

Cyanide Laser-Plasma Spectroscopy in a Flowing Gaseous Mixture

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ABSTRACT: This article communicates measurements of diatomic molecular cyanide (CN) in a flowing gas mixture. The mixture is composed of 1:1 molar carbon dioxide and nitrogen flowing at a rate of 100 mL per minute. The laser plasma is generated with Q-switched IR irradiance that is a few times above the optical breakdown threshold for the mixture. This work reports recorded spatio-temporal spectral maps. Detailed data curation reveals density, temperature profiles from the line-of-sight measurements. In addition, Abel inversion maps serve the purpose of inferences about the expanding shockwave and expanding kernel of the plasma.

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1. INTRODUCTION

Cyanide (CN) spectra produced from molecular recombination spectra are measured following optical breakdown in gases for time delays of the order of 100 ns after the end of the laser pulse [1]. CN plasma dynamics are observed by using gas mixtures. Applications of cyanide spectroscopy include potential medical and forensic applications [2].

In molecular spectroscopy, the molecular Hamiltonian determines the overall framework of the molecular emissions. The Hamiltonian accounts for electronic, vibrational, and rotational components due to vibrations and rotations associated with molecules. Additionally, the total molecular Hamiltonian contains influences due to molecular nuclei motion and nucleonic spins. This Hamiltonian consists of quantized rotations and vibrations. Spectral lines produced by vibrations and rotations correlate to transitions between quantum numbers associated with vibrations and rotations. The principal idea of a molecular emission is that there is a separate potential energy curve for each electronic state. Quantized vibrations represent splitting of these potentials and rotational splitting of these vibrations occur. Figure 1 shows the plotting of the potentials, V , versus the internuclear separation coordinate R . The potentials shown in Figure 1 are known as Morse potentials, which results from the work of Morse [3]. Electronic transitions, vibrational transitions, and rotational transitions represent the organizational structure of the spectral emission, respectively. Due to vibrational transitions being similar in electronic position, band like structures are formed.

Cyanide (CN) is a diatomic molecule and has two characteristic electronic systems. These two systems are the violet ($B^2\Sigma^+ - X^2\Sigma^+$, 344–460 nm) and the red ($A^2\Pi - X^2\Sigma^+$, 437–1500 nm), where $X^2\Sigma^+$ state represents the ground state [4]. In this notation A, B, and X represent the potential curves, 2 represents the spin of +1/2 (doublet), Σ and Π represent the angular or orbital quantum number $l=0, 1$ respectively, and + represents the unchanged symmetry

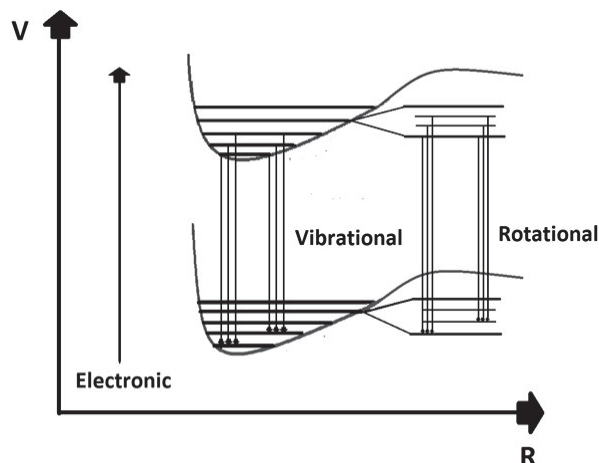


Figure 1: Electronic, vibrational, and rotational spectra as a result of molecular potentials between two electronic levels.

For this work, the CN violet-band $\Delta v=0$ system is of interest and these emissions represent vibrational transitions from a higher energy potential ($B^2\Sigma^+$) to the ground state, where $\Delta v=0$ means the difference between the vibrational quantum numbers is 0. As shown in Figure 2, if the CN molecule is in a vibrational state in the higher potential represented by vibrational quantum number $v=1$, it can only transition to the vibrational state in the ground state also represented by quantum number $v=1$, which would result in the (1,1) band. The CN violet system consists of vibrational bands (0,0), (1,1), (2,2), (3,3) and (4,4), which are 388.34 nm, 387.14 nm, 386.19 nm, 385.47 nm, and 385.09 nm, respectively. In addition to the vibrational states, rotational states exist for the CN violet system which give the system its fine structure.

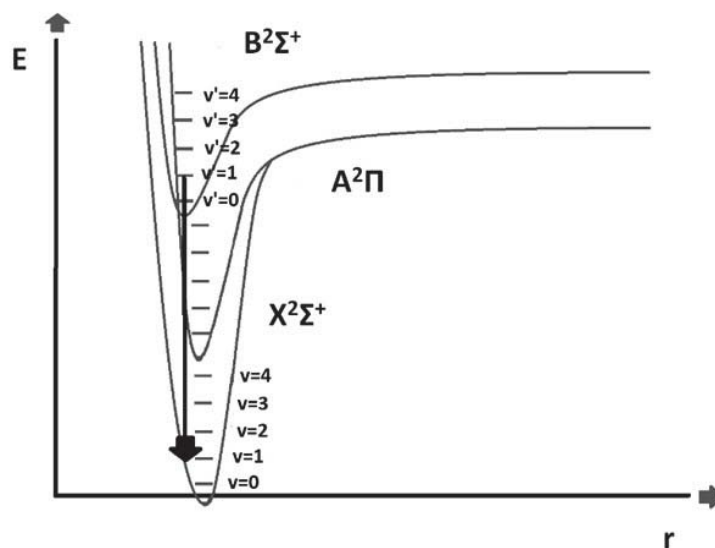


Figure 2: Potential curves of CN.

2. EXPERIMENTAL DETAILS

A set of flowing gas mixture measurements were performed using an experimental setup similar to that of a fixed gas mixture [2], however, the arrangement for laser induced breakdown spectroscopy is included in the next paragraph. A flowmeter (Cole-Parmer Instrument Company model FM112-02ST, USA) was connected to the gas outlet instead of the differential pressure gauge as mentioned in the fixed gas mixture experimental setup. The chamber was supplied with a constant flow of ultrahigh purity N_2 and research-grade CO_2 gas mixture. A flow rate of 100 mL per min of the mixture entered and left the cell, which was monitored by the flowmeter as seen in Figure 3.

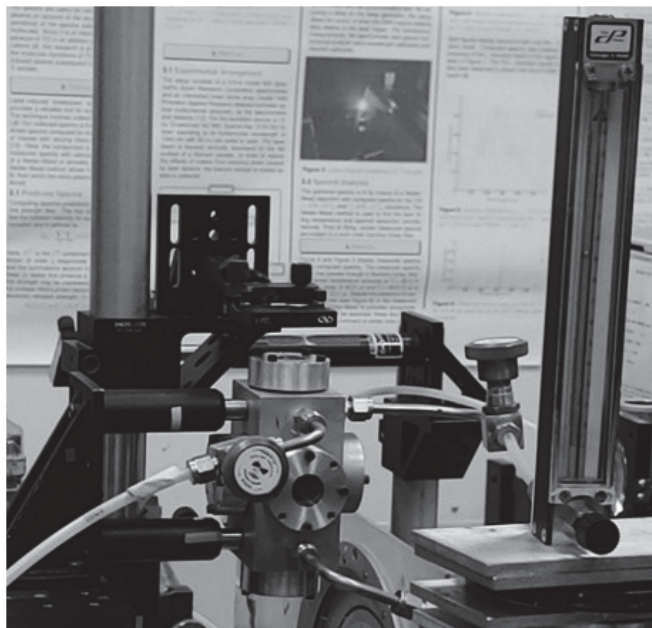


Figure 3: Picture of the gas chamber and flowmeter

Inside the chamber, optical breakdown was created at a rate of 10 Hz using a laser beam focused with $f/5$ optics from the top or parallel to the slit. Pixels of the ICCD were binned in four-pixel tracks along the slit direction, resulting in the production of 256 spectra for each time delay. Recordings of spectra were performed with and without the Order-Sorting Filter consisting of 100 accumulations collected for 21 time delays at 250 ns steps.

The standard experimental components are used for laser-induced breakdown spectroscopy and have been summarized previously, e.g., see Ref. [2], but are included for completeness. The experimental arrangement consists of a set of components typical for time-resolved, laser-induced optical emission spectroscopy, or nanosecond laser-induced breakdown spectroscopy (LIBS) [5]. Primary instrumentations include a Q-switched Nd: YAG device (Quantel model Q-smart 850) operated at the fundamental wavelength of 1064-nm to produce full-width-at-half-maximum 6-ns laser radiation with an energy of 850 mJ per pulse, a laboratory type Czerny-Turner spectrometer (Jobin Yvon model HR 640) with a 0.64 m focal length and equipped with a 1200 grooves/mm grating, an intensified charge coupled device (Andor Technology model iStar DH334T-25U-03) for recording of temporally and spatially resolved spectral data, a laboratory chamber or cell with inlet and outlet ports together with a vacuum system, electronic components for synchronization, and various optical elements for beam shaping, steering and focusing. Figure 4 displays the principal schematic of the experimental arrangement. The laser beam is focused into the cell through which the 1:1 molar carbon dioxide and nitrogen mixture is flown.

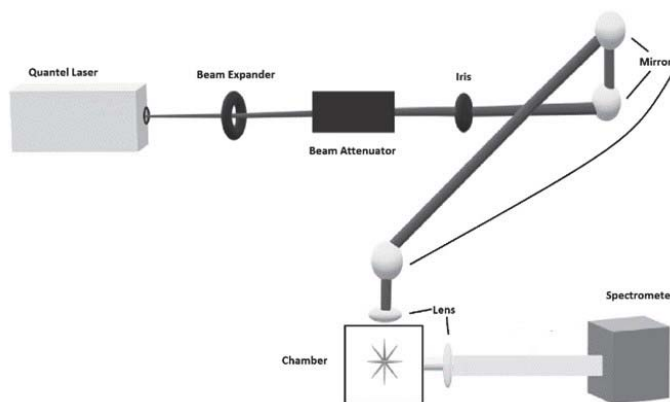


Figure 4: Modular schematic of laser-induced breakdown experiment

For the generation of optical breakdown micro-plasma, a singlet lens (Thorlabs model LA1509-C) is used close to the top entrance window of one arm of the chamber containing the 1:1 $\text{CO}_2\text{:N}_2$ gas mixture held at 3.04×10^5 Pascal (Airgas ultra-high purity N_2 and research grade CO_2). For 1:1 imaging of the plasma onto the $100 \mu\text{m}$ spectrometer slit, a fused silica plano-convex lens (Thorlabs model LA4545) is employed. For the CN experiments, the laser pulse energy is attenuated with beamsplitters and apertures from 850 to 170 mJ/pulse.

3. RESULTS AND DISCUSSION

3.1 Spatially resolved spectra

Optical breakdown was generated inside the chamber at a rate of 10 Hz, with the laser beam focused with $f/5$ optics from the top, or parallel to the slit. The detector pixels are binned in 4 tracks along the slit direction, resulting in obtaining 256 spectra for each time delay. Recording of measurements consist of 100 accumulations collected for 21 time delays at 250 ns steps. Figures 5 to 7 illustrate recorded spatio-temporal spectra that were recorded along the line-of-sight for a 1:1 molar $\text{CO}_2\text{:N}_2$ flowing gas mixture with a flow rate of 100 mL/min, and are accumulated over 100 individual laser-plasma events and the CI 193.09 nm atomic carbon line as identified in previous work [6 – 8]. Similar to the gaseous mixture of fixed volume, the CN violet system $\text{B}^2\Sigma^+ - \text{X}^2\Sigma^+$ vibrational bands of (0,0), (1,1), (2,2), (3,3) and (4,4) are clearly visible. Additionally, the overlap of the CI 193.09 nm atomic carbon line in second order and the (2,2) band head is present as in the gaseous mixture of fixed volume. At time delays greater than 2.2 μs , the CI 193.09 nm atomic carbon line in second order appears to dissipate away.

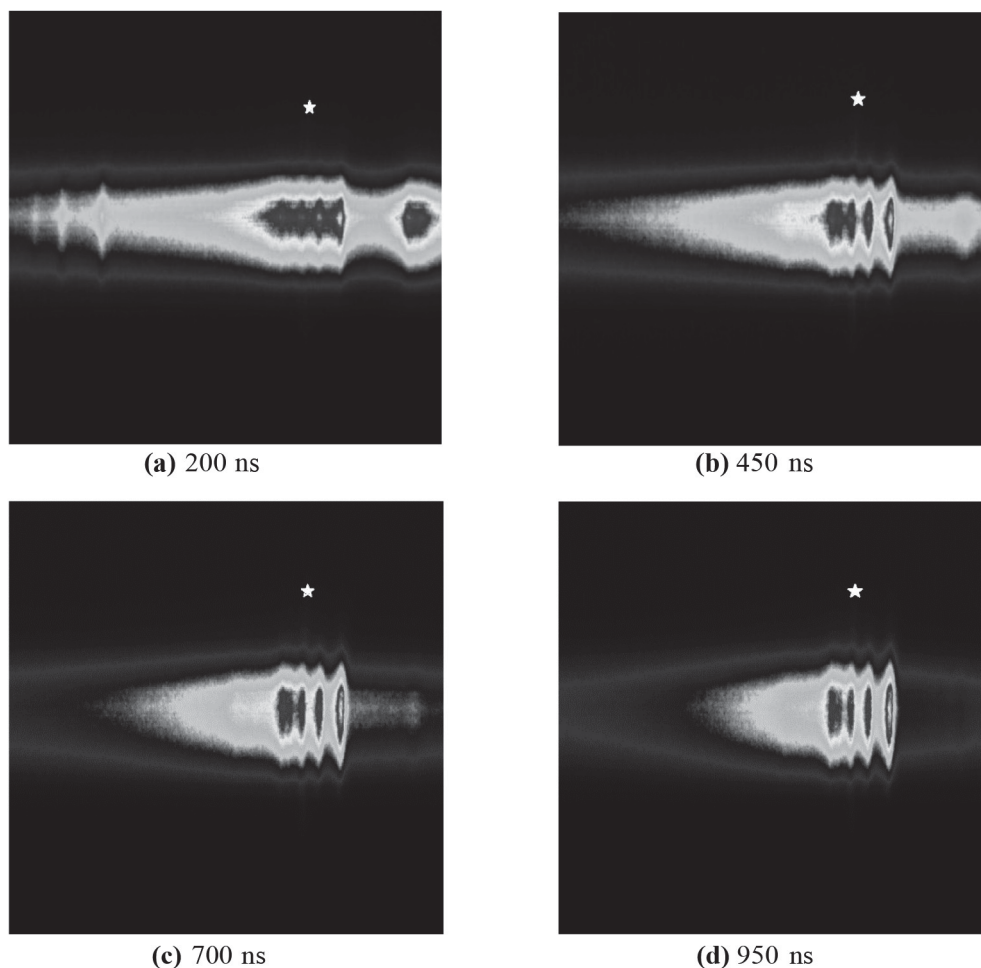


Figure 5: Optical breakdown CN spectra in a 1:1 molar $\text{CO}_2\text{:N}_2$ flowing gas mixture for time delays of (a) 200 ns, (b) 450 ns, (c) 700 ns, and (d) 950 ns. Spectrometer-detector gatewidth: 125 ns. *, 2nd order CI.

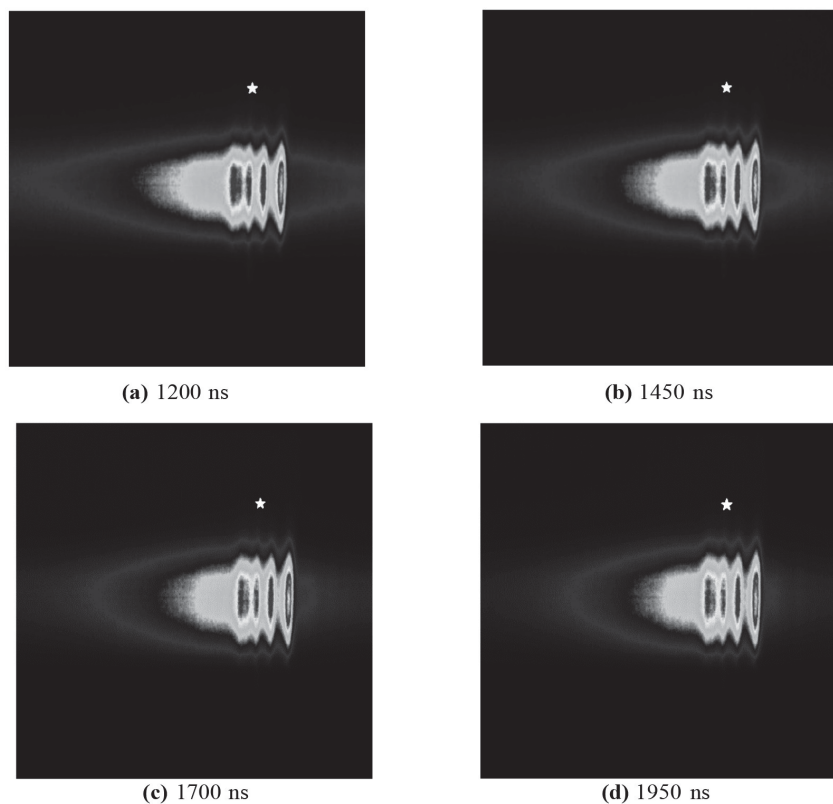


Figure 6: Optical breakdown CN spectra in a 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gas mixture for time delays of (a) 1200 ns, (b) 1450 ns, (c) 1700 ns, and (d) 1950 ns. Spectrometer-detector gatewidth: 125 ns. *, second-order atomic carbon line.

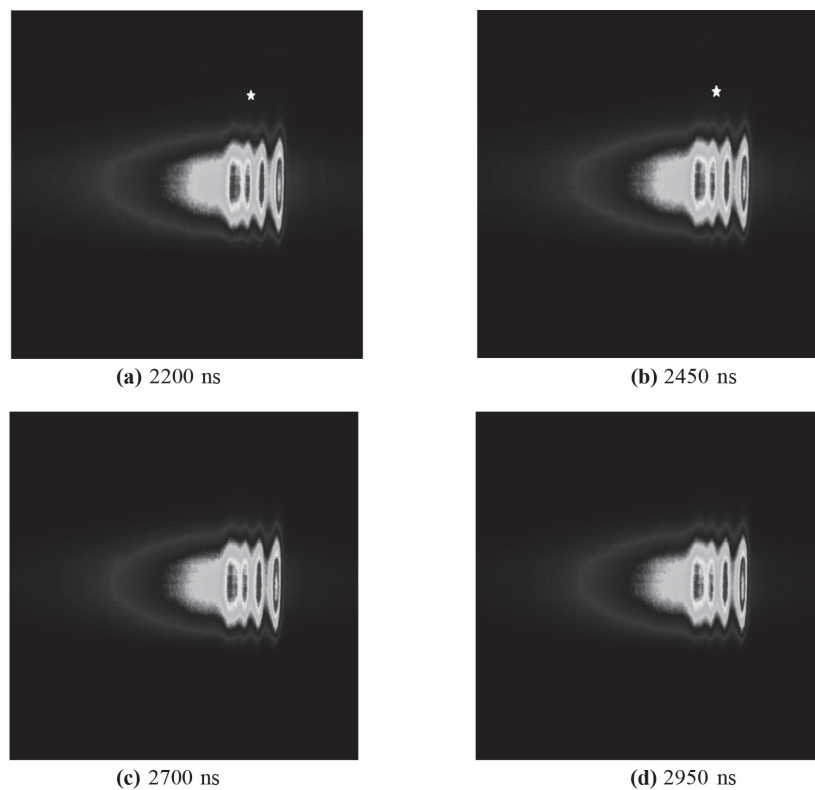


Figure 7: Optical breakdown CN spectra in a 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gas mixture for time delays of (a) 2200 ns, (b) 2450 ns, (c) 2700 ns, and (d) 2950 ns. Spectrometer-detector gatewidth: 125 ns. *, second-order atomic carbon line.

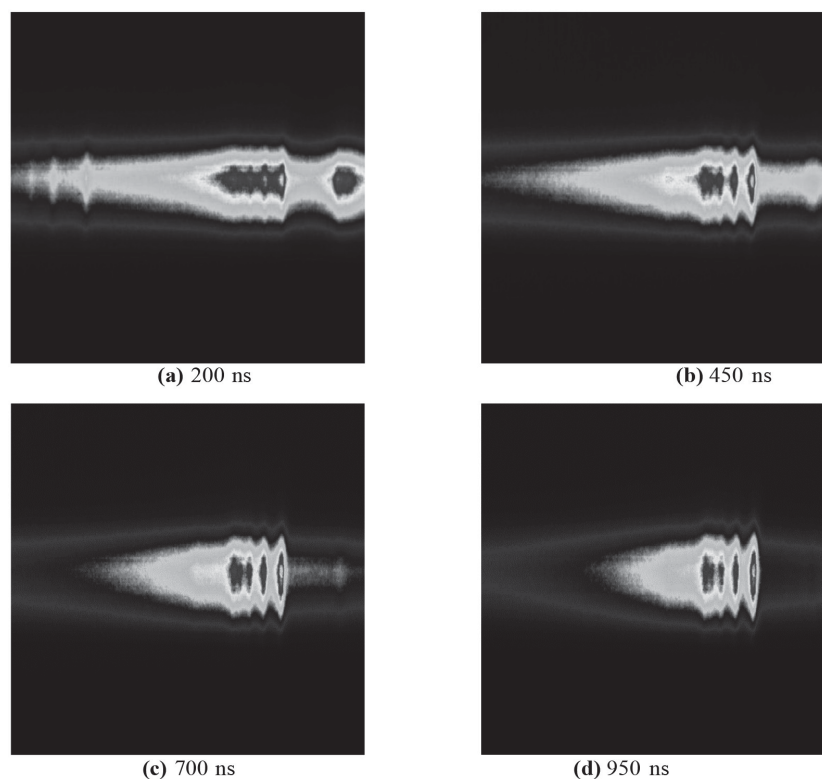


Figure 8: Optical breakdown CN spectra in a 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gas mixture for time delays of (a) 200 ns, (b) 450 ns, (c) 700 ns, and (d) 950 ns, recorded with 309 nm cut-on wavelength filter for suppression of 193.09 nm second order atomic carbon line

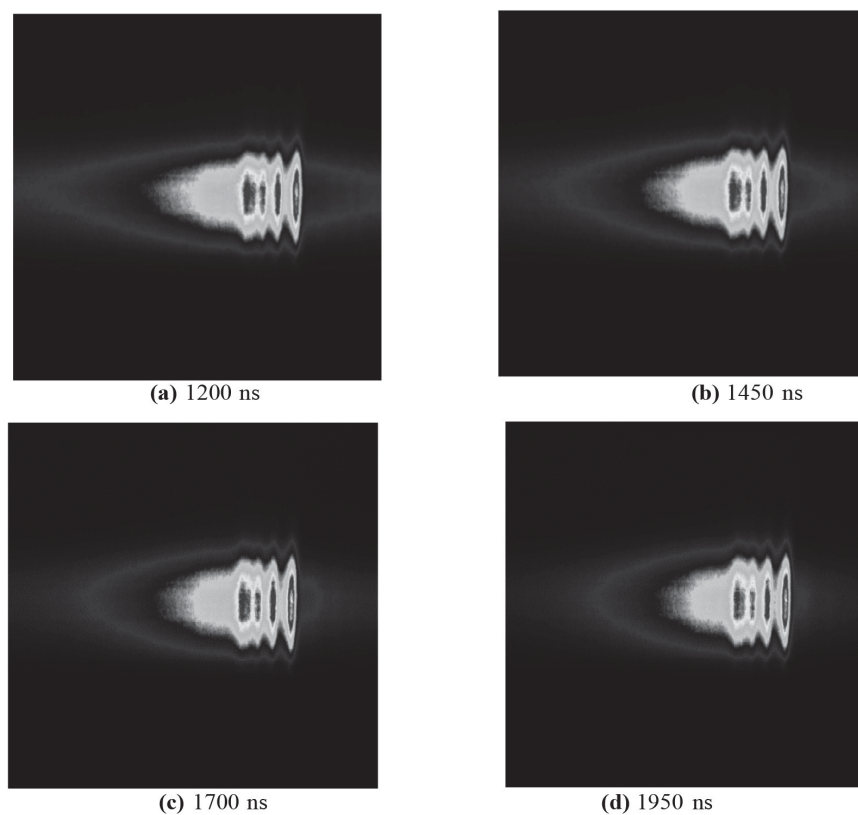


Figure 9: Optical breakdown CN spectra in a 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gas mixture for time delays (a) 1200 ns, (b) 1450 ns, (c) 1700 ns, and (d) 1950 ns, recorded with 309 nm cut-on wavelength filter for suppression of 193.09 nm second order atomic carbon line.

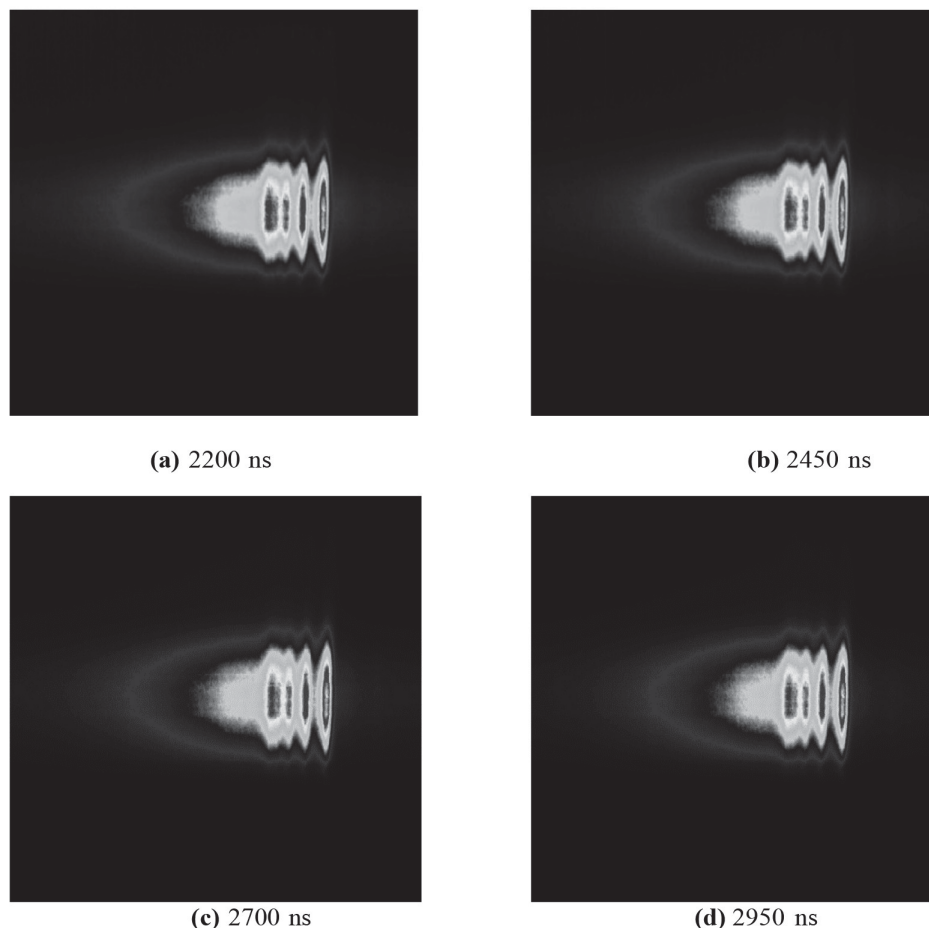


Figure 10: Optical breakdown CN spectra in a 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gas mixture for time delays (a) 2200 ns, (b) 2450 ns, (c) 2700 ns, and (d) 2950 ns, recorded with 309 nm cut-on wavelength filter for suppression of 193.09 nm second order atomic carbon line.

3.2 Cyanide Temperature

Inferred temperatures, obtained from spectrum fitting [6], of filtered line-of-sight CN spectra in the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture are plotted versus slit height of the spectrometer as shown in Figures 11 to 14. As can be seen in Figures 11 to 14, temperature variations occur in the central region, while increased temperatures are shown at the edges of the plasma similar to the 1:1 molar $\text{CO}_2:\text{N}_2$ gaseous mixture held at atmospheric pressure and 276 kPa (2070 Torr) [2]. Identical to the 1:1 molar $\text{CO}_2:\text{N}_2$ gaseous mixture held at atmospheric pressure and 276 kPa (2070 Torr), higher temperatures are seen on the edge of the plasma towards the top of slit or towards the laser side. At a time-delay of 450 ns, Figure 11, the temperatures in the central region of the plasma are between 9,500 K to 10,000 K, while the temperatures at the edges of the plasma are more than 10,000 K. At a time-delay of 700 ns time delay, Figure 11, the temperatures in the central region of the plasma cool to a range of 9,000 K to 9,500 K, while temperatures at the edges of the plasma are between 9,500 K to 10,000 K. At the 950 ns time delay, Figure 12, the temperatures in the central region of the plasma remain in a range of 9,000 K to 9,500 K, while the temperatures at the edge of the plasma towards the top of the slit increase to greater than 11,000 K. At time delays of 1.2 μs to 1.7 μs the plasma central region temperatures cool even further to a range of 8,500 K to 9,000 K, while the edge of the plasma towards the top of the slit maintain temperatures in excess of 11,000 K as seen in Figures 12 and 13. From time delays of 1.95 μs to 2.2 μs , Figure 14, the central region of the plasma sustains temperatures of 8,500 K to 9,000 K and temperatures near the edge of plasma towards the bottom of the slit are around 9,000 K, while temperatures near the edge of the plasma towards the top of slit increase to greater than 11,000 K. Also seen in Figure 14, is the plasma temperatures in excess of 11,000 K are near the edge of the plasma towards the top of the slit.

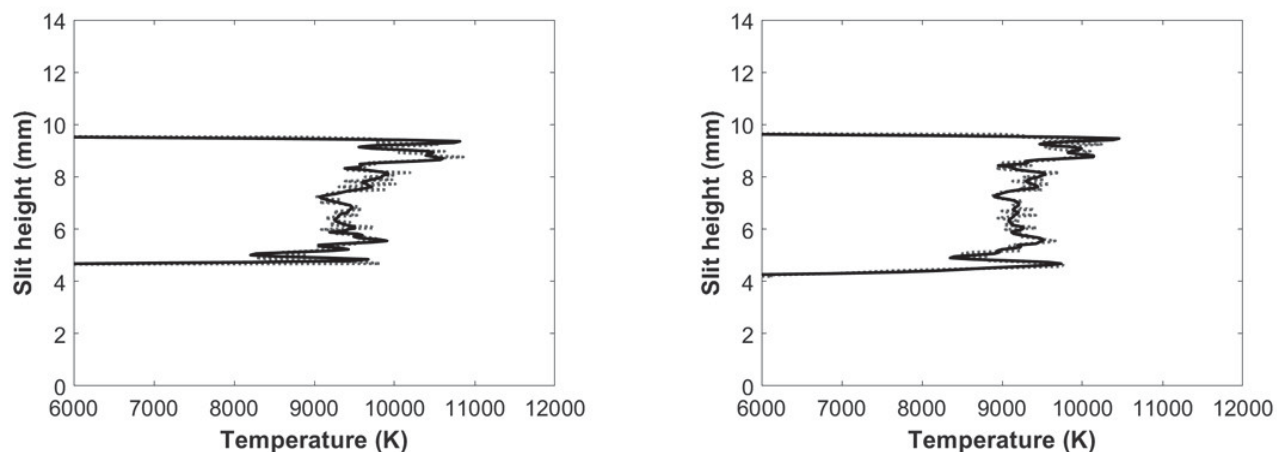


Figure 11: Temperature vs. slit height for filtered line-of-sight CN spectra for a flowing 1:1 molar $\text{CO}_2:\text{N}_2$ gaseous mixture with 450 ns (left) and 700 ns (right) time delay [7].

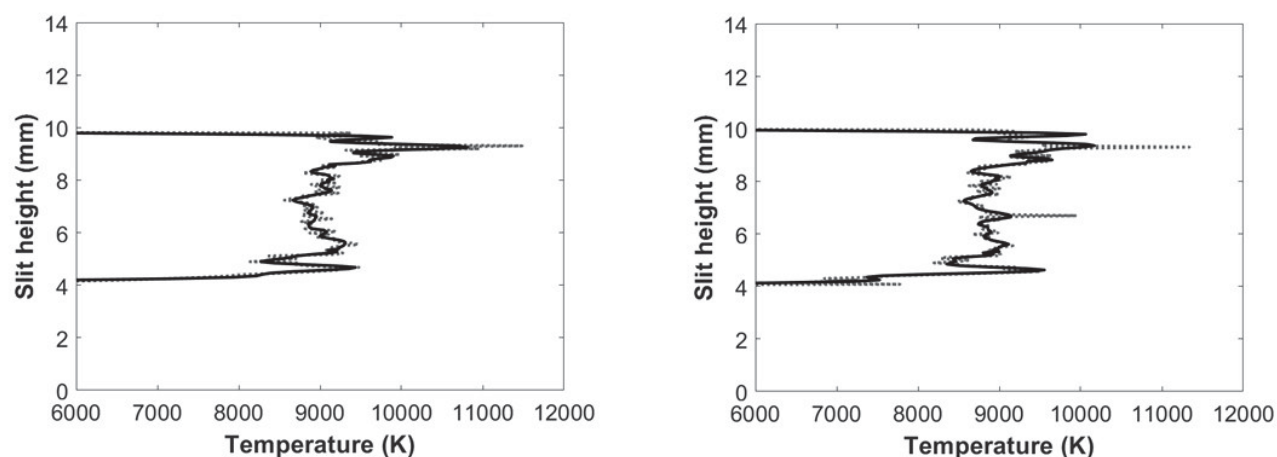


Figure 12: Temperature vs. slit height for filtered line-of-sight CN spectra for a flowing 1:1 molar $\text{CO}_2:\text{N}_2$ gaseous mixture with 950 ns (left) and 1200 ns (right) time delay [7].

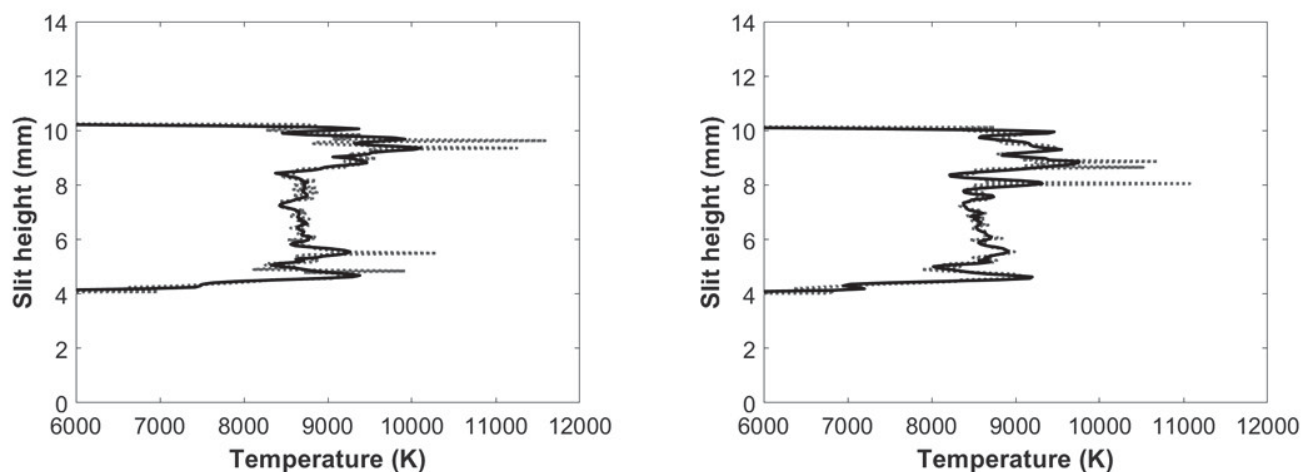


Figure 13: Temperature vs. slit height for filtered line-of-sight CN spectra for a flowing 1:1 molar $\text{CO}_2:\text{N}_2$ gaseous mixture with 1450 ns (left) and 1700 ns (right) time delay.

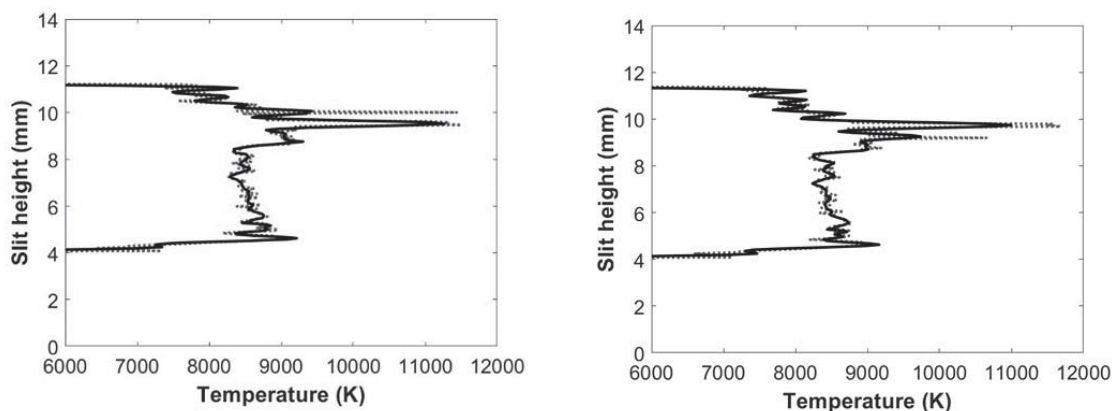


Figure 14: Temperature vs. slit height for filtered line-of-sight CN spectra for a flowing 1:1 molar $\text{CO}_2:\text{N}_2$ gaseous mixture with 1950 ns (left) and 2200 ns (right) time delay.

3.3 Stark widths and Stark shifts

Inferred Stark widths of the CI 193.09 nm carbon line in second order for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture were determined using the same technique as the gaseous mixture held at atmospheric pressure. The inferred Stark widths are plotted versus the slit height of the spectrometer, which can be seen in Figures 15 to 22. Larger Stark widths are seen towards the edges of the plasma, while smaller Stark widths are seen in the center of the plasma. For time delays of 450 ns to 2200 ns, Figures 15 to 22, the Stark widths are between 0.4 to 0.5 nm and located towards the edges of the plasma.

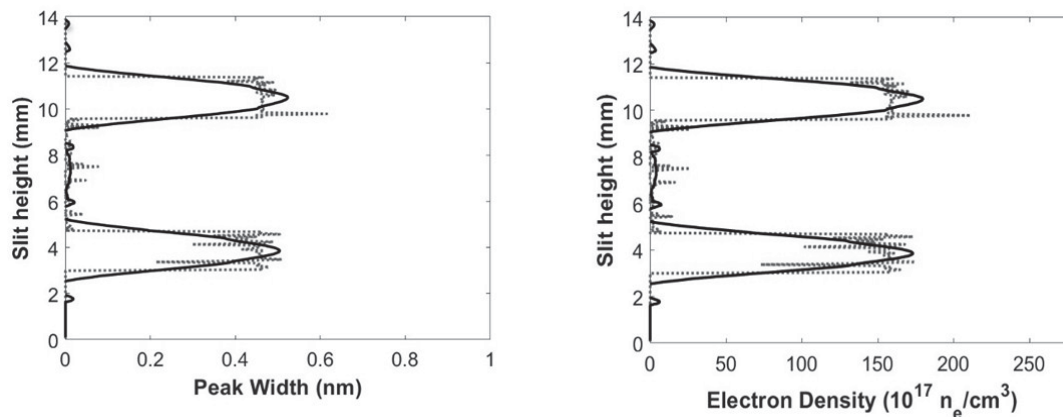


Figure 15: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 450 ns time delay.

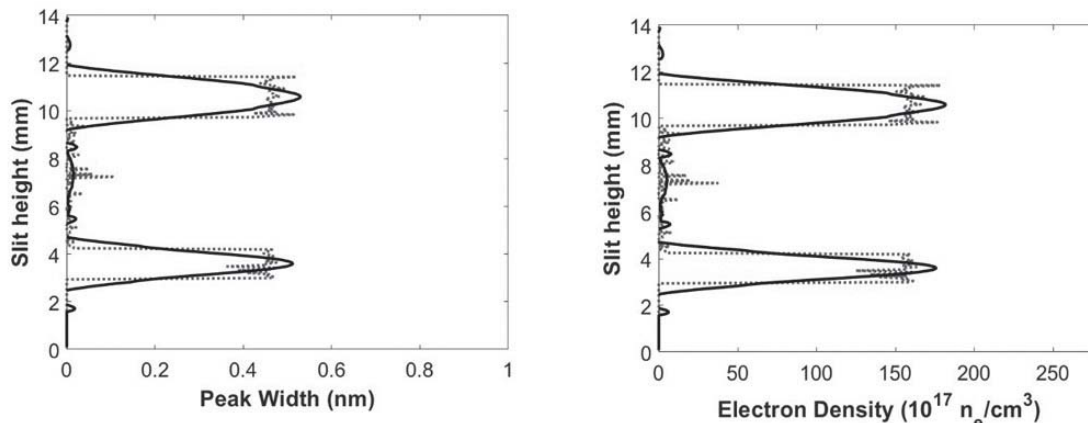


Figure 16: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 700 ns time delay [7].

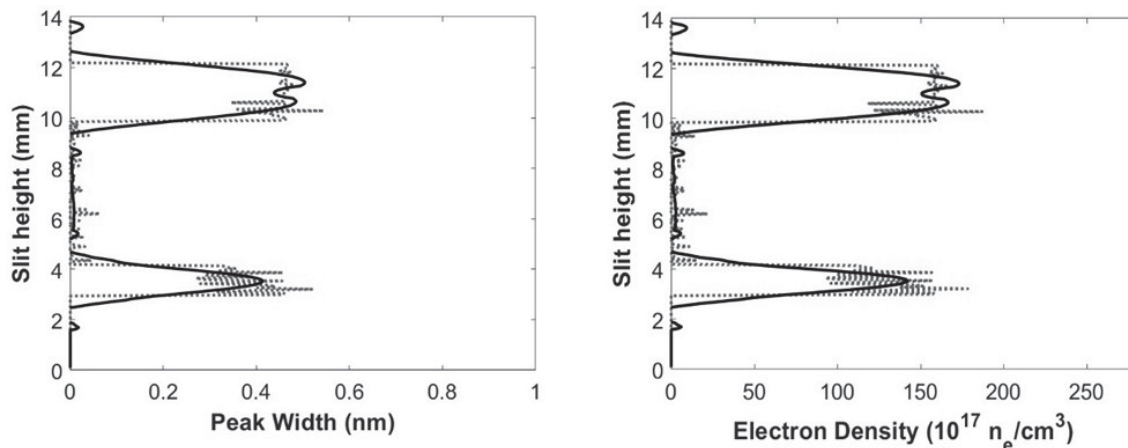


Figure 17: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 950 ns time delay [7].

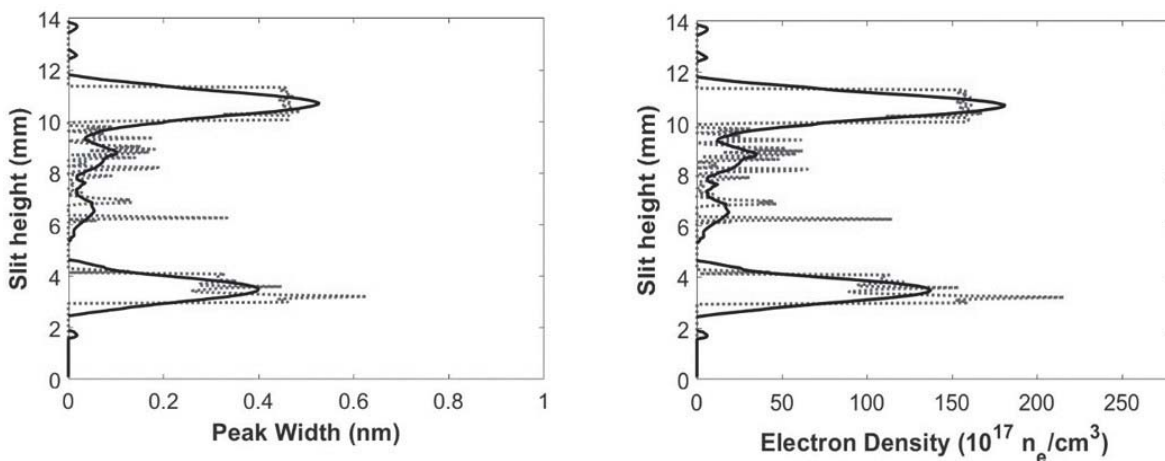


Figure 18: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 1200 ns time delay.

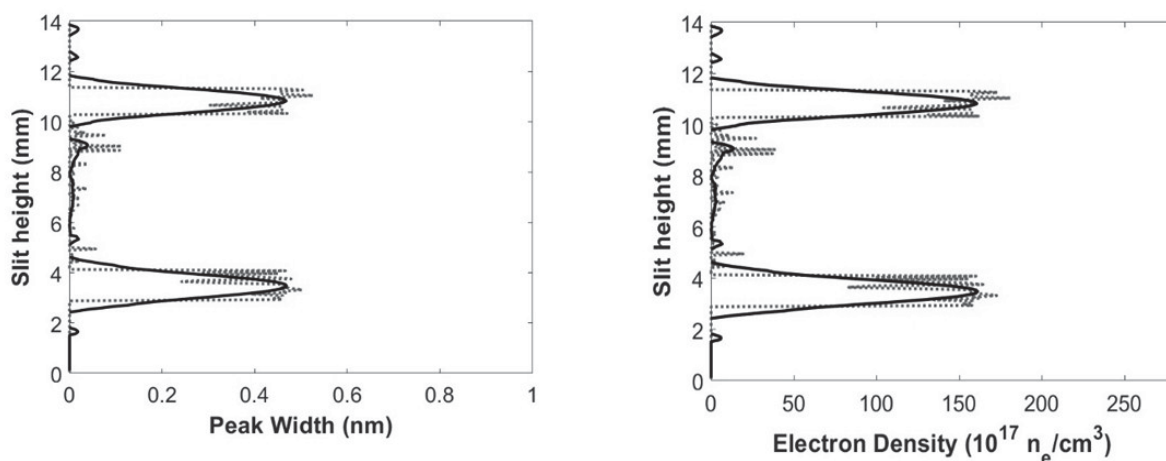


Figure 19: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 1450 ns time delay.

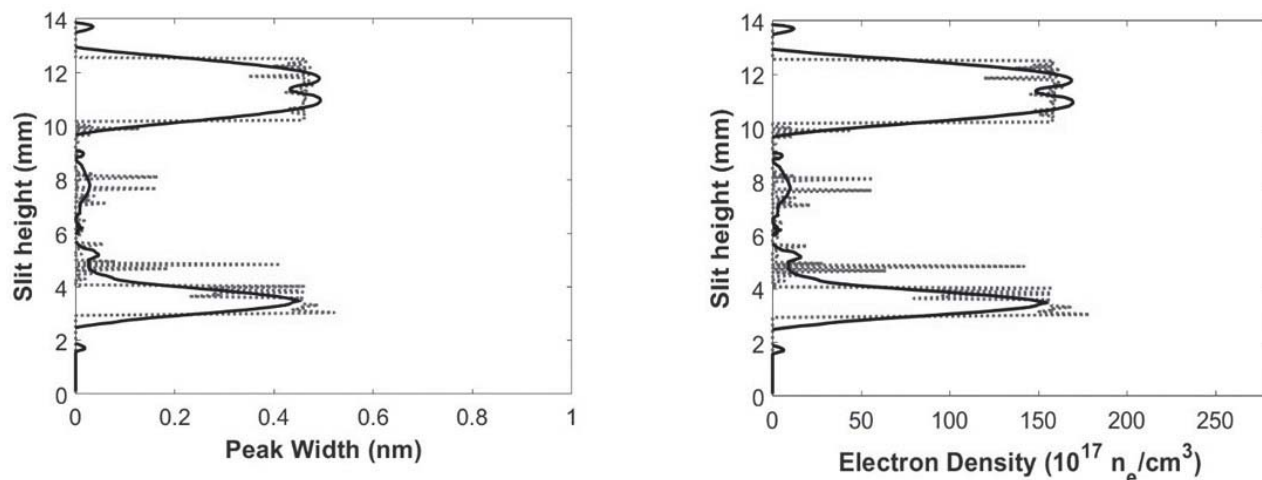


Figure 20: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 1700 ns time delay.

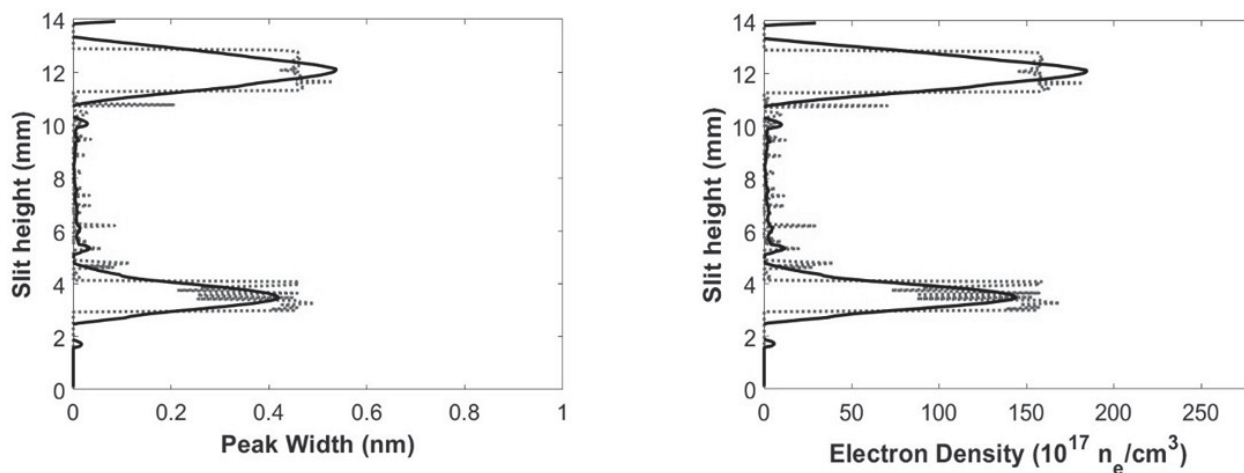


Figure 21: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 1950 ns time delay.

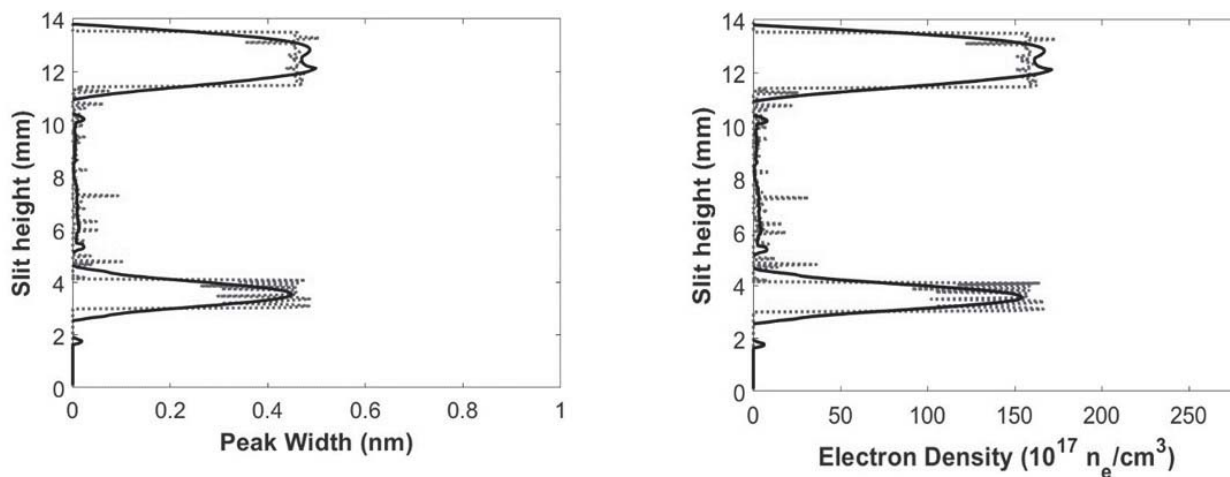


Figure 22: Inferred widths (top) and calculated electron densities (bottom) of CI 193.09 nm atomic carbon line in second order vs. slit height for the 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 2200 ns time delay.

Infer d St ea kvchie Sfehifweds eo GhCdfefCods lelh9 3fSewf likd 0elFe 1 Gns hie dchfem ifshi 9 kchSfenfve d
 0 sibnfSoebSfiihSfel: Qel nfeCods lelh9 3fSewf likd 0ne 0eo le3fefdS9 klfweS9 edhfer d St ea kvch0eg u_{r,d,s} 0e 1w
 rd St einKl0S_{Stark} 0s Nchfelfhd 0o Ss lex qpTv2, Tl9 e d9 loo Ss leClfdT0 f ihSfwde l^wes S_wfS0

$$\Delta\lambda_{\text{Stark}} \text{ (nm)} = 2 w \text{ (nm)} n_e \left(10^{17} \text{ cm}^{-3} \right), \quad \delta\lambda_{\text{Stark}} = d n_e \left(10^{17} \text{ cm}^{-3} \right),$$

a nSfehfa kvch0a 0e 1we nk0b S 9 fdS00e Sfecds bsCdfwT0p, 0d3fa $\epsilon \approx e 2, : Tpe 9 0w \approx , 2, : Tye 9 2l$ nf
 o GhCdfwefS9 er d St ea kvchi 0GdfwefS hi chfa 0cnfkmcs Nchf b fods 9 fdS0a nkon0 le3fa fflkl dFkhSfi 050s
 $: : 2x$ GhCdfwbf t fCods lewlikdfe Sfes Nchfes S_wfSs N_f $\approx \varphi$, $p^{T0} 9 R 2E$ Cods lewlikd 0Gd dS9 0 inkNl 0 l9 koehf
 i 9 fe3fn 4ks Se ichfebS4lshi C0 f lds lfwrd St Fa kvch0Gd 0N S chf0S a klmm ifshi 9 kchSf0a nSfenkmfSefeki
 iffl0da Swiechf0wfnies NchfebCi9 e lweGa fSefekieda Swiechf0f ldfSes NchfebCi9 ei l9 kC Sedchfem ifshi
 9 kchSfenfve d 0 sibnfSoebSfiihSf2l nfanmfsfCods lewlikdfe dsa Swchf0wfnies NchfebCi9 e bbf S0s 0S0a
 dhfeinsota 4f0cb lils leCa dpp0

$$R(\tau) = \left(\frac{E}{\rho_{mi}} \tau^2 \right)^{\frac{1}{5}},$$

a nSfeEeki chfe 3isSfwehGfefl fSm 0p_{mi} eki chfem iewf likd 0e lwereki chfed0 fewfC. 0S9 esbdo 03Sf twsa 12l nf
 Sfih0en S chfeos9 bhdfwe lwe9 f ihSfweS9 ein wsa nS bnileinsota 4fes wkeo le3feifflekl 30ep2l nfi f
 insota 4fes wkeo Nchfes a klmm ifshi 9 kchSfe S0 hono 0ifS0 chf0Sfwodwansota 4fes wkeo lchf0ansota 4f
 S wkeo Nchfem ifshi 9 kchSfenfve d 0 sibnfSoebSfiihSfdp: 0

Table 1: Computed shockwave radii vs. plasma radius for the 1:1 molar CO₂:N₂ flowing gaseous mixture, 170 mJ

τ 5ilL	os 9 bhdfweRd 9 9 O	9 f ihSfweRd 9 9 O
y5,	p2y	: 2y, e+e 27:
7,,	: 2,	: 27, e+e 28p
T5,	: 2y8	: 285e+e 286
p:,,	: 27:	: 27, e+e 287
py5,	: 2Ty	v2 5e+e 27:
p7,,	v2pv	v2p5e+e 275
pT5,	v2vp	v255e+e 27 7
::,,	v2y7	v27, e+e 28p

qlNSSFwer d St einKl0s Nchfex qpTv2, Tel9 0o Ss leClfdk f0s lws S_wfSs Nchfep:pe 9 sC S0x O: N 0S0a klmm ifshi
 9 kchSfa fSewf dS9 klfwhi kl nchfa 9 fedonlkqhf0 bb0fw0s chfem ifshi 9 kchSfenfve d 0 sibnfSoebSfiihSfe lwe
 $: 76d P$ 0s, 7, d sSld: 0p: 0pv0el S_wfS0rd St einKl0e Sfeiffleda Swiechf0wfnies NchfebCi9 0a nk0a 9 0S0rd St
 a kvchie Sfeiffleklchf0f ldfSes NchfebCi9 2A0chs hmer d St einKl0eo le3fehifweds ewf dS9 klfel, 0chfer d St ea kvch
 9 fchswe bbf S0s 03fe ei hbfSs S0fonlkqhf2

3.4 Abel inverted CN Spectra

A3f0kl4fSiks l0s Nchf0K0Sfwep:pe 9 sC S0x O: N 0S0a klmm ifshi 9 kchSfa i 0fS_wS9 fwe3. dkl4fSklm0 f ihSfw0l fF
 sNf kmcdw d 00fz0u l0N S0f onea 4f0l n0u000es 3d klchfes vk 0kl d flikd ewi dS3hds l0eS0u l0e oos S_wkl n0s chfes lff
 ikwvFshSfSs S0nf3. inf40sC l0s k 0 0s S0h9 dpy0

$$I(z, \lambda) = 2 \int_z^p \epsilon(r, \lambda) \frac{r}{\sqrt{r^2 - z^2}} dr,$$

PS4lshi C0 f ihSfwein wsa nS bnileins a chfebCi9 enflfS dfweklchfep:pe 9 sC S0x O: N em ifshi 9 kchSfenfve d
 0 sibnfSoebSfiihSfen ie 0Gifed0bnfS0 0n b0a n0nea sh0Ghi dN chfchif0s N03f0kl4fSiks l2A0e 0d0 ffwfC.
 sN0,, di0FkhSfe v0chfex N0wi dS3hds le bbf S0f4f1C ewi dS3hdfwe 0Sii chf0Ci9 2FS9 0d0 fewfC. i0N0y, 0i00

1950 ns, Figures 23 to 25, the CN signals begin to become stronger towards the edges of the plasma and weaker in the center of the plasma, which is consistent with the higher temperatures seen at the edges of the plasma as discussed previously. At 2200 ns time delay, Figure 23, it is clearly visible that there is a higher CN signal towards the edges of the plasma than in the center. This trend is maintained up to 5200 ns as can be seen in Figures 24 to 29. These results were projected to be analogous to the shockwave results, but inside the plasma kernel and shockwave the variations of the CN distribution were expected.

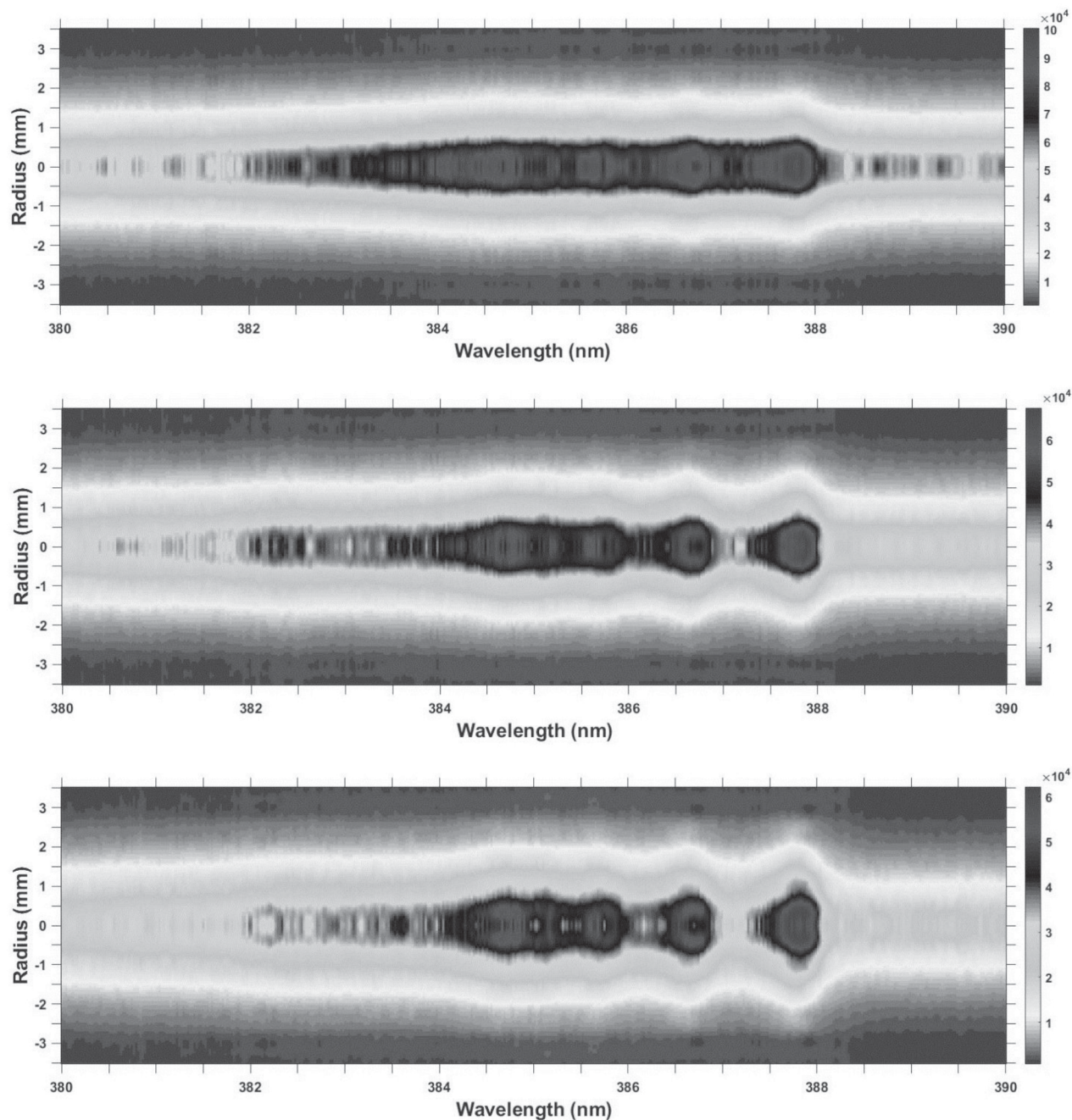


Figure 23: Abel inverted CN spectra 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 200 ns (top), 450 ns (center), and 700 ns (bottom) time delay.

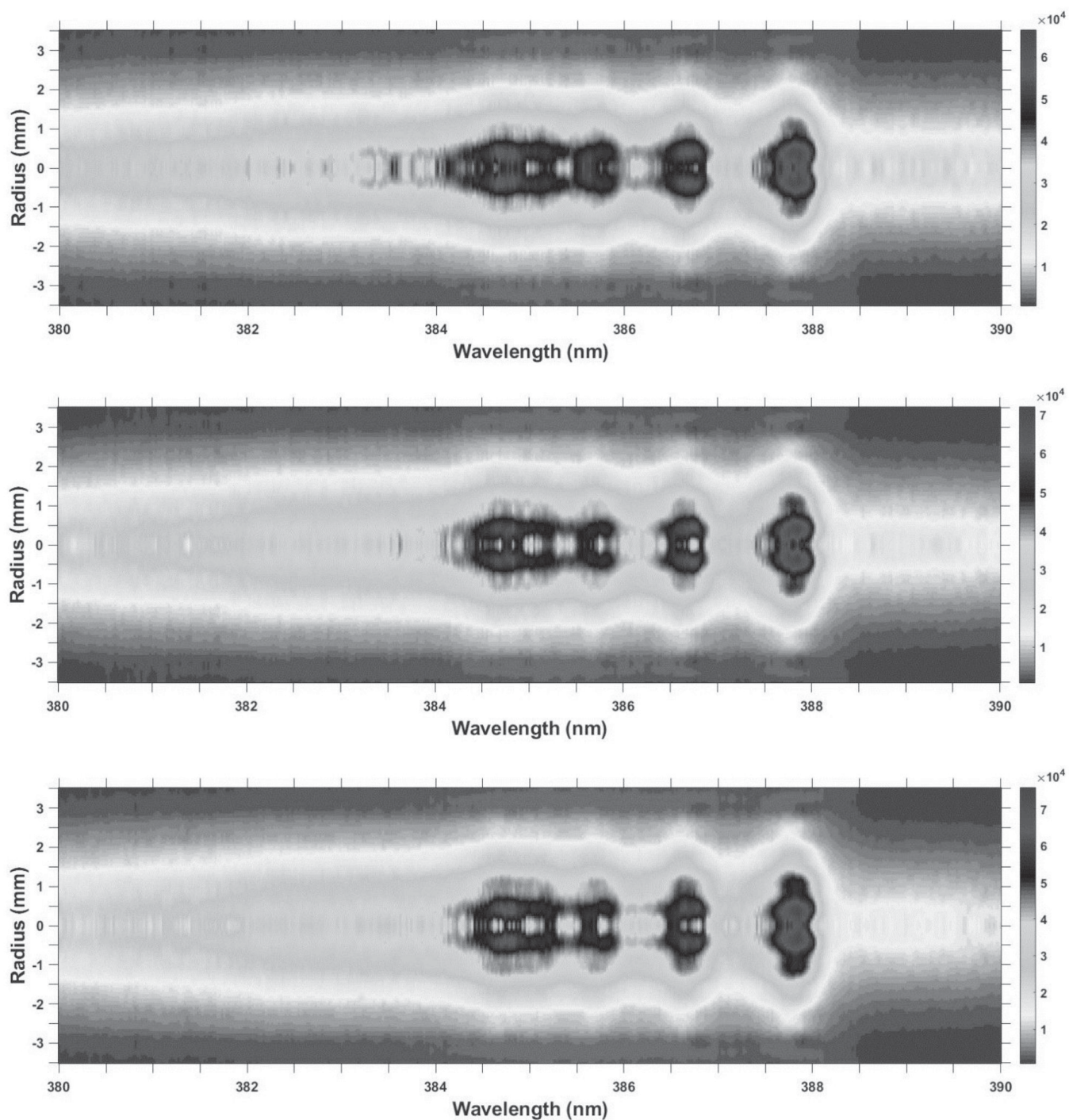


Figure 24: Abel inverted CN spectra 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 950 ns (top), 1200 ns (center), and 1450 ns (bottom) time delay.

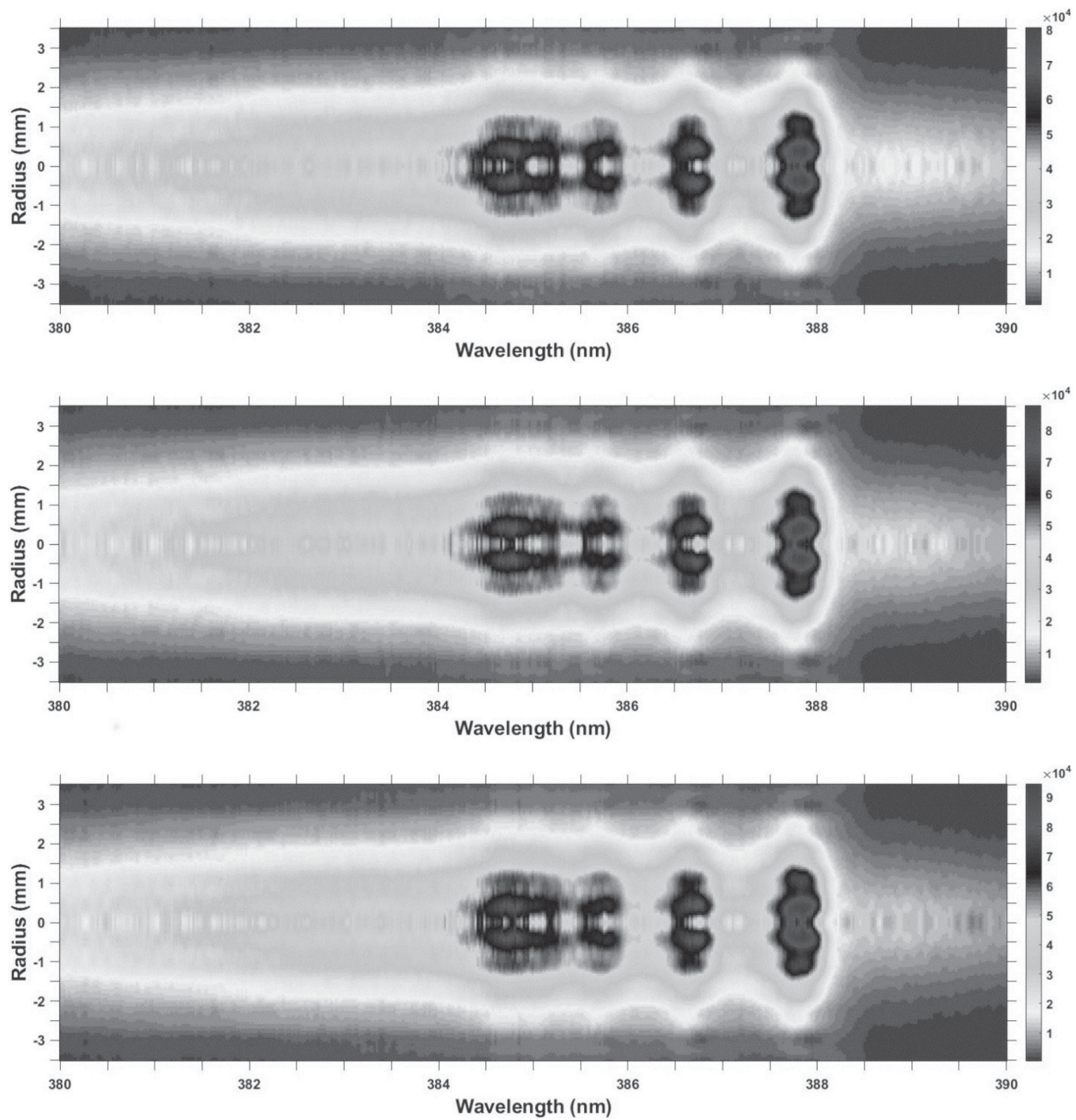


Figure 25: Abel inverted CN spectra 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 1700 ns (top), 1950 ns (center), and 2200 ns (bottom) time delay.

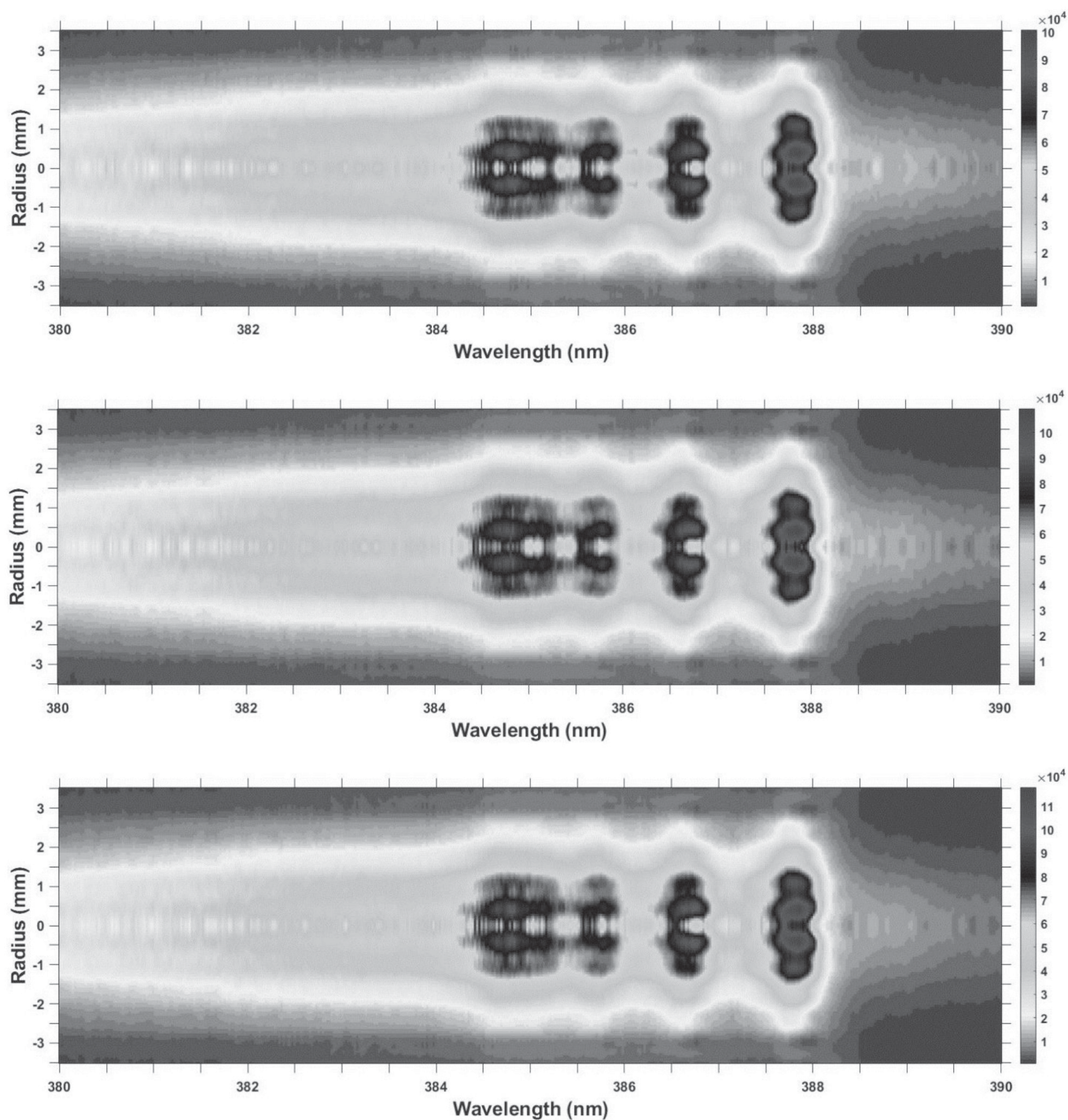


Figure 26: Abel inverted CN spectra 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 2450 ns (top), 2700 ns (center), and 2950 ns (bottom) time delay.

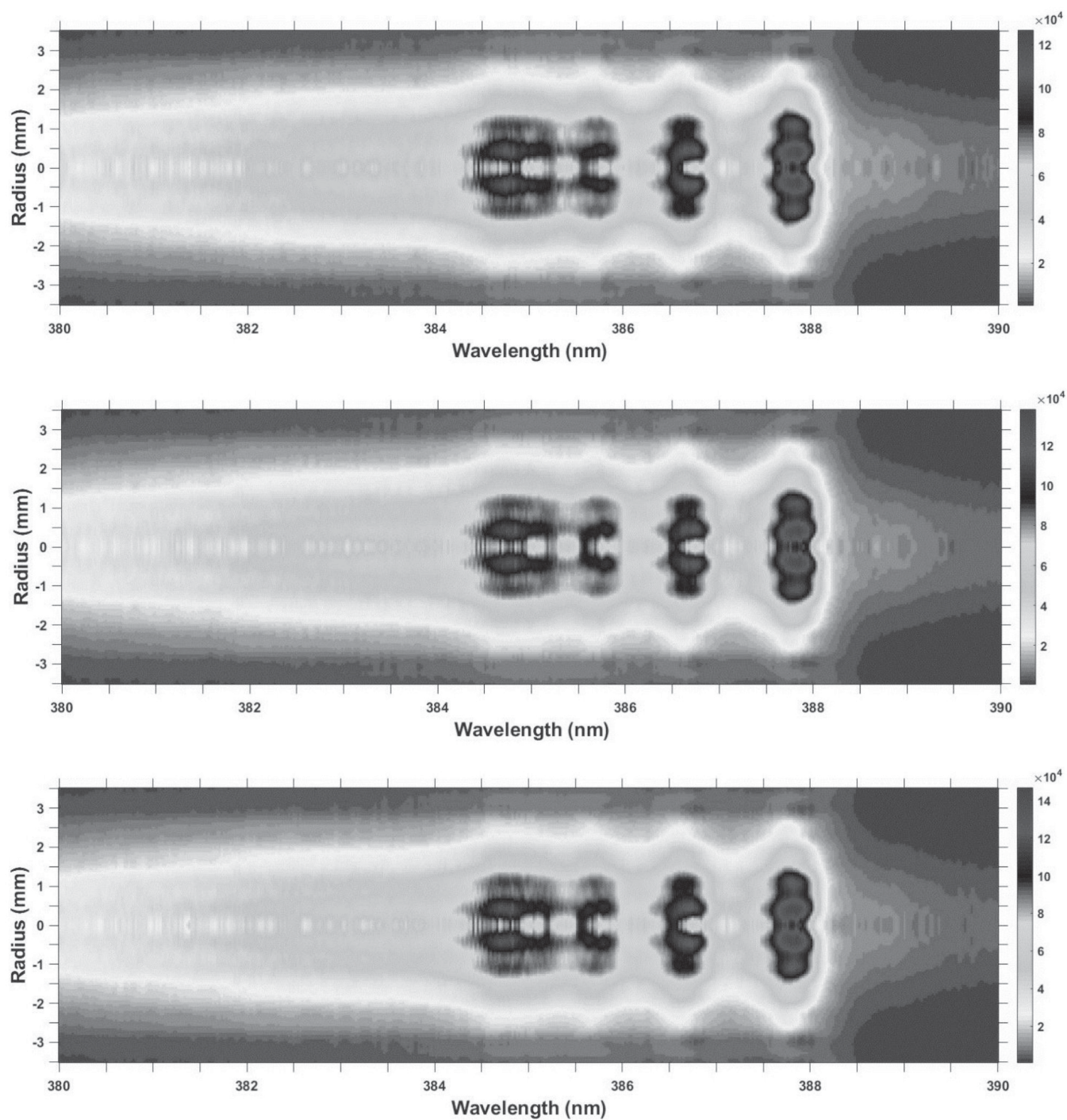


Figure 27: Abel inverted CN spectra 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 3200 ns (top), 3450 ns (center), and 3700 ns (bottom) time delay.

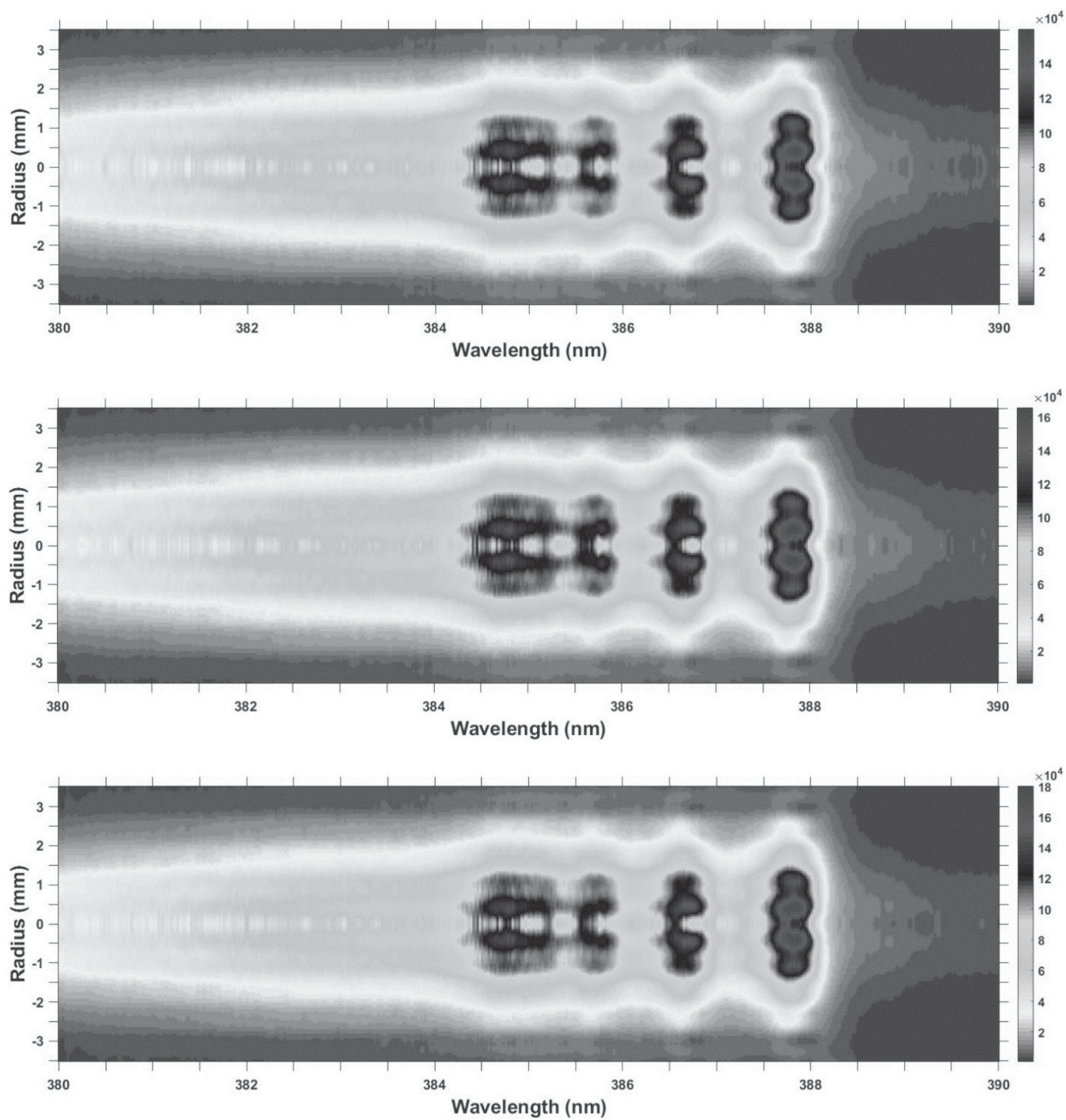


Figure 28: Abel inverted CN spectra 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 3950 ns (top), 4200 ns (center), and 4450 ns (bottom) time delay.

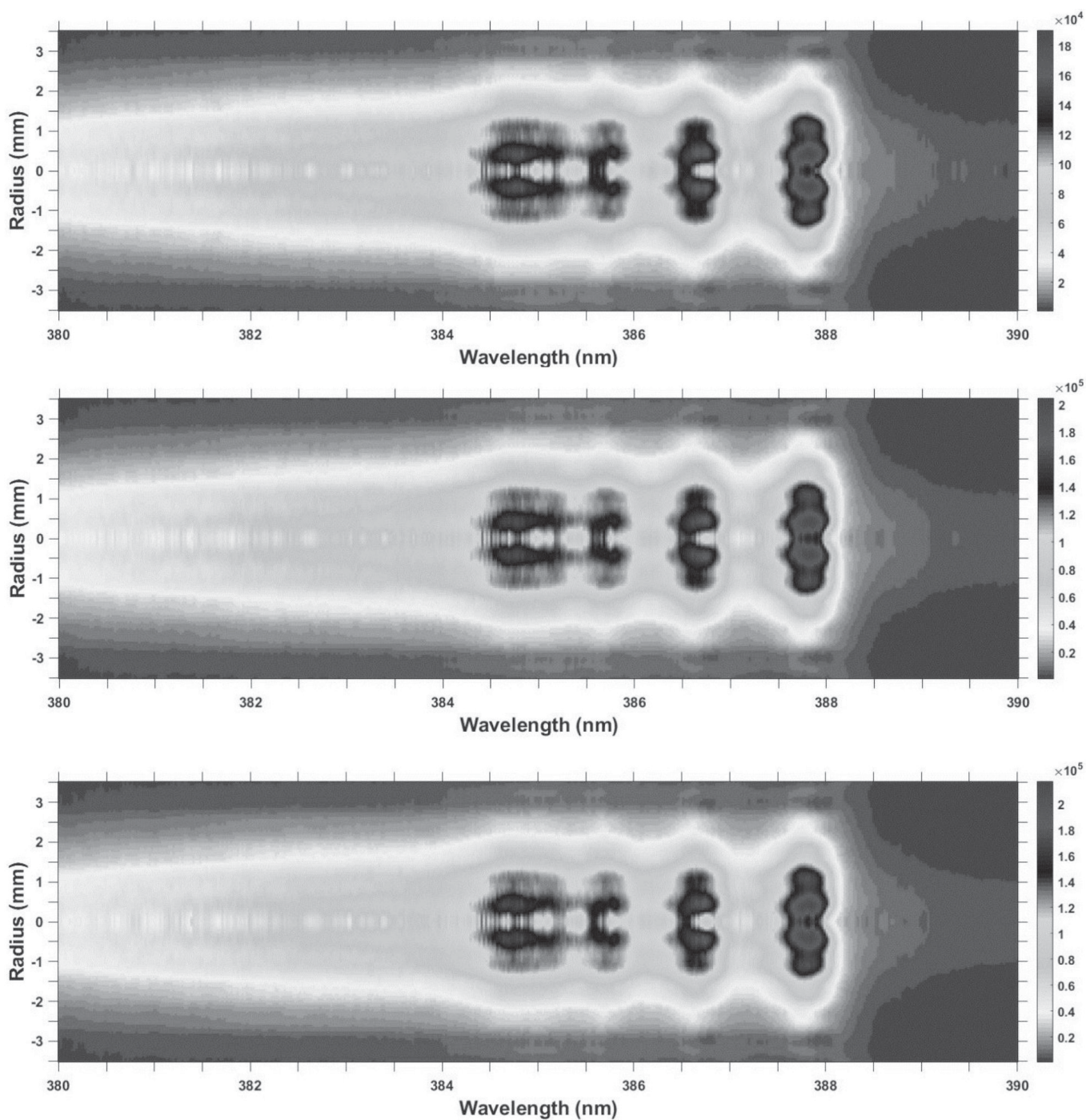


Figure 29: Abel inverted CN spectra 1:1 molar $\text{CO}_2:\text{N}_2$ flowing gaseous mixture with 4700 ns (top), 4950 ns (center), and 5200 ns (bottom) time delay.

3. CONCLUSIONS

Experiments described in this work show the effects of the shockwave on the plasma dynamics with absorbed energies in the range of 160 to 200 mJ. Particularly for the flowing gaseous mixture investigations, the shock wave radii computed from the strong-explosion formula, agree with the spectroscopic results. Line-of-sight spectra already

indicate spatial variation across the plasma, however, for the near symmetric plasma expansion, Abel inversion allows one to make further inferences about the expansion dynamics, including the spatio-temporal dynamics of the plasma kernel, or the epicenter of the laser plasma. These detailed studies are important prior to applications of diagnosis of CN with laser-induced plasma in medical and forensic applications.

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