

# Plasma-assisted Deflagration to Detonation Transition in a Microchannel with Hybrid fs/ps Coherent Anti-Stokes Raman Scattering Measurements

Madeline Vorenkamp<sup>a\*</sup>, Scott A. Steinmetz<sup>b</sup>, Timothy Y. Chen<sup>b</sup>, Xingqian Mao<sup>a</sup>, Andrey Starikovskiy<sup>a</sup>, Christopher Kliewer<sup>b</sup>, Yiguang Ju<sup>a</sup>

<sup>a</sup>*Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ 08544, USA*

<sup>b</sup>*Sandia National Laboratories, Livermore, CA 94550*

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## Abstract

This study examines kinetic enhancement by nanosecond dielectric barrier discharge (ns-DBD) plasma on fuel-lean dimethyl ether (DME), oxygen (O<sub>2</sub>), and argon (Ar) premixtures during deflagration to detonation transition (DDT) experiments in a microchannel. Non-equilibrium plasma produces active species and radicals and creates fast and slow heating of a mixture to promote ignition due to electronic and vibrational excitation. Experiments have been conducted to examine the influence of the plasma discharge on the premixture and on the resultant deflagration to detonation transition (DDT) onset time and distance through the use of high speed imaging and one-dimensional, two-beam, femtosecond/picosecond, coherent anti-Stokes Raman scattering (CARS). A high-speed camera is used to trace the time histories of flame front position and velocity and to identify the dynamics and onset of DDT. The results show that plasma discharge can nonlinearly affect the onset time and distance of DDT. It is shown that a small number of plasma discharge pulses prior to ignition result in reduced DDT onset time and distance by 60% and 40%, respectively, when compared to the results without pre-excitation by ns discharges. The results also show that an increase of plasma discharge pulses results in an extended DDT onset time and distance of 224% and 94%, respectively. Time history of the deflagration wave speed of DME and the analysis of ignition timescale under the choking condition of the deflagration front suggest low temperature ignition may play a role for DME near the isobaric choking condition of the burned gas and the DDT. Plasma-induced conversion of the reactive mixture was assessed via the O<sub>2</sub> to CO<sub>2</sub> ratio as measured through fs/ps CARS during the DBD discharges. The present experiments demonstrate the ability of non-equilibrium plasma to alter the chemistry of DME/O<sub>2</sub>/Ar premixtures in order to control DDT for applications in advanced propulsion engines.

**Keywords:** non-equilibrium plasma; deflagration to detonation transition; coherent anti-Stokes Raman scattering; dimethyl ether; low temperature chemistry

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\*Corresponding author.

## 1. Introduction

The dominant form of energy conversion in the United States is still combustion, in 2020 80% of electricity generation was from combustion processes [1]. With the reality of climate change, it is undeniable that combustion processes must become more efficient to help curb carbon emissions. Existing gas turbine engines use a constant pressure Brayton cycle, which limits their thermal efficiency. Advanced power generation using a constant volume cycle, such as a rotary detonation engine (RDE), has the potential to increase efficiency by up to 30% [2]. Unfortunately, instabilities of the detonation wave present a major development challenge which prevents the use of fuels such as those with high hydrogen content.

Detonation can be achieved through a transitional process when the leading shock wave couples with the combustion wave, causing the flame to shift from a subsonic deflagration to a supersonic detonation. Establishing control of this process, commonly referred to as DDT (deflagration-to-detonation transition) would make RDEs and other pulse detonation engines more feasible and energy efficient [3]. In part due to this potential, the dynamics of reaction and shock waves during flame acceleration (FA) and deflagration-to-detonation transition (DDT) under near-limit conditions in small channels have recently received increasing interests [4-15]

Methods of DDT control through kinetic accelerants or energy addition have already begun to be explored. Recent studies at Princeton of the kinetic enhancement of microchannel detonation transition by the addition of ozone to acetylene mixtures, showed dramatic reductions in DDT time (by up to 77.5%) and in onset distance [16]. The DDT limit was also extended from an equivalence ratio of 0.3 to 0.2. The enhancement from ozone can be explained by the Zeldovich hot spot theory and ignition gradient theory [17, 18, 19]. By accelerating ignition delay time using O radical addition from ozone thermal decomposition as well as temperature and concentration gradients, detonation onset is accelerated. The addition of O radicals, stemming from ozone thermal decomposition, accelerates chain branching and the ignition delay time. However, ozone thermally decomposes at temperatures above 450 K, making it difficult to use directly in practical engines with elevated temperatures. Therefore, it is desirable to develop a non-equilibrium plasma discharge to directly initiate and accelerate DDT in-situ. Plasma assisted combustion provides a great opportunity to accelerate ignition to detonation transition and to increase detonation stability.

Over the last two decades, plasma has demonstrated potential to enhance combustion process, to reduce emissions, and to enhance combustion in advanced engines and improve fuel reforming processes [20,21]. Fuel oxidation is carried out through a chain mechanism, and by accelerating

this mechanism it is possible to reduce ignition delay times kinetically. This can be done through the dissociation and excitation of molecules and active species production by an electron impact, for example, in a nonequilibrium dielectric barrier discharge [20-22]. Non equilibrium plasmas have demonstrated the ability to reduce ignition delay, and temperature [22-26]. However, there is still much work to be carried out to understand the effect of plasma assisted combustion on detonation transition for use in applied engines, including the use of specifically a ns-DBD non-equilibrium plasma for DDT enhancement.

In this study we aim to examine the influence of an ns-DBD plasma discharge on a combustible premixture and on the resultant deflagration to detonation transition (DDT) onset time and distance through the use of high speed imaging and one-dimensional (1-D), two beam, femtosecond/picosecond, coherent anti-Stokes Raman scattering (CARS). The plasma-induced conversion of the reactive mixture to products yields an important benchmark for the chemical activity of the DBD plasma for comparison to plasma-assisted chemistry models. Employing fs/fs CARS in-situ, we characterize this conversion by monitoring the ratio of O<sub>2</sub> to CO<sub>2</sub> as a function of number of ns-DBD pulses. Additionally, the extent of rotational and vibrational heating of the O<sub>2</sub> reactant, which may play a role in kinetic enhancement, is directly monitored through the CARS experiments [27].

For this study, a uniform nanosecond (ns) dielectric barrier discharge (ns-DBD) plasma is applied across a 1 mm tall, 4 mm wide, ~60 cm long combustion channel filled with dimethyl ether/oxygen/argon (DME/O<sub>2</sub>/Ar). A high speed camera imaging the combustion chemiluminescence is used to trace the time histories of flame front position and velocity. The applied ns-DBD plasma is studied with hybrid fs/fs one dimensional coherent anti-Stokes Raman scattering (1D CARS) measurements of rotational and vibrational temperature as well as through the consumption of O<sub>2</sub> and production of CO<sub>2</sub>.

## 2. Experimental Methods

The plasma microchannel has a rectangular cross section of 1x4 mm<sup>2</sup> and a length of 600 mm. A dielectric barrier discharge was formed across the channel between copper electrodes isolated from the discharge gap by a Kapton® film of 0.035 mm thickness. The distance between the electrodes was 1 mm, the width of the electrodes corresponded to the width of the channel (4 mm), and the length of the electrodes was 500 mm. A spark gap was installed at a distance of 50 mm from the edge of the DBD electrodes to initiate a combustion wave in the channel. The channel is filled through a needle feed-

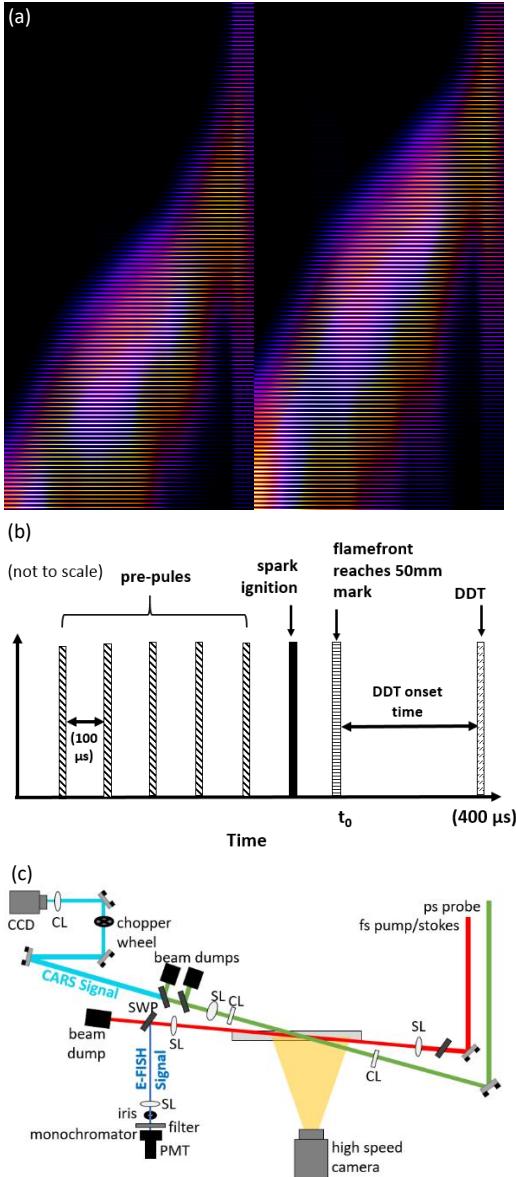


Fig.1. (a)Left panel, stacked fast frame images of flame front progression without (left) and with (right) plasma pre-pulses (b) Timing sequence of the plasma discharge, ignition, and onset of DDT. (c) Experimental layout. CL: cylindrical lens, SL: spherical lens, PMT: photomultiplier tube, SWP: short-wave pass filter, CCD: charge couple device camera

through at one end, the opposite side of the channel has a needle feed-through serving as a vent valve. Windows are fused silica to allow optical diagnostics. The assembly is clamped together with fasteners

through the frame pieces, nylon insulating tubes shield the fasteners.

The channel is mounted on a translation stage on an optical table where gas connections bring in the fuel mixture. A schematic diagram of the experimental setup is shown in Fig. 1c. For operation, the fuel and oxidizer are fed through a pressure-regulated flow controller and a mass flow controller, and then mixed over a ~5m length of tubing. For filling the test section, the channel is initially pumped down to the single Torr level with an oil-free mechanical vacuum pump, the pump is then isolated and the premixture is pulled into the channel filling to atmospheric pressure.

The plasma discharge is generated by a DC high voltage supply and pulser (Eagle Harbor, NSP-120-20-N), and is controlled by a signal generator. A trigger initiates a sequence of 10 kV, 10 kHz, 200 ns pulses ahead of ignition. A variable number of pulses may be applied, with the final pulse 100  $\mu$ s prior to spark ignition. The timing is highlighted in Figure 1b.

## 2.1 High Speed Imaging

A Phantom v1610 high-speed CMOS camera recording at 250 kHz is used to image the chemiluminescence emitted from the flame-front and trace its propagation position and velocity, as well as to locate the deflagration to detonation transition. Once the imaging sequence is complete, the data is processed through Matlab, and regions of diagnostic interest are identified.

## 2.2 Hybrid fs/ps Coherent Anti-Raman Scattering

One-dimensional (1-D), two beam, femtosecond/picosecond, coherent anti-Stokes Raman scattering (fs/ps CARS) technique is employed to probe inside the microchannel. Given the small cell dimensions, the use of fs/ps CARS helps to mitigate the effect of fluorescence and damage in the windows and allows for the direct tracking of rotational/vibrational temperatures and conversion of the reactive mixture to CO<sub>2</sub> through the action of the plasma. Previous work using nanosecond pure-rotational CARS in an open burner demonstrated the ability to simultaneously track concentrations of O<sub>2</sub>, N<sub>2</sub>, CO and CO<sub>2</sub> in complicated mixtures [28]. The theory and general methods of fs/ps CARS is detailed in [29] and as such, will not be included here. The geometry of the microchannel poses a significant challenge in terms of optical access. The probe region of interest is 1mm tall, and 4 mm wide with 6 mm thick windows immediately adjacent. Due to the proximity of the windows to the probe region, substantial white light is generated by the fs beam at energies high enough to produce CARS if approaching the cell in a simple orthogonal orientation. This ultimately led to the development and use of an optical configuration similar to that employed in [30] with the exception of using the two-

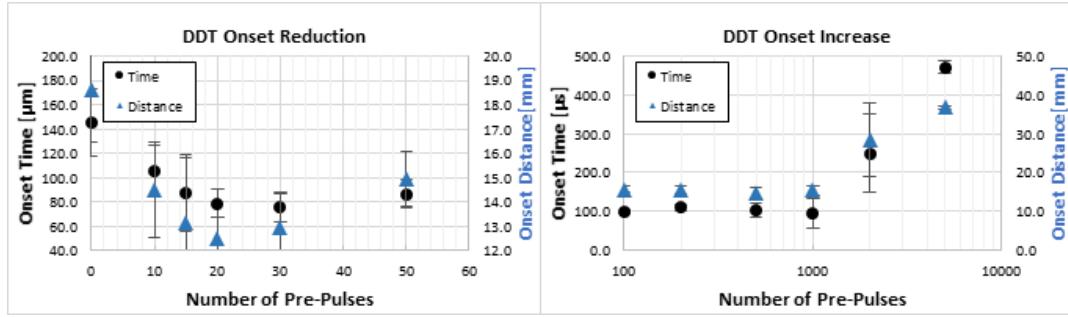


Fig.2. Average DDT onset reduction (left) and increase (right) in distance and time with error bars for standard deviation.

beam phase matching approach [31]. The ps probe is focused to a horizontal sheet with a cylindrical lens of 500 mm focal length, and the fs laser beam is focused to a point using a 700 mm spherical lens which intersects the probe sheet going across it, forming a 1-D region approximately parallel to the length of the microchannel that emits the CARS signal. The ps and fs beams enter the channel at 10° and 5° respectively. These shallow incidence angles effectively increase the channel width, placing the focal point at a greater path length from the windows. Despite the fact that a significant percentage of the beam energy is thus reflected at these angles, the probe region receives a higher amount of energy than it would from beams with energy sufficient to avoid white light, at steeper angles. Additionally, due to the significant intensity of reflected beams, the channel was angled in the azimuthal direction to avoid generation of CARS from reflected beams outside of the channel, which would copropagate with the desired signal.

The generated 1D CARS signal was relay imaged to the entrance slit of a 550mm spectrometer (Horiba iHR550). The signal was separated from the residual probe beam by two angle-tuned short-pass filters (Semrock 561RU). A 2400 line/mm grating dispersed the signal onto a CCD (Andor Newton). For the assessment of vibrational heating, the fifth-order diffraction from the grating was used to increase spectral resolution. The first order diffraction resulted in a spectral resolution of 0.70 cm<sup>-1</sup> FWHM, while the higher order diffraction yielded a resolution of 0.56 cm<sup>-1</sup> FWHM. The small improvement in the slit function allowed for improved resolution of rotational transitions arising from the ground vibrational state and first vibrational state as discussed later.

Once the deflagration regions of interest had been identified through the high speed imaging, the channel was positioned accordingly with the translation stage, relative to the fixed diagnostics. Single-shot 1D CARS images were collected with probe energy of 2 mJ spread over a 1D line of ~2 cm and pump/stokes energy of 200 μJ inside the channel.

### 3. Results and Discussion

#### 3.1 High Speed Imaging of DDT

For this study, a test case of zero ns-DBD pulses prior to ignition, as well as cases of 10, 15, 20, 30, 50, 100, 200, 500, 1000, 2000, and 5000 pre-pulses are studied. Two batches (trials of 6) are conducted for the cases with fewer pulses (10-30) and one batch for the higher pulse cases (50-5000). Four batches are conducted for the no-plasma case. These batches are cycled to ensure repeatability. The initial time used in determining the time to onset of DDT is defined as the time at which the flame front has propagated 50 mm such that the onset time is the time required for the flamefront to travel from the 50 mm mark to the point of DDT, and the onset distance is the distance traveled after 50 mm to the point of DDT, this is to ensure the results are independent of variations in initial ignition kernel development [8]. The resultant average onset time and distance are plotted in Fig. 2. It is seen that the cases of 15 to 30 pre-pulses provide the strongest plasma enhancement of DDT. At peak, onset distance is reduced from an average of 18.6 cm to 12.5 cm, a 33% reduction. The onset time is reduced from 146 μs to 76 μs, a 48% reduction. Interestingly, with a further increase of pre-pulse numbers, we start to see the DDT onset time and distances extended by up to 224% and 94%. This suggests that there is an optimum pre-pulse number in plasma assisted DDT.

To better understand these results, it is helpful to examine the typical velocities of the deflagration fronts over time for the various plasma conditions in Fig. 3. In the control case with no plasma pulses, two distinct velocity peaks occur, one around 70 μs and one around 115 μs. This is unique for DME, which has low temperature chemistry, and was not observed for C<sub>2</sub>H<sub>2</sub> DDT experiments. These are suspected to be caused by the two-stage ignition chemistry of DME. The first stage may be caused due to the presence of low temperature chemistry and the heat loss near the wall, and the second due to the high temperature auto-ignition and DDT where a steep overdriven velocity profile is observed before the front settles down to

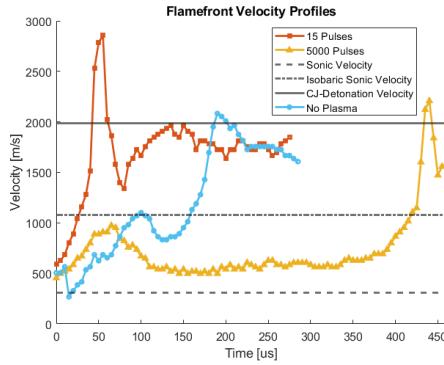


Fig.3. Flame-front velocity as a function of time for the control case, no plasma pulses, as well as the cases resulting in the greatest reduction and increase in DDT onset distance and time, 15 pulses and 5000 pulses respectively.

Chapman-Jouguet velocity. When 15 ns-DBD pulses are applied, exhibiting the highest degree of DDT enhancement, we see that this gap between stages closes and the high temperature chemistry stage proceeds immediately from the low temperature chemistry stage. The region of negative acceleration is effectively eliminated and there is only one peak in the wave velocity profile before detonation. This is probably due to the plasma assisted ignition chemistry. When 5000 pulses are applied to the mixture prior to ignition, two velocity peaks are once again observed, and the second acceleration event is greatly delayed. This is caused by the over consumption of the fuel by the pre-ignition plasma discharges and thus the reduction of the chemical heat release in the deflagration process. This finding underscores the importance of assessing the degree of chemical conversion induced by the plasma pulses, which we elucidate through fs/ps CARS as discussed below.

To understand the physics and chemistry of DDT of DME, in Fig. 4, we show an example of a DDT time history without plasma. Here both the flame front's velocity and the two-stage ignition delay time are plotted against the time elapsed since the flame reached the 5 cm mark of the microchannel. We see that the initial velocity spike appears to peak around the isobaric sonic velocity, before accelerating and settling around the C-J velocity at DDT. The match between the first flame front peak velocity and the isobaric sonic speed suggests that the first peak occurs near or at the critical conditions that the burned gas after flame front is choked and enables strong pressure-velocity coupling if there is a sufficient chemical heat release rate at the flame front. Interestingly, near this choking condition, we see the presence of low temperature chemistry in the region, which supports that near the first spike, the low temperature chemistry may affect the DDT processes

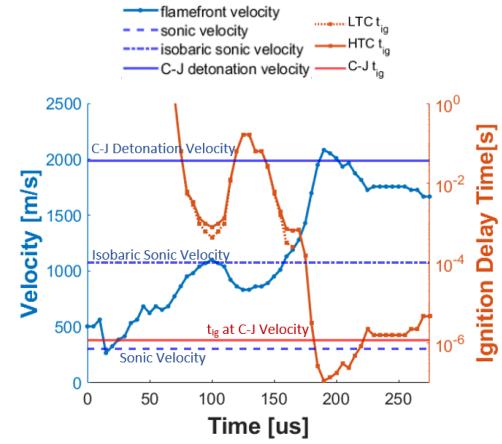


Fig.4. A typical flamefront velocity profile as a function of time plotted against CHEMKIN simulations of low temperature chemistry (LTC) and high temperature chemistry (HTC) ignition delay time.

shown in Fig. 3. It is also seen that near 150  $\mu$ s, as the flame front speed increases, there is a low temperature chemistry induced negative temperature coefficient phenomenon. Following this, hot ignition occurs and ignition delay time becomes so short that the burned gas is choked again, leading to a strong ignition and pressure coupling to detonation transition. In addition, it is also seen that after the DDT, the ignition delay time predicted by the measured wave front speed is very close the ignition delay time of the C-J condition. This suggests that the analysis using the ignition delay time at the wave front velocity helps to understand the timescale and chemistry in DDT. Moreover, the experimental C-J velocity is slightly below the theoretical velocity due to losses like friction and heat to the wall.

From the analysis in Fig. 4, in the cases with fewer pre-ignition plasma pulses, we see that plasma enhances DDT because of the shortening of the ignition delay time by the active species production in plasma. However, in the cases with a higher number of pulses, although the ignition delay time is reduced, due to the fuel oxidation in plasma discharge and the heat release in the deflagration wave front is reduced which weakens coupling between combustion and the compression wave necessary for DDT. As a result, the DDT is delayed. The nonlinear plasma enhancement of DDT is a direct result of the competition between the kinetic enhancement of ignition and the weakening of heat release rate due to pre-ignition plasma assisted fuel oxidation, and can ultimately be leveraged to control DDT onset time and position.

### 3.2 Hybrid fs/ps Coherent Anti-Raman Scattering

To assess the chemical action of the ns-DBD plasma pulses on the combustible mixture, fs/ps

CARS was implemented to follow the conversion of reactants to products. This is achieved by recording a time-resolved set of CARS spectra for a given number of plasma pulses and evaluating for relative  $[CO_2]/[O_2]$  ratio according to the following equation:

$$\frac{[CO_2]}{[O_2]} = \sqrt{\frac{S_{CO_2}}{S_{O_2}}} \times \frac{\sigma_{O_2}}{\sigma_{CO_2}} \times e^{-t(\frac{1}{2\tau_{O_2}} - \frac{1}{2\tau_{CO_2}})} \quad (1)$$

where  $S$  is the integrated CARS intensity of the particular species,  $\sigma$  is the relevant Raman cross section,  $t$  is the probe delay time and  $\tau$  is the dephasing constant. In the current study, all data were collected at a constant probe delay of 200 ps at near 300 K. Thus, both the ratio of Raman cross sections and dephasing correction terms of Eq. 1 are constant. In this case, calibration experiments in a cell with known pressure ratio of  $CO_2$  and  $O_2$  enable the absolute calibration of the evaluated concentration ratio. As can be seen in Fig. 5(a), the series of ns-DBD plasma pulses clearly results in the production of  $CO_2$ . Fig. 5(b) displays the rate of reaction as a function of DBD pulse number. This data is in agreement with the

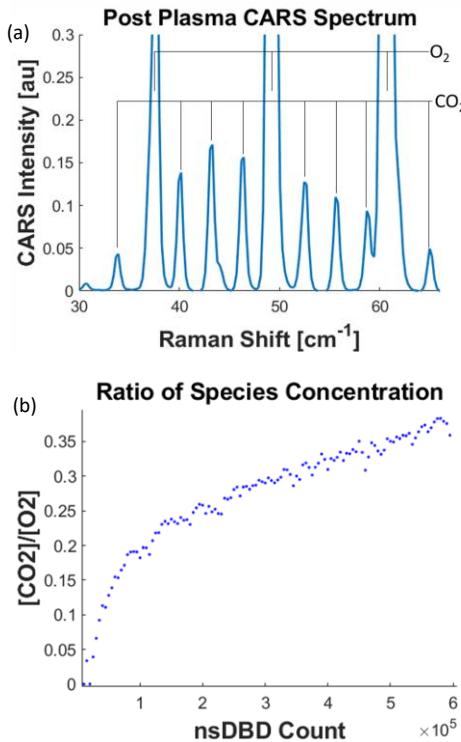


Fig. 5. (a) fs/ps CARS spectra demonstrating the growth of  $CO_2$  and depletion of  $O_2$  during 1 minute of ns-DBD pulses operated at 10 kHz. (b) Calibrated concentration ratio as a function of DBD pulse number in the reactive mixture.

overall observation from Fig. 2 that plasma-induced fuel oxidation eventually increases DDT onset time.

In order to assess the plasma-induced molecular vibrational and/or rotational heating of  $O_2$ , the CARS spectra were evaluated for both rotational and vibrational heating 200 nanoseconds after the end of the plasma pulses. Because of rotation-vibration coupling, rotational transitions in vibrationally excited  $O_2$  molecules are slightly red-shifted relative to the rotational transitions of molecules in the ground

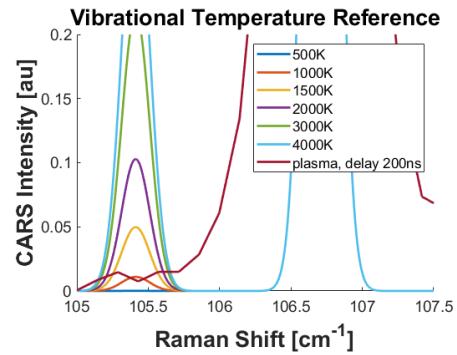


Fig. 6. Pure-rotational fs/ps CARS spectrum of molecular  $O_2$  200 ns after a ns-DBD plasma pulse. Only the  $N''=17$  transition is displayed. Simulations of the spectral location and intensity of the rotational transition in  $v=1$  clearly indicate that no observable vibrational heating is induced above 1000 K vibrational temperature.

vibrational state. This shift may be used to evaluate for plasma-induced vibrational nonequilibrium as was recently demonstrated for  $N_2$  in a bare pin-to-pin discharge [27]. In that work, a minimum vibrational heating of approximately 1000 K was required to evaluate the population in the first vibrationally excited state. Because of the close spacing of the transitions, the 5<sup>th</sup> order diffraction from the 2400 groove/mm grating was used to enhance the spectral resolution of the spectrometer. As seen in Fig. 6, no significant vibrational heating of molecular  $O_2$  is induced by the plasma in this system. Furthermore, the evaluated rotational temperatures increased by ~30 K following plasma discharge, indicating that this ns-DBD plasma is not significantly vibrationally exciting the  $O_2$  molecules. Thus, the decreased DDT onset times observed does not result from vibrational excitation, and more likely is caused by the production of reactive species that enhance low temperature chemistry.

#### 4. Conclusion

Deflagration to detonation transition experiments in DME/ $O_2$ /Ar mixture of atmospheric pressure have

been carried out. The results show that transversal ns-DBD can chemically sensitize the DME mixture and accelerate the DDT transition both in space and time.

The results show that plasma discharge can nonlinearly affect the onset time and distance of DDT. It is shown that a small number of plasma discharge pulses prior to ignition result in reduced DDT onset time and distance while a large number of pre-ignition discharges will delay the DDT. This nonlinear effect is explained by the competition between the plasma enhanced kinetic effect on ignition and the reduced heat release rate of the wave front due to the partial fuel oxidation with excessive pre-ignition plasma discharge. For DME mixtures, two distinct flame front peaks were observed. The present experiments and analysis suggest that this unique feature may be caused by the low temperature chemistry of DME.

The present study demonstrates a quantitative effect of the plasma on the premixture temperature

and species concentration. In-situ fs/fs CARS measurements evaluated the plasma-induced rate of conversion to  $\text{CO}_2$  and determined that minimal vibrational heating of molecular  $\text{O}_2$  is induced.

The ability to control DDT with plasmas is promising for the development of next generation rocket engines and opens the opportunity to demonstrate further improvement in lean burn flame stability, low temperature fuel oxidation and processing, as well as in emission reduction.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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