# HIGH KINETIC ENERGY (1-10 eV) LASER SUSTAINED NEUTRAL ATOM BEAM SOURCE

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Development of high energy (1-10 eV) neutral atom beams has relied primarily on the use of charge exchange or DC/RF discharge techniques. A new source is described which uses a laser sustained plasma technique for producing high intensity  $(>10^{15}/\text{cm}^2 \text{ s})$  and high translational energy (>1 eV) atomic beam species for use in gas-surface scattering experiments. Laser sustained plasmas have demonstrated temperatures of 10 000 K in xenon, 18 000 K in argon, and a predicted temperature of 30 000 K in helium. Combining this plasma with hydrodynamic expansion techniques should produce atomic beam velocities greater than 10 km/s for many species. Initial experiments with xenon using 70 W of CO<sub>2</sub> laser power have demonstrated beam kinetic temperatures of 8–9000 K with Mach numbers of 4–6 resulting in peak velocities for xenon of 1.5 km/s (1.5 eV). Extrapolation of these results to helium predicts that velocities in excess of 10 km/s are possible but will require laser powers in excess of 1.3 kW.

## 1. Introduction

The production of intense  $(10^{19} / \text{s sr})$  high energy (1-10 eV) beams of heavy (>100 amu) molecules and/or atoms is straightforward using heated nozzle helium seeding techniques [1]. The production of intense high energy low mass (<40 amu) beams, however, is a difficult challenge. The two main techniques which use charge exchange [1] and DC arcs [2] have a number of disadvantages. Charge exchange suffers from intensity limitations at low energies (<100 eV) because of space charge defocusing. Although attempts to overcome this problem using electron neutralization of the beam have been attempted, this source is used primarily for producing large beam fluxes at energies greater than 100 eV. Direct current arcs [2] have been used extensively in gas phase scattering experiments but exhibit instabilities due to electrode erosion and have large power (10-20 kW) and water cooling requirements. This source also produces reduced velocities for reactive species (4 km/s for O-atoms) which have to be added to the lower temperature flow downstream of the arc in order to protect the electrodes from catastrophic erosion. Light species such as O-atoms produced by radio frequency [3a] and high pressure microwave sources [3b] are limited to about 1 eV translational energy.

## 2. Background

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It has been demonstrated (see ref. [4] for an extensive listing of laser sustained discharge references) that a free standing discharge of high temperature can be produced by focusing the beam from a sufficiently

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powerful cw-CO<sub>2</sub> laser (wavelength =  $10.6 \mu$ m) in inert and/or molecular gases at atmospheric pressures or greater. The discharge is located near the focal point of the laser beam and does not require electrodes or coupling coils for production. Since the focused power density of the cw-laser beam needed to sustain the discharge is  $10^6 - 10^7$  W/cm<sup>2</sup>, a factor of about 10 lower than typical gas breakdown power densities, a suitable spark is required for initiation. This can be provided with a Tesla-coil-electrode arrangement or a pulsed laser which produces a spark within the focal volume of the cw-laser beam. The pulsed plasma contains a high density  $(>10^{16} \text{ cm}^{-3})$  of electrons which act as an absorption center for the cw-laser beam. Once started, the plasma operates continuously as long as sufficient power is supplied to the focal volume. At laser frequencies (30 THz) which are an order of magnitude above the plasma frequency [5], absorption occurs primarily by free-free transitions (inverse bremsstrahlung) associated with electron-ion collisions. Power loss results from electron-ion recombination, radiation, and thermal conduction. The very high temperature of the laser sustained plasma is due to its higher power density as compared to conventional RF sources. At RF frequencies, which are below the plasma frequency, mainly the outer layer of the plasma is heated because of the very large cross section for direct EM wave plasma interaction with the interior being heated by conduction. A typical inductively coupled 1.5 kW plasma with a volume of 8 cm<sup>3</sup> has a power density of 188 W cm<sup>-3</sup> compared to  $5 \times 10^3 \,\mathrm{W \, cm^{-3}}$  for a 1 mm diameter plasma sustained by a 45 W laser beam. Since the laser sustained plasma is highly dependent on power density, the mode quality of the laser beam and the optical properties of the laser beam focusing system determine



the laser powers needed to sustain the discharge. Typical minimum laser powers needed to maintain the discharge using a simple ZnSe 2.54 cm focal length lens and a Gaussian laser beam also depend upon the properties of the gas (ionization potential, etc): xenon (50 W), argon (250 W), 50% oxygen-50% argon mixture (500 W), neon (1-1.3 kW) and helium (>1.3 kW). Spectroscopic temperatures of 10 000 K and 18 000 K have been measured [4] for xenon and argon. A temperature of 30 000 K is predicted for a helium plasma.

#### 3. Nozzle source and results

Fig. 1 shows schematically the beam source designed to test the idea of using the laser sustained discharge technique for producing extremely high temperature neutral atomic species for use in gas-surface scattering experiments. Since only a 70 W CO<sub>2</sub> laser was available for the time-of-flight (TOF) experiments, xenon was chosen as the primary carrier gas because of its low maintenance threshold power (low ionization potential). The tungsten nozzle was brazed into the copper body of the source and all other fittings and components were glued in place with high temperature epoxy. As the total power input was less than 70 W, only modest water cooling was needed to cool the lens and source body. To start the discharge, the ZnSe lens was positioned to place the focal volume between the high voltage and ground

Fig. 1. Laser sustained discharge atomic beam source. Main body is water cooled to prevent overheating. First experiments used a tungsten nozzle and 70 W of laser power in 3 atm of xenon. Later experiments using a 50% argon and 50% oxygen mixture at 500 W laser power required the development of a

electrodes, the system was pressurized to 3 atmospheres of xenon, the laser was turned on, and a Tesla coil was applied to the high voltage electrode. The spark formed between the electrodes provided a "seed" plasma to start the discharge. After the continuous discharge was operating, the lens was moved forward to position the plasma volume in the throat of the nozzle. A bright luminescence was observed downstream of the nozzle during operation due to emission from electronically excited Xe. Fig. 2 shows a portion of the xenon spectrum (upper trace) taken with a half-meter monochromator, the quenching (lower trace) of the xenon emission by the addition of oxygen to the xenon, and neutral atomic oxygen luminescence.

Time-of-flight analysis was performed on the xenon (fig. 3) using the apparatus shown in fig. 4 as a function of the distance between the discharge and the nozzle. The TOF distributions were converted to velocity space P(V) by performing a least squares fit to eq. (1) [1]

$$P(V) = c(V/A_0)^3 \exp - [V/A_0(1 + [B - 1]/2M^2)^{1/2} - (B/2)^{1/2}M]^2, \qquad (1)$$

where  $A_0 = (2kT/m)^{1/2}$ , c is a constant, V is velocity. B is the ratio of specific heats, M the Mach number, and m is the atomic mass, taking into account the Jacobian for the transformation from time to velocity space and that the detector output is proportional to number density. Equating the beam energy  $0.5mV^2$  to

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Fig. 2. Laser sustained discharge atomic spectrum. A section of the 3000-8000 Å spectrum is shown here. The pure xenon spectrum is shown at the top and the bottom spectrum shows the effect of added oxygen. The intense 5S-5P transition in the O I band is observed along with appreciable quenching of the xenon lines.

2kT where k is the Boltzmann constant and T the temperature, a best fit is determined for temperature and Mach number. Fig. 5 shows both the effective beam temperature and Mach number for the best fit. The data indicate that very small changes in the position of the discharge near the nozzle produce large changes in the effective beam temperature. At the shortest distance, the discharge could be seen protruding through the nozzle into the low pressure downstream side of the nozzle. At still shorter distances, the discharge was extinguished due to operation in a low pressure region of the nozzle and the inability to increase the laser power.

Fig. 6 shows the TOF distributions of Xe, O<sub>2</sub>, and

O-atoms obtained from a mixture of 4% oxygen in xenon. These data were taken under conditions in which the discharge was accidentally offset slightly from the original nozzle hole thus causing erosion of the nozzle. In order to operate the discharge with a higher percentage of oxygen in argon a more powerful laser was used and a platinum nozzle was developed capable of withstanding laser powers up to 1.5 kW and high concentrations of oxygen. The nozzle was tested with a mixture of 50% oxygen in argon at a laser power of 500 W. The more powerful laser has not been coupled to the molecular beam apparatus shown in fig. 4 so TOF analyses have not yet been performed but exposure of organic based films to the beam indicated a high percentage of O-atoms present by the extent of surface etching. In addition, extensive ozone formation detected when the nozzle was vented into the room air indicates a high percentage of  $O_2$  dissociation.

Fig. 7 presents preliminary results of xenon scattering from a very rough stainless steel surface. The translational energy of the direct beam was set at 5752 K and it had an intensity of  $10^{15}$  /s cm<sup>2</sup>. The specular scattered signal was averaged for 6 min and showed a 43% loss of energy suggesting the well depth of the gas-surface interaction was much less than the beam energy and that only a small fraction of the molecules were equilibrated to the surface temperature (300 K). Future work will involve positioning the beam source a factor of 10 closer to the surface producing a 100-fold increase in signal.

### 4. Conclusion

The results using xenon as the carrier gas show that neutral beam translational temperatures (8–9000 K) near the spectroscopic temperature (9–10 000 K) can be obtained with Mach numbers ranging from 4–6. The results using xenon-oxygen and argon-oxygen mixtures indicate that diatomic species can be dissociated and the atomic species extracted into a beam. Estimates of the absolute intensity of the xenon beam show that  $10^{19-20}$  particles/s sr can be produced. Assuming that conduction is a minor pathway for discharge energy loss, an upper limit on the velocity  $V_m$  of another carrier gas of mass *m* at temperature  $T_m$  can be obtained from eq. (2)

$$V_m = V(132/m)^{1/2} (T_m/T)^{1/2} , \qquad (2)$$

where V, T, and 132 are the velocity, temperature and atomic mass in the xenon plasma. Using eq. (2) we obtain 3.6 km/s for argon, 6 km/s for neon, and 14.8 km/s for helium. 1-2% xenon seeded into a helium discharge at 30 000 K would have a beam energy of >86 eV. One area of investigation that is underway is to determine the extent of metastable atom formation. In



Fig. 3. Xenon time of flight (TOF) spectra. Shows TOF spectra at various distances between the discharge and nozzle. TOF (f) was taken with 200 Torr of xenon, the discharge and laser off, and a 300 K nozzle temperature. TOF (c) was obtained with the discharge off, the laser on at 60 W and the lens focal volume positioned at the electrodes. All other TOFs were obtained with the discharge operating in 3 atm of xenon with decreasing discharge to nozzle distance in the order (d) to (a). Experimental flight times are not corrected for timing mark offset and ion flight times through the mass spectrometer.



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from the rotatable lid of the main vacuum chamber for the measurement of angular distributions. Also shown is a time-of-flight chopping wheel which provides 50% transmission efficiency using cross correlation techniques. Pumping of the system is accomplished by a 1500 1/s turbo on the scattering chamber, a 500 1/s turbo on the differential pumping chambers, and ion pumps on the detector along with a liquid helium cryopump.

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Fig. 5. Beam temperature and Mach number as a function of discharge to nozzle distance: Beam temperature and Mach number are determined by a least squares fit of eq. (1) (see text) to the TOF data of fig. 3. These two quantities are plotted versus the distance between the nozzle and discharge.



Fig. 6. TOF data for Xe,  $O_2$ , and O-atoms. Taken with 4% oxygen in the discharge. Discharge was found to be displaced from the nozzle axis by 3 nozzle diameters (0.3 mm).

the case of oxygen, <sup>1</sup>D O-atom formation will probably be suppressed through collisional deactivation [3a].



Fig. 7. Xenon surface scattering. Direct beam intensity at the surface with the nozzle one meter away was estimated to be  $10^{15}$  /s-cm<sup>2</sup>. Surface was stainless steel with a great deal of visible roughness which produced a very broad angular distribution. Note that 43% of the translational energy has been lost at the specular angle from the beam and it has not equilibrated to the surface temperature of 300 K. The scattered signal was averaged for 6 min. Future work will place the source 10–15 cm from the surface thus increasing the signals by about 100.

Future work will be performed using a  $1.5 \text{ kW cw-CO}_2$  laser coupled to the beam apparatus in order to verify the scaling equation and to characterize in detail the production of metastable atoms and molecules.

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- [4] D.A. Cremers, F.L. Archuleta and R.J. Martinez, Spectrochimica Acta 40B (1985) 665.
- [5] The plasma frequency  $f_p = [(e^2 n_e)/(4\pi^2 \varepsilon_0 m_e)]^{1/2}$ , where  $e n_e$ , and  $m_e$  are the electronic charge, density, and mass, respectively, and  $\varepsilon_0$  is the vacuum permittivity. Electromagnetic waves of frequency  $f_w$  interact directly with the plasma to produce heating when  $f_w < f_p$ , but when  $f_w > f_p$  heating occurs by interaction between the waves and individual electrons and ions. For an electron density of  $8 \times 10^{16}$  cm<sup>-3</sup>,  $f_p = 2.5$  THz which is a factor of 10 below the 30 THz laser frequency.