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Ar⁺ LASER EXCITED FLUORESCENCE OF
DIATOMIC COMBUSTION RADICALS IN A FLAME

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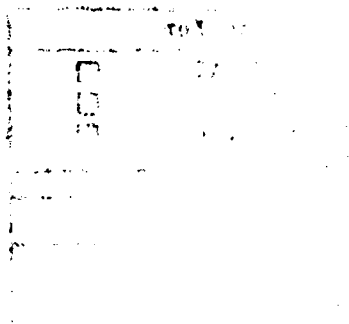
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I. INTRODUCTION

At various times over the past ten years we have encountered laser excited fluorescences during the course of performing spontaneous Raman spectroscopy on atmospheric pressure premixed flames. These fluorescences come from accidental coincidences of ro-vibrational electronic transitions with prism-selected lasing lines of Ar^+ or Kr^+ ion lasers. At the elevated temperatures of a flame the combustion radicals have many more populated states than at room temperature, increasing the chance for a coincidence with a laser line. The combustion diatomic radicals that have been excited in this manner in a $\text{CH}_4/\text{N}_2\text{O}$ premixed flame are $\text{CN}^{1,2}$, OH^3 , NH^3 and $\text{C}_2^{2,4-6}$. Recently, ion laser technology has been improved to allow higher power ultraviolet lasing to be commercially available. We report here a further investigation of coincidental fluorescence excitation of combustion diatomics from ultraviolet lines of an Ar^+ ion laser.

II. EXPERIMENT

Two different experimental arrangements were used in this study due to the limitation of available focusing optics. Fig. 1 illustrates the previously used intracavity arrangement. The excitation source is a Coherent Innova 200 - 25/7 argon ion laser which

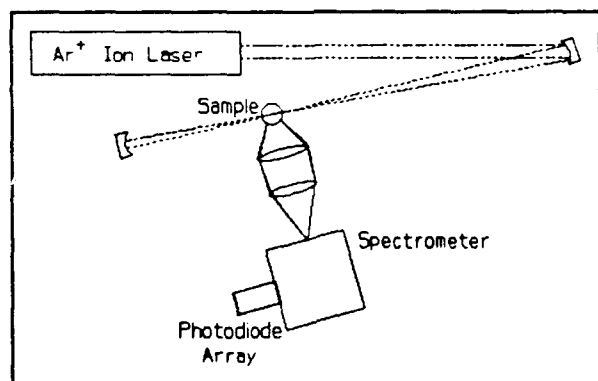


Figure 1. Experimental setup showing the sample placed at the intracavity focus of the laser.

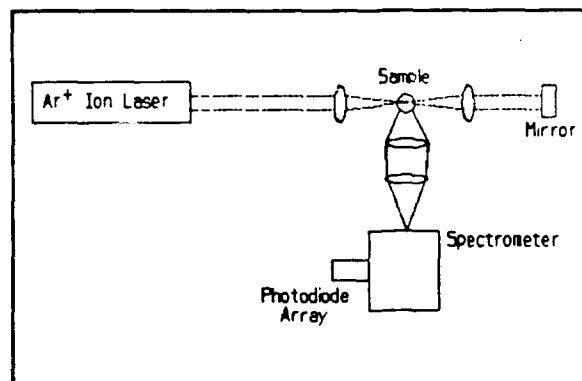


Figure 2. Experimental setup showing the sample placed at an extracavity focus of the laser.

is operated in an intracavity mode. That is, the output coupler is removed from its normal position in the laser frame and the lasing cavity is extended (focusing provided) with two concave mirrors having radii of curvature of 100 and 30 cm. Depending on the amount of absorption of the sample and the lasing line the intracavity lasing power can be more than two orders of magnitude larger than the extracavity power. These intracavity mirrors, however, are not highly reflective below 350 nm; thus for those laser lines the experimental arrangement for the excitation source is shown in Fig. 2. The laser output coupler is now in the normal position and the output laser radiation is focused and recollimated with 20 cm focal length convex lenses. A flat mirror returns the radiation through the same path effectively doubling the amount of radiation in the sampling region.

Raman and fluorescence signals are gathered at right angles to the direction of the excitation source for both arrangements shown in Figs. 1 and 2. An f1 10 cm focal length quartz convex lens is the collection optic and an f2 20 cm focal length quartz convex lens is used to interface the collection optic to the 0.25 m Jarrell-Ash spectrometer. These optics magnify the sample region by a factor of two which fills the 0.1 mm entrance slit. Spectra are detected and recorded with an EG&G Princeton Applied Research Model 1456 intensified photodiode array which is coupled to the spectrometer. This detection system captures spectra of differing wavelength range depending on the order and line spacing of the grating used in the spectrometer. As an example a 1200 groove/mm grating operating in first order will simultaneously disperse a wavelength increment of about 40 nm over the approximately 700 intensified pixels of the photodiode array. A premixed laminar $\text{CH}_4/\text{N}_2\text{O}$ flame is used for the source of hot combustion radical species. The N_2O oxidizer facilitates the production of radical species which contain single C and/or N atoms (e.g. CN). This flame is supported on a small multi-hole metal burner head of 0.4 cm diameter. Although the flow conditions were not well regulated the flame composition was lean with a flame temperature around 2400 K (as determined by absorption spectroscopy).

III. RESULTS

There are twelve commercially advertised uv lasing lines of the Ar^+ laser used in this study; all except two (385.8 and 308nm) were made to lase. It is unclear why these two lines did not lase; perhaps there were problems with mirror coatings. However, an unadvertised Ar^+ line at 379.5 nm was found to lase. Bridges and Chester⁷ have previously

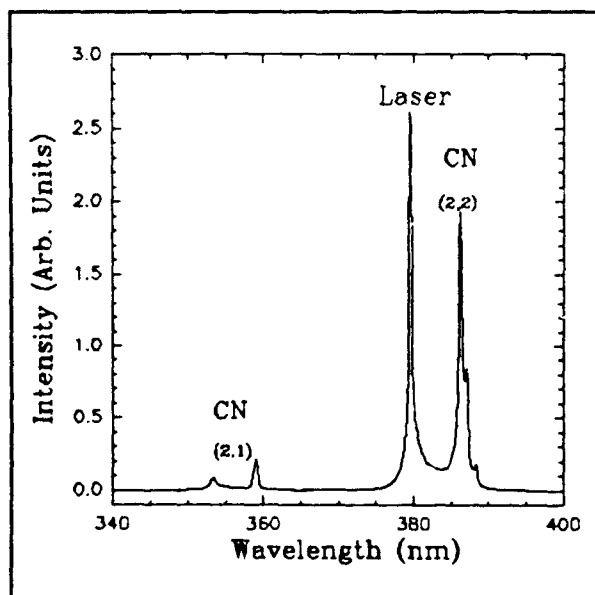


Figure 3. Fluorescence of CN from a $\text{CH}_4/\text{N}_2\text{O}$ flame. The $\Delta v=0,1$ CN transitions and the laser line can be readily observed.

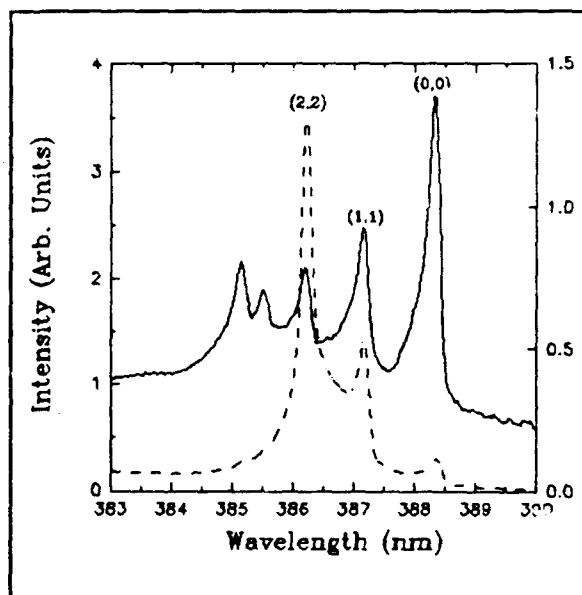


Figure 4. Emission and fluorescence of CN in a $\text{CH}_4/\text{N}_2\text{O}$ flame, expanded scale. Only the $\Delta v=0$ sequence can be observed at this resolution.

Table 1. Ar⁺ laser pumped fluorescence excitation of selected diatomic combustion radicals.*

Discrete Ar ⁺ Laser Lines (nm)	Species	Excitation Transition
528.7	-	-
514.5	C ₂	d ³ π _g - a ³ π _u (0,0) R ₂ (10) + Q ₁ (20)
501.7	C ₂	d ³ π _g - a ³ π _u (1,1)R ₂ (46) and (0,0)R ₂ (54)
496.3	-	-
488.0	-	-
476.5	-	-
472.7	C ₂	d ³ π _g - a ³ π _u (1,0)R ₁ (5)
465.8	C ₂	d ³ π _g - a ³ π _u (2,1) R ₂ (34) + R ₃ (34)
454.5	CN	B ² Σ ⁺ - X ² Σ ⁺ (1,3) R ₁ (20) + R ₂ (20)
** 379.5	CN	B ² Σ ⁺ - X ² Σ ⁺ (2,2)
** 363.8	-	-
** 351.4	-	-
** 351.1	-	-
** 335.8	NH	A ³ π - X ³ Σ (0,0)
** 334.5	-	-
** 333.6	-	-
** 305.5	-	-
** 302.4	OH	A ² Σ - X ² π (0,0) S ₂₁ (12)
** 300.2	-	-
** 275.4	-	-

* Rotational transitions are given in terms of the ground state rotational quantum number N"

** Present work

made this line lase in a pulsed dc discharge of argon. It was found that this line excites CN; the fluorescence excitation spectra are shown in Figs. 3 and 4 and described in Table 1. Note that all of the fluorescence excitation spectra presented here have the flame

emission contribution subtracted; and when there are two spectra on the same graph the vertical axis for the dashed line is on the right. Table 1 contains the laser lines, the combustion diatomic species excited and the excitation transition. Fluorescence excitation of CN together with the laser excitation line are clearly seen on Fig. 3. The wavelength coverage is sufficient to show both the $\Delta v = 0$ and 1 transitions for CN. The grating in the monochromator was changed from 1200 to 2400 grooves/mm for the spectrum shown in Fig. 4 and the $\Delta v = 0$ region for CN is shown with better detail. The solid line represents only the CN flame emission whereas the dotted line represents only the laser excited fluorescence from CN. It can be clearly seen that the predominant P-branch fluorescence peaks for $\Delta v = 0$ and 1 occurs in the (2,2) and (2,1) bandheads of CN, respectively. The R-branch excitation can be observed for the $\Delta v = 1$ sequence but is hidden under the laser line in the $\Delta v = 0$ sequence. It is clear that the primary fluorescence is coming from the $v' = 2$ (excited electronic) state. The energy of the laser line (379.5 nm) suggests that some rotational line in $v'' = 2$ (ground electronic) state is being pumped. A program⁸ which generates CN spectra from spectral constants indicates that the laser excitation line pumps the $B^2\Sigma - X^2\Sigma$ (2,2) $N = 55$ transition of CN. These results are not consistent. That is, although the calculation gives an R-branch transition within experimental error of the laser line, it gives a wavelength for the corresponding P-branch transition which should be resolvable from the P-bandhead; and this is not experimentally observed. Consequently, the rotational level being pumped has not been assigned. One explanation for the failure to determine the proper rotational quantum number is that the spectral constants used in the generation of CN line positions cannot be reliably extrapolated to such high N values.

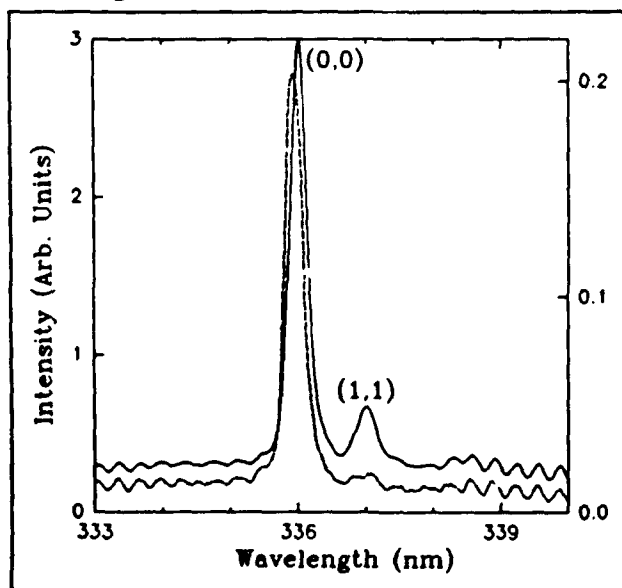


Figure 5. Fluorescence and emission of NH in a $\text{CH}_4/\text{N}_2\text{O}$ flame.

The 335.8 nm Ar^+ laser line is very close to the $A^3\Pi - X^3\Sigma$ (0,0) bandhead of NH and thus it is not surprising that this line pumps NH. Computer generated NH spectra⁸ indicate that there are 16 transitions within 0.05 nm of this laser line. These are transitions where the value of N ranges from 1 - 8. The NH emission from the flame is shown as a solid line on Fig. 5 and the (0,0) and (1,1) vibrational bandheads are labelled. The laser induced fluorescence and laser line are shown as the dashed line. The peak shifted slightly to the blue of the (0,0) emission bandhead contains contributions from both fluorescence and the laser line. No fluorescence was observed for NH in the $\text{CH}_4/\text{N}_2\text{O}$ flame when using either the 334.5 or the 333.6 nm laser lines. These

results are tabulated in Table 1.

There are three prominent Ar^+ uv laser lines around 300 nm which we made lase simultaneously with properly coated mirrors. We could not, however, obtain prism selected single line lasing. Thus, for the OH fluorescence results, the excitation line is

inferred from the prominent fluorescence lines. Both OH flame emission and laser induced fluorescence can be seen in Fig. 6 where again the emission is a solid line and the fluorescence a dashed line. The wavelength range included in Fig. 6 shows only one of the laser lines, which is at 305.5 nm. The other two laser lines are at 300.25 and 302.40 nm. The line that appears to pump OH is the 302.40 line which matches closely with the $S_{21}(12)$ transition. This transition wavelength is given as 302.399 nm by Dieke and Crosswhite⁹. Excitation here would produce fluorescence in the $R_2(13)$, $Q_2(14)$ and the $P_2(15)$ lines which occur at 306.97, 311.68 and 316.96 nm, respectively⁹. The locations of these lines coincide very well with the observed fluorescence emission peaks which are shown by the dashed line in Fig. 6.

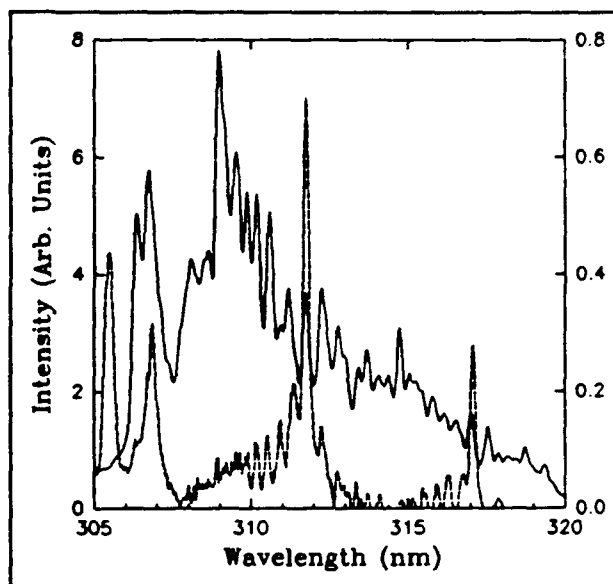


Figure 6. Fluorescence and emission of OH from a CH_4/N_2O flame.

Twelve out of the twenty lines listed in Table 1 produced no noticeable excitation of the hot combustion diatomics (C_2 , CN, CH, OH and NH). None of the Ar^+ laser lines excited CH. It should again be mentioned that for the Ar^+ uv laser lines below 350 nm the experiment was conducted extracavity. This arrangement produces low laser power which results in weaker fluorescence signals.

IV. CONCLUDING REMARKS

When comparing fluorescence excitation sources, prism selected lines of ion lasers are much simpler to use than various forms of tunable dye lasers. With this in mind, two situations are envisioned in which incorporating ion laser excitation as described in this paper would be the preferred choice: (a) experiments where the most important criterion is to track a species by its fluorescence signature irrespective of what transition or transitions are pumped and (b) experiments where the coincidentally pumped transition is of importance in the study.

This technique has been previously used to detect CN^1 in a propellant flame burning in room air and we have used the 379.5 nm laser line in attempts to spatially profile CN in a propellant flame burning at 1.5 MPa nitrogen pressure. Thus far a clean well-defined fluorescence signal has not been seen and changes such as lowering the pressure and/or incorporating optics up for intracavity lasing may be necessary.

This technique is not limited to diatomic species. For larger species the number of transitions dramatically increases, and for two triatomics, NCO^{10} and NH_2^{11} there have been coincidental excitations reported for Ar^+ prism selected lines.

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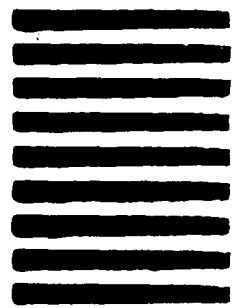


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