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LASER SPECTROSCOPY OF PLASMAS(U) CALIFORNIA UNIV  
BERKELEY DEPT OF MECHANICAL ENGINEERING J W DAILY  
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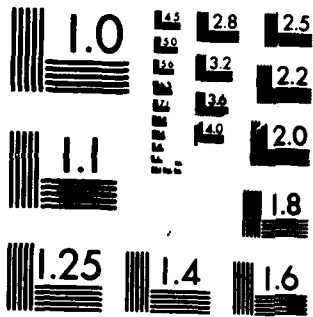
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| <p>During the past year, work was initiated to develop novel advanced laser spectroscopy plasma diagnostic methods. The methods are based on observing the doppler shift in the absorption lines of ionic species. Two methods under study are Velocity Modulated Laser Spectroscopy and Two-Beam Doppler Shift Laser Spectroscopy. The scientific goal of the work is to increase understanding of plasmas by making in-situ measurements of ion drift velocities, concentrations and temperatures in a non-intrusive fashion. The scientific approach is to combine conventional laser spectroscopies with velocity detection. Using a method such as Rayleigh, fluorescence, or Raman scattering, one probes the Doppler profile of the species of interest, observing shifts in the Doppler profile that arise because of the presence of a electric field. The shift may be related to the ion mobility, and thus conductivity, if the electric field is known, or to the electric field if the mobility is known. Temperature and concentration may be recovered by the conventional means. <i>Keywords:</i></p> |   |
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## I) Introduction

The purpose of our work has been to develop advanced laser spectroscopy methods to diagnose partially ionized plasmas. We have focused on methods that are based on observing the Doppler shift in ionic spectra due to the presence of an ion drift velocity. Two particular methods we are working with are Velocity Modulated Laser Spectroscopy (VMLS) and Two Beam Doppler Shift Laser Spectroscopy (TBDSLS).

The scientific goal of our work is to increase understanding of the role of flow non-uniformities and plasma/wall interactions in plasma devices by making in-situ measurements of electric field strength, ion mobilities, concentrations and temperatures in a non-intrusive fashion that allows point, one, and two dimensional imaging.

The scientific approach is to use conventional laser spectroscopic methods such as Rayleigh scattering, Raman scattering, or fluorescence, to probe ion absorption line profiles. If there is an electric field present, the ions will experience a net force and undergo drift, resulting in a shift in the position of the line profile. If the ion mobility is known, then the electric field component along the probe direction can be calculated. If the electric field driving the plasma is modulated, one will observe an oscillating shift in the line profile that arises because of the oscillating force imposed on the ions. The shift may be related to the ion mobility, thus conductivity.

Temperature and concentration may be recovered by conventional laser spectroscopic means. The methods are species

and state selective, allowing one to make measurements on more than one species and to study the effect of internal mode nonequilibrium.

The merit of the methods lies in their ability to provide simultaneous measurements of important parameters in plasmas. The methods are well suited to multi-dimensional imaging. One may use an array detector to image lines and planes in addition to the more conventional point configuration.

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## II) Theoretical Basis

The basic of Doppler shift methods for studying plasma conditions is that ionic species will experience a drift velocity when exposed to an electric field. The velocity experienced is of the form

$$v = \mu_i E$$

where  $\mu_i$  is the ion mobility. Since the observed frequency at which an atom or molecule radiates is shifted according to the Doppler relation

$$dv = (v_0/c) v$$

where  $v$  is the component of ion velocity along the line of sight, by measuring the shift in frequency of the ions absorption line one can obtain the drift velocity. By appropriate interpretation of the experimental results, one may then obtain information regarding the mobility and the electric field.

The configurations we are studying are Velocity Modulated Laser Spectroscopy (VMLS) and Two-Beam Doppler Shift Laser Spectroscopy (TBDSLS).

The VMLS configuration is useful for studying AC plasmas and for measuring mobility under known plasma conditions. As illustrated in Figure 1, one brings a laser beam into the plasma in a direction parallel to the electric field and then collects scattered fluorescence at ninety degrees. By using laser induced fluorescence to detect the ion and using a laser that has a line width narrow with respect to the absorption line width, the LIF signal can be used to probe the line. That is, as

# Velocity Modulated Laser Spectroscopy

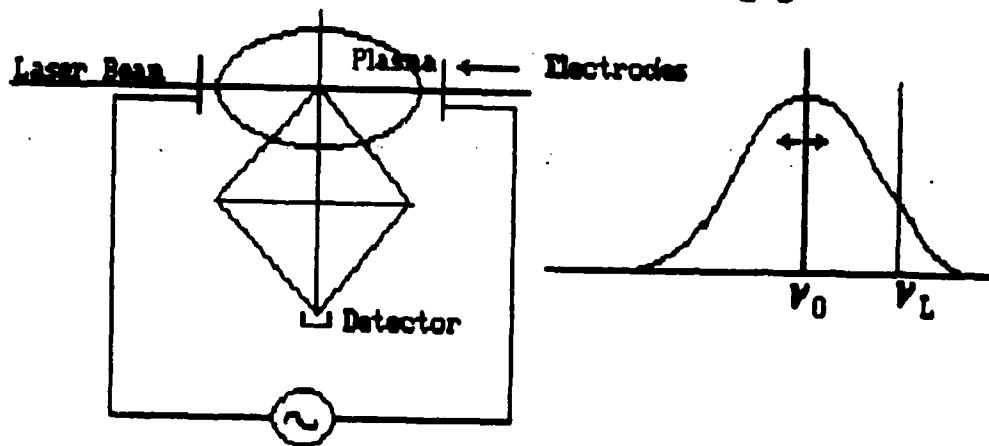


Figure 1

the laser frequency is scanned past the line, the signal will trace out the line shape. If one assumes that the line is Doppler broadened, then the signal will be proportional to

$$\text{Sig} \propto (2\sqrt{\ln 2}/\Delta v_D/\pi) \exp(-((2\sqrt{\ln 2}/\Delta v_D)(v-v_0))^2)$$

The effect of velocity modulation on the Doppler profile is felt as a modulation of  $v_0$ . By differentiating the signal twice, one obtains the result that the signal is most sensitive to changes in  $v_0$  when  $v_L = v_0 \pm 0.425 \Delta v_D$  and the fractional sensitivity is

$$d\text{Sig}/\text{Sig} = 2.35 dv_0/\Delta v_D$$

One may recast this expression into a detectability limit for the detection of a given modulation voltage

$$E_{0\text{Det}} = 1.08 (Q/q) P (d\text{Sig}/\text{Sig})_{\text{Det}}$$

If one assumes a moderate one percent for the signal detectability limit, and a typical ten square Angstroms for the collision cross section, then one obtains

$$E_{0\text{Det}}(\text{V/m}) = 0.00675 P(\text{Pa})$$

At one atmosphere, the detectable electric field is only 6.75 V/cm, a field readily achievable in the laboratory. By utilizing phase sensitive detection, one can achieve significantly lower detectability limits.

In principle, any method which probes the Doppler profile with sufficient resolution can be used to determine a DC line

shift. If only one probe is used, however, there may be a problem of absolute frequency calibration. By using two beams in opposite directions, an in-situ calibration is achieved.

Consider the arrangement illustrated in Figure 2. Assume that there is a drift velocity in the positive x direction. If the laser frequency is scanned through the line, then the rightward running beam will probe the profile labeled R in the lower figure, while the leftward running wave will probe the profile labeled L.

As the laser frequency is scanned through the line (or lines as it were,) from  $\nu < \nu_0$  to  $\nu > \nu_0$ , signal will first appear from the right running beam. As the frequency is increased, the leftward running beam will begin to contribute. If the two signals are independently observed, they will look like that of Figure 2. (By chopping the beams at a rate fast compared to the frequency scan rate, one may use the same imaging optics to detect both signals.) The peak separation is just twice the Doppler shift and the crossing point of the two signals is the unshifted line center.

The detectability limit for the method is determined by the resolution with which the distance between the two peaks can be resolved. If the precision with which the line shape signal can be measured is  $\Delta s$ , then for Doppler broadening the precision with which the separation in peaks, and thus electric field, can be resolved is

$$\Delta E/E = (1/\sqrt{2 \ln 2}) (\Delta \nu_D / \Delta \nu_E) (\Delta s/s)^{1/2}$$

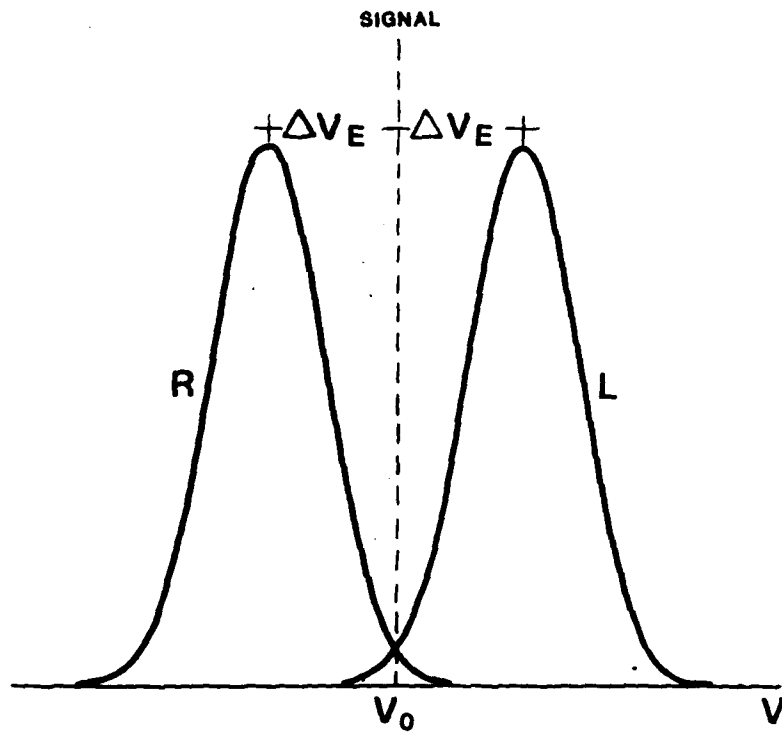
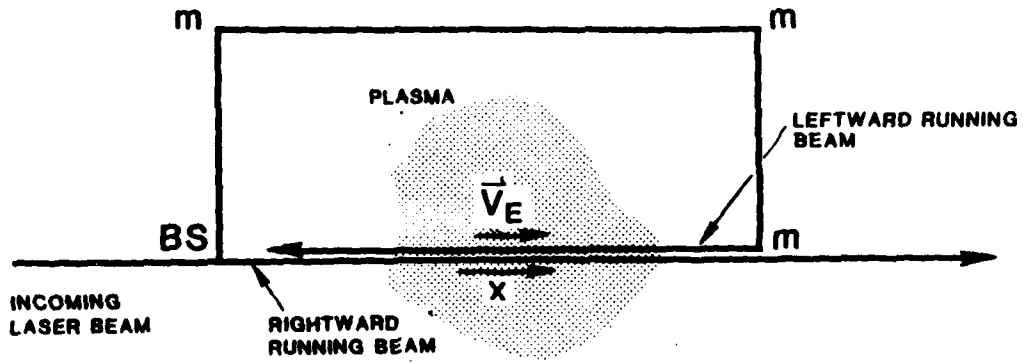


Figure 2

If we define the detectability limit as that value of E for which a precision of 10% is achieved when the spectroscopic signal is detected with a precision of 1%, then we can write approximately

$$E_{\text{det}}(\text{V/cm}) \approx 0.04 P(\text{Bar})/T(\text{K})^{1/2}$$

(Assuming that  $Q \approx 10A^2$ ,  $m$  is the mass of a proton, and the line center is at 5000A.) Thus at a pressure of 0.01 Bar and 5000K, the detectability limit is about 1 V/cm.

It should be noted that least squares fitting the line shape to the appropriate profile as suggested above, would significantly increase the precision with which the line center is detected and thus reduce the detectability limit for the electric field. Also, if a suitable transition is found, saturation spectroscopy could be used to locate the line peak with high precision, although the in situ calibration for the zero shift position would be lost.

#### IV) Progress

Our grant initiated in December, 1985. During the first year of the grant we have worked on developing the theory for the methods and assembling the equipment to begin experiments to demonstrate Velocity Modulated Laser Spectroscopy (VMLS) and Two Beam Doppler Shift Laser Spectroscopy (TBDSLS).

##### Theory

The theory necessary for application of Doppler methods includes two major aspects. These are the relationship between the optical probing signal and the concentration of ions with a specific velocity component and the relationship between the velocity distribution of the ionic specie and the plasma conditions.

For laser induced fluorescence as the probe method, there is extensive documentation on the relationship between the signal and the population of the velocity class being probed, much of it worked out for flames by our group. We have adopted this theory for the present case and worked out detectability limits, etc. As indicated in the previous section. calculations show that the methods will lead to excellent detectability for measurement of electric field (or ion mobility), species concentration, and temperature.

To determine the relationship between the measured velocity or velocity distribution of a ionic species and the plasma state requires a model of the plasma. The equations which describe the plasma dynamics depend in kind and complexity on whether the

plasma or its electrode sheaths can be treated as continuum or rarified flow. Questions of interest include the nature of the ion velocity distribution, spatial distribution of drift velocity, and variations in concentration and temperature. If convection is important, then the mass average velocity of the plasma as a whole must also be known to extract the drift velocity.

In developing the diagnostic, the approach we are taking is that we should start working in an environment that is fairly well defined. To that end we have chosen to work with a flame assisted plasma at a pressure high enough to ensure continuum behavior throughout. The flame assisted plasma, when seeded with a readily ionizable atom such as barium, offers a rapid ionization rate that is dominated by neutral collisions and a three body recombination rate for which the third body is also dominantly neutrals. Furthermore, by suitable arrangement, the convective velocities in the direction parallel to the applied electric field can be kept at a minimum.

We have worked out a simplified theory of the flame assisted plasma, and have started to develop a numerical model to solve the more complete equations.

### Experimental

We have assembled an experimental system. We have an argon ion laser (Spectra Physics 171-19) pumped ring dye laser (Coherent 699-21). The ring laser is fully stabilized and has a line width of less than one megahertz. The output of the laser

is directed through a Fabry-Perot interferometer (Coherent Model 251 Spectrum Analyzer) as a means of measuring the relative wavelength precisely. A small portion of the beam is also split off and sent to a wavemeter (Burleigh Model WA 10) for absolute wavelength calibration.

The beam is then directed into the test section in either the VMLS or TBDSLS configuration. In either case, the beam (or beams) pass through small holes in the electrodes. A power supply can deliver several hundred volts DC modulated at up to 100 VAC.

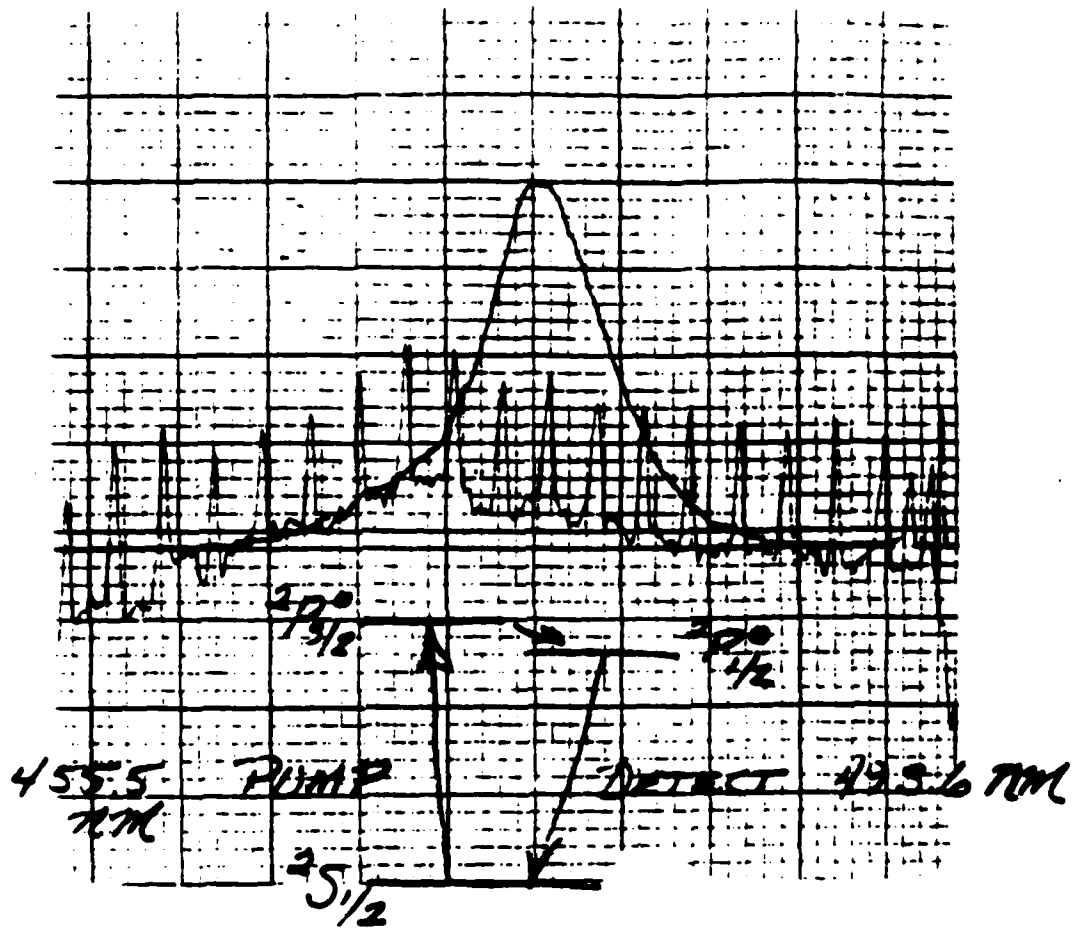
The signal is detected by observing the fluorescence at ninety degrees. The signal is collected with F/9 optics and passed through a 1/4 meter Jarrell-Ash monochromator. A RCA 1P28 photomultiplier is used as the detector.

The plasma used in the proof of principle experiments is a flame generated, partially ionized plasma in which the dominant ion is barium. Barium was chosen as the test ion because of the low ionization potential of the neutral and because it has a convenient structure with readily accessible absorption lines. We pump at 455 nm and detect collisionally induced fluorescence at 493.5 nm to avoid scattering interference.

After considerable experimenting with different burners we have chosen a capillary, diffusion flame burner designed by Krupa, et. al. of the University of Florida. The burner, manufactured in our shop, allows a wide range of operating conditions without the normal difficulties associated with using acetelyne and oxygen as reactants. The burner is mounted in a

low pressure vessel and can be operated down to about 20 Torr. Barium tetrachloride is seeded into the flame using a Perkin-Elmer aspirator modified to operate at lower pressures.

In our first experiments we observed fluorescence from the barium ion and measured the 455 nm line width at atmospheric pressure. At atmospheric pressure the linewidth is dominated by pressure broadening. Figure 3 shows a typical line shape profile. The series of sharp peaks is the Fabry-Perot signal superimposed to provide a measure of the frequency. The peaks are 1.5 GHz apart, thus the line is about 4.5 GHz wide. We have revised the detectability calculations to account for the pressure broadening. We started the new year conducting experiments at low pressures.



LIF Signal at atmospheric pressure

Figure 3

VI) Publications

a) AFOSR sponsored refereed publications appearing or accepted during the past grant period:

Daily, J. W., "Electric Field Measurement by Two-Beam Doppler Shift Spectroscopy," Applied Optics 25, 1378-1380 (1986).

b) Review paper in progress

Daily, J. W., "Laser Induced Fluorescence Spectroscopy in Flames," Invited review for Progress in Energy and Combustion Science, In progress.

## VII) Personnel

The principle investigator is Professor John W. Daily. Professor Daily is currently an Associate Professor of Mechanical Engineering (with tenure.)

At present two graduate students are working on the project. These are Mohamed Sassi, a Ph.D. student, and Carolyn Lee, a Masters student.

## VIII) Interactions

### a) Meetings

"Plasma Studies Using Doppler Shift Laser Spectroscopy,"  
AFOSR Contractors Meeting, Stanford University (16-17 June  
1986)

"Electric Field Diagnostics Using Doppler Shift  
Spectroscopy Meeting, Stanford University (16-17 June  
1986)

"Electric Field Diagnostics Using Doppler Shift  
Spectroscopy," Conference on Quantitative Spectroscopy and  
Laser Diagnostics, University of California, San Diego, La  
Jolla, CA (7-8 July 1986)

"Measurement of CH Radical Concentrations in an  
Acetylene/Oxygen Flame and Comparisons to Modeling  
Calculations," (With R. G. Joklik and W. J. Pitz) 21st  
International Symposium on Combustion, Munich, West Germany  
(3-8 August 1986)

"Plasma Diagnostics for Arcjet Plume Studies," Arcjet Plume  
Diagnostics Technical Workshop, Jet Propulsion Laboratory,  
Pasadena, California (2-3 October 1986)

"Doppler Shift Methods for Electric Field and Mobility  
Measurement in Plasmas," Invited paper, OSA Annual Meeting,  
Seattle, WA (21-25 October 1986)

### b) Seminars

"Combustion Diagnostics at Berkeley," Industrial Liason  
Program, UC Berkeley, CA (12 March 1986)

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