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CO_2 LASER SUSTAINED CW DISCHARGE ATOMIC BEAM SOURCE

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ABSTRACT

A high pressure, supersonic, laser sustained plasma nozzle beam source has been developed for the production of intense ($>10^{19}$ particles s^{-1}-sr^{-1}) beams of atomic and/or radical species having kinetic energies in the range of 1-10 eV. A high plasma temperature (10-30,000 K) is produced in the throat of a hydrodynamic expansion nozzle by sustaining a cw optical discharge in a gas using a high power cw CO_2 laser. Gas mixtures are expanded through the nozzle/discharge region creating energetic atoms and molecules. An oxygen atom beam has been produced with a kinetic energy of 2-3 eV and an intensity of $\sim 10^{18}$ O-atoms $s^{-1}sr^{-1}$. O-atom collisions (1 eV) from an uncharacterized nickel surface shows strong specular scattering with approximately 50% energy loss to the surface. Argon beams having kinetic energies between 5-10 eV with intensities of $>10^{19}$ atoms $s^{-1}sr^{-1}$ have also been produced.

I. INTRODUCTION

The production of intense beams of low mass, highly reactive species in the energy range of 2-10 eV is difficult due to reactions of the species with the production apparatus and the characteristics of the production methods. For example, a source developed by Knuth[1] has been used for the production of atomic oxygen[2] but due to reactions with the electrodes, the oxygen is admitted down stream of the discharge region resulting in a maximum kinetic energy of 1 eV and an intensity of 3×10^{17} $s^{-1}sr^{-1}$. Radio frequency discharges[3,4] have been used to produce oxygen atoms with kinetic energies of 0.1-1.0 eV and intensities of 10^{17-18} $s^{-1}sr^{-1}$. The production of high mass high kinetic energy species is accomplished by seeding and heating in helium[5] expansions but light species (<20 amu) are limited to roughly 1-2 eV in kinetic energy. Charge exchange methods excel at energies > 100 eV but suffer from space charge limitations[6] below 10 ev producing beam intensities orders of magnitude less than the previously mentioned techniques. Due to the low duty cycle of pulsed beams, techniques using pulsed laser breakdown require peak intensities of 10^3-10^4 that of cw beams in order to effectively equal cw time averaged intensities. The plasmas mentioned above are produced by

electric fields having a range of frequencies from constant (d.c. arcs). <1 kHz (a.c. arcs), 20-50 MHZ (R.F.), and 2.5 GHZ (microwaves). These sources require some physical device to support the plasma. D.C. arcs require electrodes, rf plasmas require an induction coil, and microwaves require a resonator or waveguide. Because rf and microwave plasma heating occures by direct plasma-electric field interactions characterized by large absorption coefficients with the outer layers of the plasma, these modes of plasma production are characterized by low power density (<200 W cm-3), modest temperatures (<8000 K except for d.c.arcs-20,000K). Power inputs ranging from 15 kW for d.c. arcs to 100's of watts for RF and microwave plasmas are required.

II. LASER SUSTAINED PLASMAS

In the early 1970's, it was hypothesized [7,8] and then demonstrated that a free-standing continuous discharge could be produced by focusing the output of a sufficiently powerful cw-CO_2 laser in inert[9] and molecular[10] gases at one atmosphere or above. The discharge resides near the focus of the laser and operates above the plasma frequency at 3 THz where the electric fields interact with individual electrons and ions to heat the plasma via free-free transitions (inverse Bremsstrahlung)[11].

The laser power maintenance threshold depends upon the type of gas, the total pressure, whether the laser beam is horizontal or vertical (convection sweeps the hot gas out of the laser focal volume), the focusability or coherence of the laser beam, and the optical quality of the lens system. For example, our work uses a 1 inch focal length ZnSe meniscus AR coated lens operated in the horizontal position with a transverse flow 1.5 kW CO_2 laser. The gases xenon,argon, and neon require 50, 300, and 1300 watts respectively for maintenance of the discharge. Because the focused power of the cw laser is in the range 10^6-10^7 W cm^{-2} (i.e. several orders of magnitude smaller than typical breakdown thresholds) a high energy external spark is needed to initiate the discharge. This can be provided by a conventional electric spark[12] or, as in this work, a pulsed CO_2 laser. The primary advantages of the laser sustained discharge in creating energetic atomic beams are the high temperatures produced by the high power densities (10^4 W cm^{-3}), the ability to sustain the discharge independant of nozzle material, and low total input power. Preliminary results using xenon have been reported in Ref.12.

III. SOURCE CONSTRUCTION AND OPERATION

A cross sectional view of the source is shown in Fig. 1 and consists of two portions, the lens holder and nozzle holder. A 1 inch focal length 1 inch diameter ZnSe AR coated meniscus lens is clamped to the end of a water cooled copper tube. Indium gaskets are used to cushion and seal the lens as well as to provide maximum heat transfer to the copper assembly. A threaded copper clamping ring is tightened onto the lens while heating the lens, holder, and indium assembly to 50-70 C. This procedure provides excellent sealing of the lens onto the indium. The nozzle holder is made entirely of copper with all joints being welded rather than brazed. The nozzle body is made from a 3.2 mm platinum rod 3.2 mm long, drilled to within 0.76 mm of one end with a diameter of 2.39 mm. The nozzle body is brazed into the end of the nozzle holder and a 0.2 mm diameter nozzle is then electron discharged machined through the 0.76 mm wall. A 3 mm thick copper wall separates the platinum nozzle from the water cooling jacket. A double viton O-ring is used to seal the lens holder to the nozzle holder and to locate the lens concentric with the nozzle.

Fig.1. Laser Sustained Discharge Atomic Beam Source: Discharge is initiated using a pulsed (0.5j) TEA CO_2 laser and sustained with a 1.5 kW CO_2 laser.
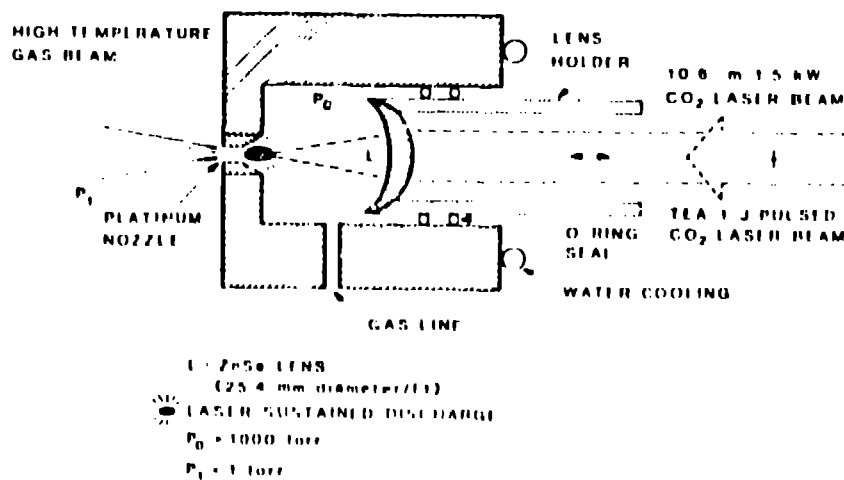
Fig. 2 shows the source mounted in the molecular beam scattering apparatus aligned with both the plasma sustaining cw (maximum power 1.8 kW) and plasma initiating pulsed (0.5 joule) CO_2 lasers. The output of the cw laser traverses the length of a 3.2 m laser table and is reflected back and turned 45 degrees to enter the source assembly. Both copper turning mirrors (M1 and M2) are water cooled. The pulsed laser beam is placed nearly coaxial to the cw beam using the set of copper coated glass mirrors (M3, M4, and M5). Initial alignment of the cw, pulsed, and HeNe laser beams is accomplished by burning a pin hole in a 0.1mm thick nickel foil using the cw laser and then adjusting the pulsed laser turning mirrors to place it through the same hole. The cw CO_2 laser is then turned off, the mirror M6 placed in the path of the cw beam, and the HeNe laser is aligned to pass through the pin hole.

Fig. 2 Alignment of pulsed and cw CO_2 lasers with the nozzle. The 5 cm diameter water cooled mirrors M1 and M2 are used to align both the pulsed and cw lasers with the nozzle. Mirrors M3, M4, and M5 are used to align the pulsed laser with the cw laser. Mirror M6 employs a kinematic magnetic mount to place the HeNe laser beam coaxial to the cw laser beam. When operating the cw laser mirror M6 is removed. The photocell detects the plasma light emitted from the nozzle assembly and interrupts the cw laser operation if the plasma is extinguished.
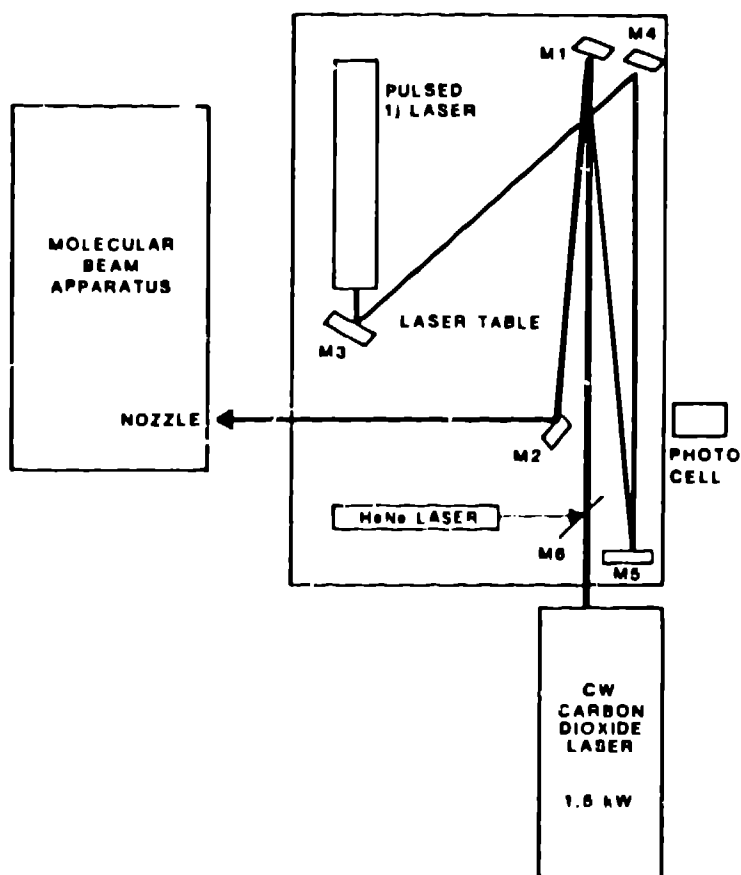

Final alignment of the cw laser beam with the nozzle is accomplished through operation of the source with argon and optimizing the time of flight distributions for maximum velocity by

moving the discharge radially using the final turning mirrors M1 and M2 and axially by movement of the ZnSe lens. After initiating and aligning the discharge using argon, other gases are mixed with the argon to obtain radical species with velocities < 5 km/s. If velocities greater than 5 km/s are desired, the argon is replaced with neon yielding velocities <10 Km/s.

III. BEAM CHARACTERIZATION

Beam characteristics are depicted in the TOF distributions of Figures 3,4, and 5. The time of flight (TOF) analyzer used for these studies consisted of a 12.5 cm diameter disk rotated at 310 Hz with 8 equally spaced 1 mm slots located on its circumference. The TOF analyzer is calibrated using low pressure (200 torr) room temperature expansions of neon, argon and krypton gases. The ion energy in the quadrupole mass spectrometer detector[12] was found to be 10 eV (25 cm path length) and the neutral flight path length was 19.5 cm. The entrance to the detector was a 0.152 mm diameter round hole. Slots (0.2 mm wide) below each 1 mm slot are used to obtain timing signals from a light bulb and photocell detector. This timing signal is used to control a 256 channel multichannel scaler having a 2 microsecond dwell time/channel which was used to record the time of flight (TOF) spectra. The TOF spectra shown have not been corrected for ion flight time, timing mark offset (12 microseconds), or instrumental broadening. The reported velocities were obtained by correcting for ion flight times and timing mark offsets but not instrumental broadening.

Fig. 3 Argon and neon time of flight distributions. Flight times have not been corrected for timing mark offset or detector ion flight times. The source pressures are indicated along with the calculated source temperature corresponding to the observed peak velocities.
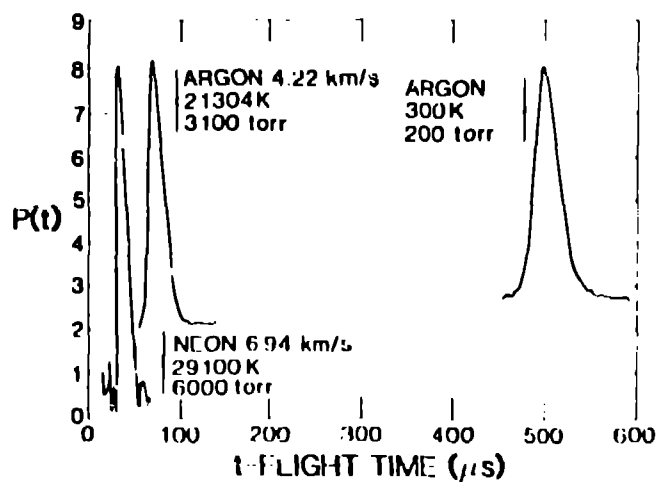
The calculation of molecular dissociation follows that of Lee[3].

$$R = \frac{N_O}{N_{O_2}} = \frac{(x_D/x_{O_2})}{n} \left(\frac{x_{O_2}}{x_O}\right) \left(\frac{I_O - n\, I_{O_2}}{I_{O_2}}\right) \tag{1}$$

Percent Dissociation = R/(R+2) (2)

where I_O and I_{O_2} are the experimentally observed number density signals at mass 16 and 32 with the discharge on, n is the ratio of number densities of mass 16 and 32 with the discharge off, x_D is the dissociative ionization cross section of O_2 to form O^+ taken as $x_D = 0.88\ A^2$, and the x_i are the ionization cross sections which we have taken as $x_{O_2} = 1.52\ A^2$ and $x_O = 1.15\ A^2$.

Figure 3 shows argon TOF distributions with the discharge off and on. The argon velocity with the discharge on was calculated to be 4.2 km/s and the neon velocity of 6.9 Km/sec was found with the discharge backed away from the nozzle. Our previous work[12] with xenon predicted that argon and neon velocities would be 3.6 km/s and 6 km/s respectively, indicating that a crude estimate of other gas velocities (V_m) can be obtained using the formula[12]

$$V_m = V_{Ar}\,(40/m)^{1/2}\ (T_m/T_{Ar})^{1/2} \tag{3}$$

where m is the mass of a carrier gas, the subscript Ar refers to argon, and T_m is the plasma spectroscopic temperature[10] of the carrier gas.

Figures 4 and 5 show TOF spectra for mass 16 and 32 with oxygen mixtures in argon of 40%, and 49% respectively. Essentially 100% dissociation of O_2 into O-atoms was observed with the 40% oxygen/argon mixture while a 49% mixture produces 98% dissociation indicating that increasing amounts of oxygen may produce recombination within the nozzle.

The extent of dissociation is highly dependent upon the placement of the discharge within the nozzle; i.e. small changes in the radial or axial position can easily produce ratios of O-atoms to O_2 of 50%. Figure 6 shows the effect that a 0.5 mm axial change in the discharge placement in the nozzle has on the gas velocity distribution. As the discharge is moved farther into the nozzle, the velocity distribution becomes broader and peaked at higher velocities. The plasma acts as a plug when placed in the nozzle and the initial gas density (10^{20} cm^{-3}) drops by a factor
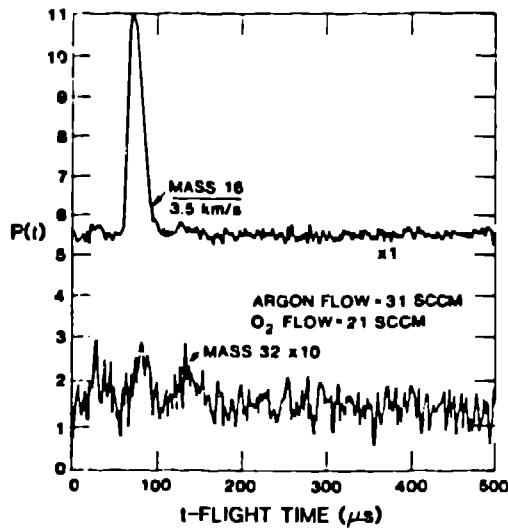
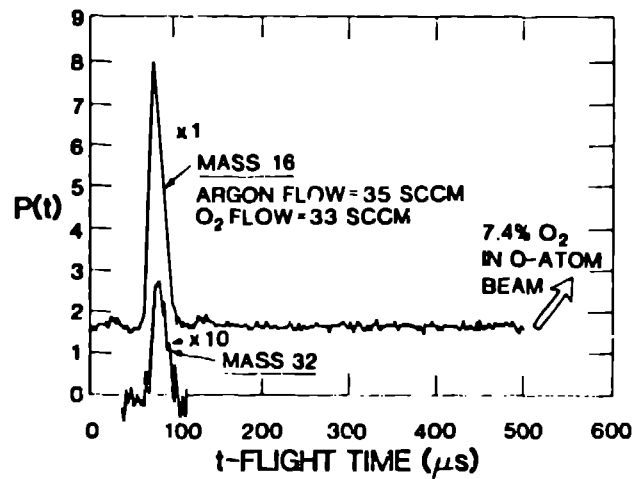Fig. 4. TOF Distribution of mass 16 and 32 using a 40% O_2, 60% argon mixture.

Fig. 5. TOF Distribution of mass 16 and 32 using a 49% O_2, 51% argon mixture.

of roughly 100 to values of 10^{18} to 10^{17} cm^{-3} due to the high temperature of the plasma. At the higher plasma temperature a decrease in the total collision cross section would also be expected[13]. This creates a condition in which the nozzle is operating at a Knudsen number close to unity or in the transition region between hydrodynamic and free molecular flow thus causing a broadening of the velocity distribution and lowering of the Mach number. The higher peak velocities are observed because cooler boundary layers in front of the discharge are reduced in intensity.

Fig. 6. TOF distributions of mass 16 using a 30% O_2, 70% argon mixture Distribution A was taken with the plasma at the entrance to the nozzle while distribution B was taken with the plasma moved 0.5 mm farther into the nozzle.
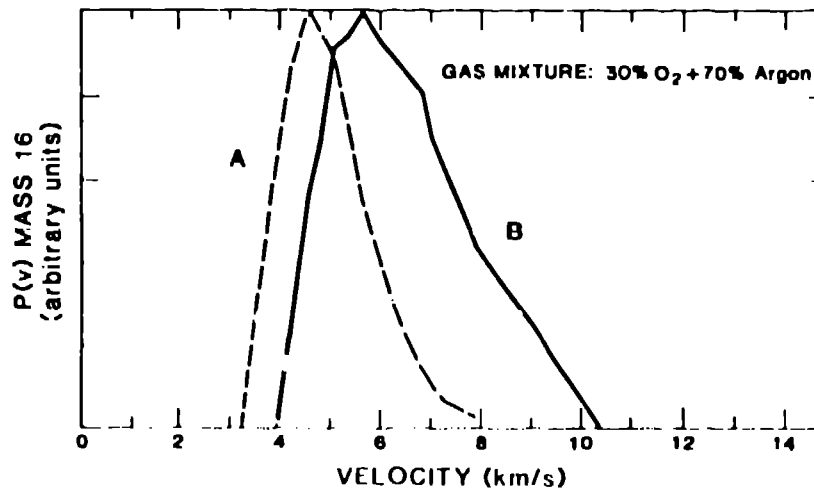
IV. GAS-SURFACE SCATTERING

Initial results of gas scattering from an uncharacterized Ni(111) surface are presented in Figures 7 and 8. The molecular beam apparatus described in reference 12 was used along with a pseudorandom sequence TOF chopper. The TOF detector is operated at 400 Hz with the multichannel scaler operating with a dwell time/channel of 10 microseconds. Angular distributions were obtained by modulating the direct beam at 400 Hz with a tuning fork chopper while data was accumulated with a phase locked pulse counter. The TOF chopper was kept in operation during angular distribution measurements. Counting times of 2 min/angle were used for angular distributions and 5 min/angle for TOF acquisition. These initial experiments focused on large incident angle (70 degree) scattering because of the ease of observing both the direct and scattered beams. The discharge source was operated with a 50% mixture of oxygen and argon with the axial position of the discharge slightly back of the nozzle (velocity was not optimized).
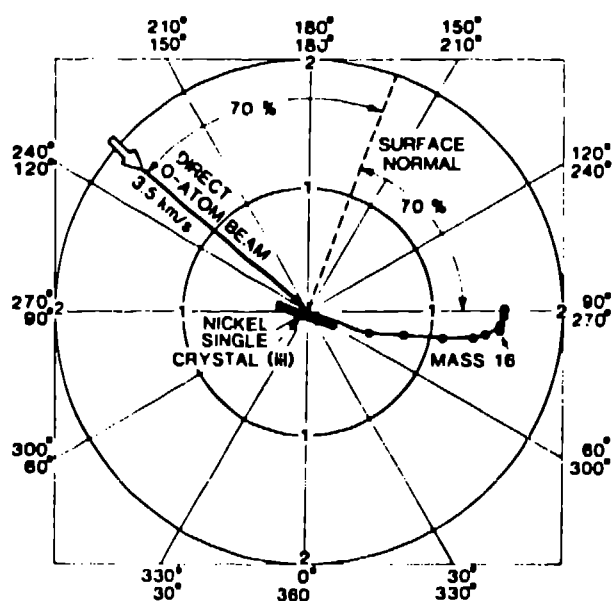
Fig. 7. Angular distribution of O-atoms scattered from an uncharacterized nickel crystal. Error bars on the data points are approximately the size of the plotting symbol. The surface temperature was 300 K
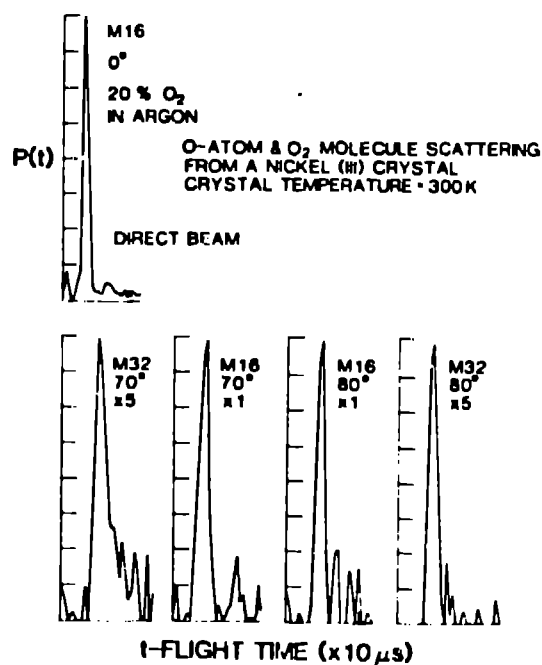
Fig. 8. TOF distributions at 70 and 80 degrees from surface normal. The mass 16 distribution when converted to energy indicates that roughly 50% of the energy was transferred to the solid.

Figure 7 shows strong specular scattering of atomic oxygen over the angular range accessible to the detector indicating predominently direct scattering with surface residence times on the order of the collision time. Figure 8 shows TOF spectra taken at the specular angle 70 degrees and at 80 degrees from the surface normal for both atomic and molecular oxygen. The data of Figure 8 when converted to translational energy indicates that approximately 1/2 the initial beam energy was lost to the solid. The Ni surface was not characterized but most likely consisted of nickel oxide with overlayers of O_2. Further experiments are in progress to obtain angular distributions near the surface normal (to determine the extent of energy accomodation) and to fully characterize the actual surface.

V. CONCLUSION

A new beam source has been described which uses a laser sustained plasma technique for producing high intensity ($>10^{19}$ particles $s^{-1}sr^{-1}$) and high tranlational energy ($>1eV$) beams. Data indicates that beam temperatures near the plasma spectroscopic temperature can be obtained and that data from one rare gas can be used to predict results from others. Atomic oxygen beam energies of 2.5-3 eV have been produced with intensities of $\sim 10^{18}$ $s^{-1}sr^{-1}$.

REFERENCES

[1]   Knuth, E.L.; Winicur, D.H. : J. Chem. Phys. 46(1967) 4318.
       Knuth, E.L.; Rodgers, W.E.; Young,W.S.: An Arc Heater for Supersonic Molecular Beams,
       Rev. Sci. Instr. 40 (1969) 1346.

[2]   Silver, J.A.; Freedman, A.; Kolb, C.E.; Rahbee, A.; Dolan, C.P.: Supersonic Nozzle Beam
       Source of Atomic Oxygen Produced by Electric Discharge Heating; Rev. Sci. Instr.
       53(1982) 1714.

[3]   Lee, Y.T.; Ng, C.Y.; Buss, R.J.; Sibener, S.J.: Development of a Supersonic $O(^3P_J)$,
       $O(^1D_2)$Atomic Oxygen Nozzle Beam Source, Rev. Sci. Instr. 51(1980)167.

[4]   Grice, R.; Gorry,P.A.: Microwave Discharge Source for the Production of Supersonic Atom and Free Radical Beams, J. Phys. $\underline{E12}$(1979) 857.

[5]   Campargue, R.: Progress In Overexpanded Supersonic Jets and Skimmed Molecular Beams In Free-Jet Zones of Silence; J. Phys. Chem. $\underline{88}$(1984)4466.

[b]   Ardenne, M.V.: *Tabellen der Elektronenphysik, Ionenphysik and Ubermikroskopie.*, Veb Deutscher kVerlag Der Wissenschafter, Berlin (1956). Pg. 507

[7]   Razier, Y.P.: The Feasibility of an Optical Plasmatron and It's Power Requirements; ZhETF Pis. Red. $\underline{11}$(1970)195{JETP Let. $\underline{11}$(1970)120}.

[8]   Razier, Y.P.: Subsonic Propagation of a Light Spark and Threshold Conditions for Maintenance of a Plasma by Radiation; Zh. Eksp. Teor. Fiz. $\underline{58}$(1970)2127{Sov. Phys. JETP, $\underline{31}$(1970)1148}.

[9]   Razier, Y.P.: *Laser Induced Discharge Phenomena.* Consultants Bureau, New York (1977).

[10] Kozlov, G.I.; Kuznetsov, V.A.; Nasyukov, V.A.: Sustained Optical Discharge in Molecular Gases; Zh.Tekh.Fiz.,$\underline{49}$(1979)2304{Sov. Phys. Tech. $\underline{49}$(1979)1283).

[11] Hughes, T.P.: *Plasmas and Laser Light.* John Wiley, New York(1975).

[12] Cross, J.B.; Cremers, D.A.: High Kinetic Energy (1-10 eV) Laser Sustained Neutral Atom Beam Source; Nuc. Instr. and Methods, $\underline{B13}$(1986)658.

[13] Levine, R.D.; Bernstein, R.B.: *Molecular Reaction Dynamics*; Oxford University Press, New York(1974), pg. 25