Christopher J Kliewer[2], Joakim Bood[1], Andreas Ehn[1] and Elias Kristensson[1]

[1] *Division of Combustion Physics, Department of Physics, Lund University*
[2] *Combustion Research Facility, Sandia National Laboratories, Livermore, California*

Abstract: Rotational coherence lifetime imaging of nitrogen in the vicinity of a laser-induced air spark at atmospheric pressure is measured by coherent anti-Stokes Raman spectroscopy (CARS) with Frequency Recognition Algorithm for Multiple Exposures (FRAME). A 25-fs laser sheet at 800 nm is employed as pump and Stokes beams while the coherence lifetime is extracted by exponentially fitting of the 2D CARS signals which are probed by uniquely modulated picosecond laser pulses at 532 nm.

Keywords: lifetime imaging, 2D CARS, FRAME, laser-induced air spark

1.Introduction

Laser plasmas have been applied in various applications since the invention of lasers in 1960s. In particular, the lifecycle of air sparks generated by an Nd:YAG laser has been studied by Harilal at el. [1] with shadowgraphy and emission spectroscopy, where the generation, expansion and collapse of a spark is presented. However, much of the physical and chemical process of the plasmas in either solid or gaseous mediums are still unclear due to the many complex and transient non-linear phenomena induced by the intensive electrical field.

Coherent anti-Stokes Raman spectroscopy (CARS) has been widely used for accurate temperature measurements and species concentrations in complex gaseous systems [2]. As the CARS signal is coherent, i.e., has high signal collection efficiency, it is extremely advantageous for optical diagnostics of plasma-related phenomena as the strong and broadband plasma emission can be efficiently suppressed by a short camera gate.

Molecular dynamics of plasma-related phenomena are transient and usually too fast to be captured with existing highspeed camera systems. Frequency Recognition Algorithm for Multiple Exposures (FRAME) [3] has been developed to be able to capture ultrafast events on the GHz to THz timescale by labelling the ultrafast light pulses with spatial intensity modulations such that a computer algorithm can subsequently extract the desired information.

In this work, FRAME is implemented into fs/ps hybrid CARS to capture the coherence lifetime of rotationally excited nitrogen around an air spark generated by an Nd:YAG laser. A rotational coherence state of nitrogen is established by a femtosecond pulse at 800 nm, while the state is probed by spatially coded picosecond laser beams (532 nm) at different delays, yielding two spatially modulated CARS signals within a single camera exposure. Exponentially fitting the intensity of said CARS signals at each pixel yields a coherence lifetime map of the nitrogen within the probe volume.

2.Experiment setup

The optical setup of the experiment is shown in Fig. 1a. An air spark is produced by focusing a 5-ns laser pulse at the probe volume with a spherical lens of f = 100 mm. The laser pulse is generated by a standard Nd:YAG laser system (Q smart 850) with a pulse energy of ~30 mJ at 532

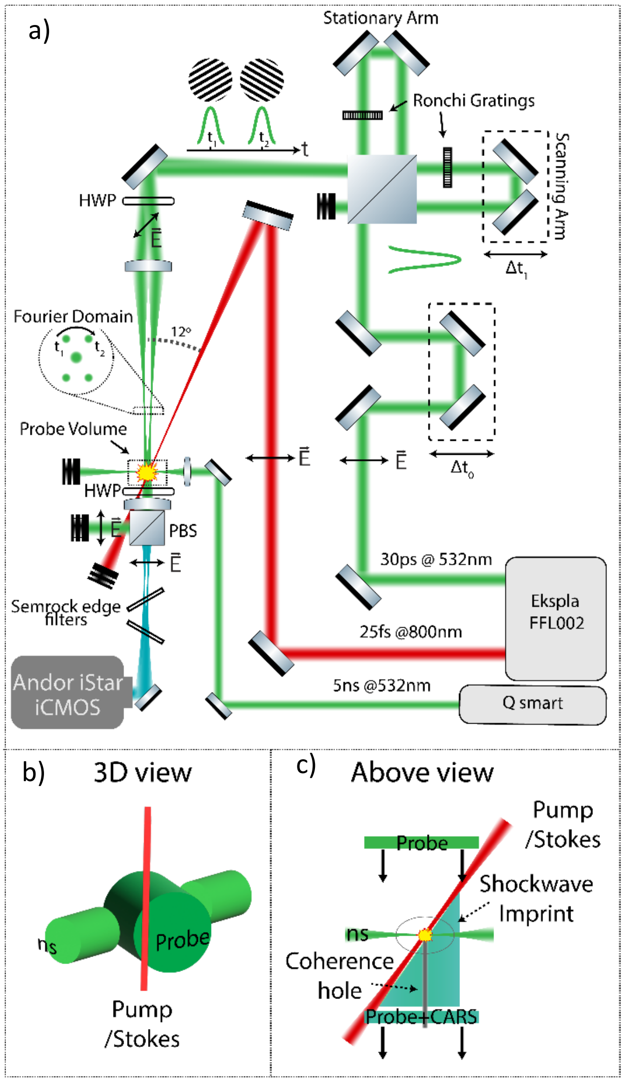

Fig. 1. Optical arrangement of the setup and the geometry of the beams at the probe volume.

nm. The 25-fs pump/Stokes beam and 30-ps probe beam are both generated by Ekspla FFL002 laser system. The probe beams are guided to be perpendicular to the plasma while the pump/Stokes beam has an angle of 12° to the probe. The geometry of the beams is demonstrated in Fig. 1b and 1c. The CARS signal, which carries the molecular information of the nitrogen, will be generated from the intersection where the pump/Stokes and probe overlap. The intensity modulation of each probe beams is created by Ronchi gratings with a line density of 10 lines/mm. More detailed discussion about the creation of the frequency modulation can be found on previous publications by the authors, such as [3].

As the CARS signals have a similar direction as the probe beams, a polarizing beam splitter and two slightly tilted Semrock edge filters (526 nm) are applied to filter out the probe wavelength. The CARS signals are then captured by an Andor iCMOS camera with a gate width of 5 ns. A 10 MHz signal generator (BNC 575) is used to synchronize the lasers while transitional stages are inserted in the probe beams for accurate adjustment with a temporal resolution of 0.5 ps. It is noted that although only two probe beams are used in this initial demonstration, the energy of the picosecond laser is strong enough to be split into at least four beams while keeping a moderate level of signal intensity of the CARS signal.

3. Methodology

As shown in Fig. 2a, CARS involves four beams: a pump, a Stokes, a probe and the CARS signal. If the frequency difference between the pump and Stokes beam matches the Raman shift of certain molecule, a coherence will be formed between different rotational energy levels. This can be done either by two narrowband laser beams with a set frequency difference or by one broadband pulse with a bandwidth broad enough to cover several rotational levels. In this work, a femtosecond laser beam with a FWHM bandwidth of ~40 nm acts both as the pump and the Stokes beam to establish the rotational coherence of the nitrogen.

To measure the lifetime of the coherence of nitrogen that has been established by the pump/Stokes, probe beams are sent consequently with difference time intervals after the pump/Stokes. The temporal profile of the plasma generated by a nanosecond laser, the femtosecond pump/Stokes beam and two probe beams are illustrated in Fig. 2b.

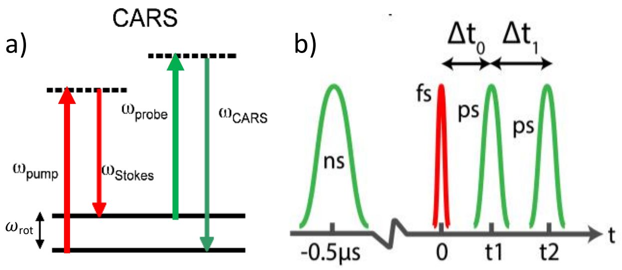

Fig. 2. (a) Energy diagram of CARS. (b) Sketch of the temporal profile of the plasma, pump/Stokes beam and probe beams.

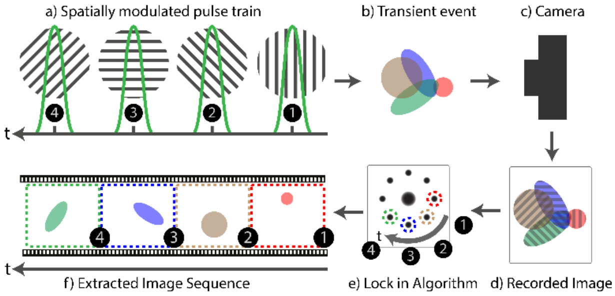

Fig. 3. Illustration of the FRAME technique.

Since the coherence lifetime of nitrogen is reported to be around 65 ps with J = 2-15 [4], the time difference between each probe and CARS pair can only be few tens of picoseconds, meaning that GHz frame rate is required to capture each CARS signals separately. In this work, high frame rate videography is achieved by a technique called FRAME.

As shown in Fig. 3, FRAME is implemented as such: (a) the intensity profile of each probe beams is modulated with a sinusoidal pattern with different orientations. (b) The probe beams are sent into the probe volume and all CARS signals are captured within a single camera exposure (c). The recorded CARS signals will be tagged with the same intensity modulation as their corresponding probe beams (d). To distinguish each signal from the recorded image, a lock-in algorithm is performed (e) resulting in an extracted ultrafast video (f). Finally, a lifetime image can be extracted by exponentially fitting the signal intensity on each pixel of the captured video. More details about how the lock-in algorithm is applied will be discussed in the Sec. 4.1.

4.Results

4.1 Lifetime scan of the CARS signal

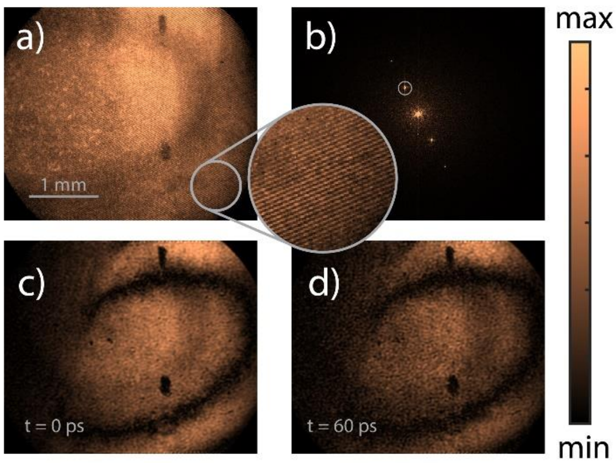

Fig. 4. (a) Raw 2D CARS image with intensity modulation. (b) Fourier domain of the raw signal. (c-d) Extracted images of the two CARS signals with a time difference of 0 ps and 60 ps after the pump/Stokes.

To validate the method, an experiment was first carried out without any plasma and with only one modulated probe beam. The raw image captured by the camera is shown on Fig. 4a, with a magnified area for a better demonstration of the modulation of the signal. The Fourier domain of Fig. 4a is shown on Fig. 4b where clusters of frequency components due to the intensity modulation can be clearly observed. To extract the modulated signal, the frequency components of interest is shifted to the centre and filtered out with a Gaussian filter. Thereafter, the temporally resolved images can be extracted by an inverse Fourier transform. The two extracted images, having a time difference of 60 ps are shown on Fig. 4c-d. The CARS signal acquired later in time compared to the pump/Stokes is weaker, indicating a decay of the coherence.

To obtain the rotational coherence lifetime profile of the nitrogen, the time difference between the probe and pump/Stokes, i.e., Δt_0 as in Fig.2b, is scanned by a translation stage. The normalized CARS intensity scanned across 300 ps is plotted in Fig. 5. The exponential fit of the CARS signal is plotted in red in Fig. 5, yielding a coherence lifetime of 80 ps, which is similar as previously reported by others [4].

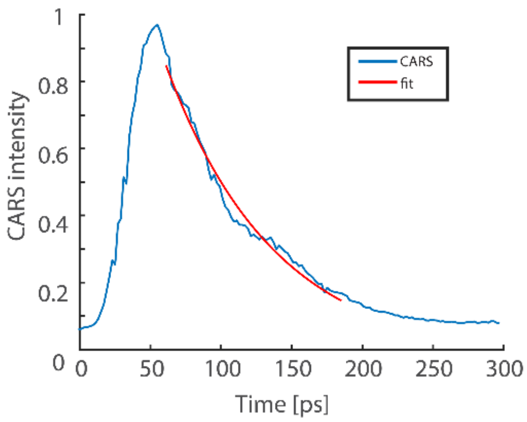

Fig. 5. Plot of the lifetime scan of the integrated CARS signals (blue) and the exponential fit (red).

4.2 Coherence lifetime imaging

Finally, coherence lifetime images are obtained at 0.5 μs, 1 μs and 1.5 μs after an air spark is induced in the probe volume. The raw images captured by the camera and the coherence lifetime maps for each time step are shown in Fig. 6. A growing black hole of the signal is observed from where the air spark is generated, while a shock wave, which has the speed of sound, is captured around the hole. Shock waves with similar shapes have also been reported in [1] whereas the black hole is unique for the CARS signal. While further investigations are required to understand the physical and chemical interactions inside the hole, a possible assumption could be that the energy/temperature of the nitrogen is too high to be probed by the bandwidth of the experiment as the nitrogen is rotationally excited to high J numbers.

For a better exponential fitting, one of the probe arms is tuned close to the position from which the strongest CARS signal is yielded, i.e., temporally overlap with the pump/Stokes, and is thus referred to as the stationary arm. The other arm, which is referred to as the scanning arm will be scanned around the stationary arm. The integrated CARS intensity of the two arms, scanned across 400 ps, is plotted in Fig.6a. The coherence lifetime images are extracted by the frame in which the time difference between the two arms are 120 ps, where the intensity of the scanning arm is about 30% of the stationary arm.

Coherence lifetime images are shown in Fig. 6b, where a threshold is applied if the signal to noise ratio of any extracted images is less than 10. As the coherence lifetime is calculated point by point, the resolution of the lifetime

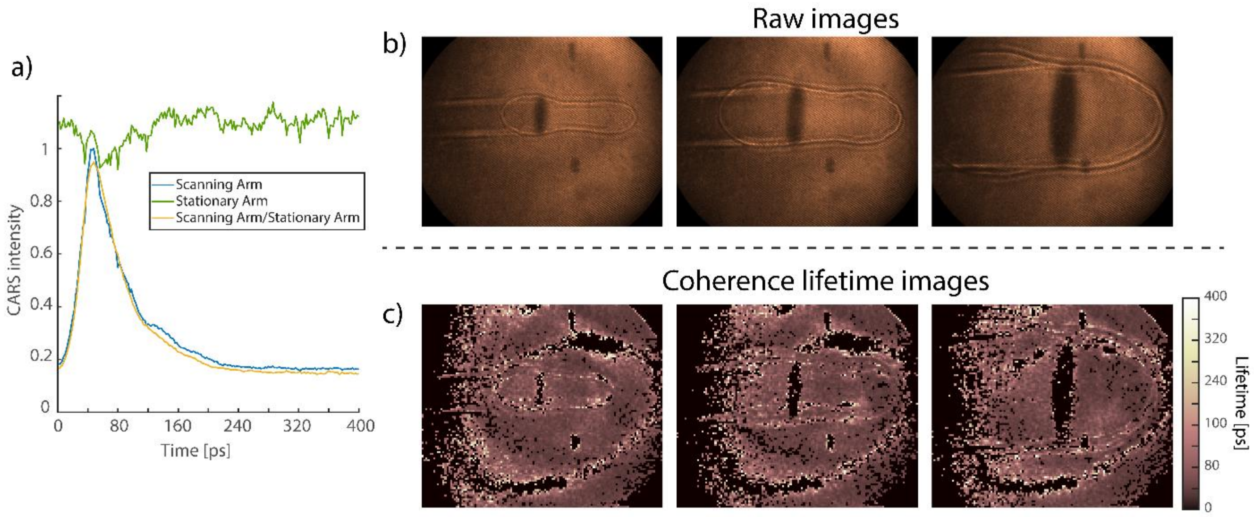

Fig. 6. (a) Plot of the lifetime scan of the integrated CARS signals. Blue: Scanning Arm, Green: Stationary Arm, Yellow: Ratio between the scanning arm and the stationary arm. (b) Raw images and coherence lifetime imaging of nitrogen at 0.5 μs, 1 μs and 1.5 μs after an air spark.

imaging is reduced to 10 % of the raw images (in order to save computing time). As shown in Fig. 6b, the coherence lifetime of nitrogen is mostly similar to what has been measured in Fig. 5, i.e., around 80 ps, while longer lifetime is noticed around the shock wave and the black hole. However, since only two arms are applied in this initial demonstration of the technique, considerable uncertainty can be induced from the exponential fitting. A discussion of the coherence lifetime will be given when four arms are applied.

5.Discussion

In this work, FRAME-enhanced CARS has been developed for coherence lifetime imaging of nitrogen in the vicinity of an air spark. With two modulated beams, a coherence lifetime map is successfully generated at 0.5 μs, 1 μs and 1.5 μs after an air spark. The dynamics of the shock wave around the laser-induced plasma and the measured coherence lifetime of nitrogen both agree with previous reported results by other researchers, while a growing black hole of the CARS signal of nitrogen is observed for the first time. This is also the first time where 2D CARS is used for coherence lifetime imaging. As two more beams will be implemented to the setup, results with better accuracy will be expected in near future.

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References

[1] Harilal, S.S., Brumfield, B.E., Phillips, M.C., Phys. Plasmas 22, 063301 (2015).
[2] Roy, S., Gord, J. R., & Patnaik, A. K., Progress in Energy and Combustion Science, 36(2), 280-306 (2010).
[3] Ehn, A., Bood, J., Li, Z., Berrocal, E., Aldén, M., & Kristensson, E., Light: Science & Applications, 6(9), (2017).
[4] Hosseinnia, A., Ruchkina, M., Ding, P., Bood, J., & Bengtsson, P. E., Proceedings of the Combustion Institute, 38(1), 1843-1850 (2021).