Short-pulse laser interferometric measurement of absolute gas densities from a cooled gas jet

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We report on the use of a novel technique to measure the gas density from a pulsed gas jet. Deuterium gas is fully ionized with an intense picosecond laser, and the resulting electron density is measured by interferometric probing with a second picosecond pulse. We have applied this technique to characterize a cryogenically cooled, high-density gas jet.

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Pulsed gas jets are currently used in a wide range of physics experiments, particularly in many laser plasma\(^1\) and high-field nonlinear optical experiments such as high-order harmonic generation.\(^2\) Such gas jets are also routinely employed to produce molecular beams of van der Waals bonded clusters.\(^3\) For this application the gas jet is often cooled to enhance the cluster formation.\(^3\) Recently pulsed gas jets were investigated as a debris-free laser target to produce x-rays for lithography applications.\(^4\)

Such experiments usually require knowledge of the gas density immediately below the output of the jet nozzle. These measurements are difficult to make, and a number of techniques have been devised in recent years to characterize the density profiles of such jets.\(^5\)–\(^10\) Most techniques rely on optical methods,\(^6\)–\(^8,10\) although a density measurement technique based on nuclear Rutherford backscattering has been reported as well.\(^9\) Many characterization methods require a comparison of some quantity such as laser-induced plasma fluorescence\(^6\) or third-harmonic generation\(^10\) between the gas jet and a static filled cell of known density. The disadvantage of these techniques is that differences in the density profiles of the gas jet and the static cell can lead to substantial errors in the comparison of signals because of laser propagation effects. The scattering technique of Ref. 9 does not have this problem; however, an accurate determination of the absolute gas density with this approach is difficult. Interferometric techniques are superior because they yield a direct determination of absolute gas densities without the need for comparison with reference gas cell. Altucci et al. previously conducted interferometric measurements directly on the gas jet.\(^10\) Their method is difficult, however, because the refractive index of the gas is close to 1. If the gas is first ionized, on the other hand, this problem can be averted because of the large deviation of the plasma’s refractive index from unity. In this Letter we report the measurement of the absolute gas density from a cryogenically cooled deuterium gas jet by a novel technique based on picosecond ionization and interferometry.

Our density measurements use an intense picosecond laser to ionize the gas in the gas jet. The resulting plasma is then probed interferometrically with a second picosecond probe pulse timed within 20 ps of the initial ionizing pulse, permitting an accurate determination of the electron density in the plasma. Using a low-Z gas such as hydrogen or helium that can be ionized to a known charge state permits a direct determination of the gas density from the electron density. Use of picosecond pulses with this technique is advantageous compared with the use of long, nanosecond pulses because the short pulse effectively freezes the motion of the plasma. Because plasma produced by a laser in the gas will expand on a nanosecond time scale and thereby rarely, an accurate determination of the initial density is difficult. The use of a short probe pulse timed to within a few picoseconds of the ionizing pulse circumvents this problem by probing the plasma before it can expand or recombine.

In our experiments we measure the densities from a modified commercial solenoid valve sonic gas jet. This jet has been optimized for cryogenic cooling and operation with high backing pressure in high vacuum systems. Our jet is used primarily for atomic cluster generation and plasma physics experiments. The jet has a 500-\(\mu\)m-diameter nozzle and is operated with an opening time of 3 ms, ensuring that the gas flow is essentially steady state during the gas pulse. The density is probed 1.5 ms after the valve has been opened, although variation between the valve opening and the laser pulse could be used to measure the time variation of the gas plume as well. For all the experiments reported here, deuterium is used to back the gas jet. This gas is used because it permits complete ionization with modest laser intensity and is a species utilized in many of our plasma experiments. This interferometric technique is, in principle, quite accurate (better than 10\%); however, because of shot-to-shot fluctuations in the gas jet itself at a given backing pressure and reservoir temperature, some uncertainty about the measured gas density arises.

To conduct the measurements we use a 2-ps pulse (full width at half-maximum) with a wavelength of 526 nm from a frequency-doubled Nd:glass laser. 25 mJ of energy from this laser is focused into the gas jet to a peak intensity of \(1 \times 10^{16}\) W/cm\(^2\), well above the intensity required for complete saturation of the ionization of the D\(_2\) molecules in the gas jet. Thus the measured electron density is simply a factor of 2 greater than the initial molecular density. The
position of the laser focus below the jet nozzle can be varied to probe the gas density as a function of distance from the jet nozzle. A small amount of the initial 526-nm light is split from the main beam, sent through a delay leg, Raman shifted in an ethanol cell to 620 nm, and propagated through the vacuum chamber at a right angle to the main, focused beam. This probe backlights the plasma and is imaged by a telescope with 20× magnification to a CCD camera. The probe beam traverses the plasma 15 ps after the main pulse has ionized the gas. After the imaging telescope, before the CCD camera, the probe beam is sent into a Michelson interferometer with a right-angle prism in one leg. With this configuration it is possible to interfere the part of the beam phase shifted by the plasma with a part of the beam that passed underneath the plasma.

A typical interferogram from this setup is shown in Fig. 1. To derive the plasma electron density we determine the phase from the image by using fast-Fourier-transform techniques, and we Abel invert the cylindrically symmetric phase image to retrieve the electron density in the center of the plasma channel. The electron density radial profile from the center of this image is also plotted in Fig. 1. The radial profile of this plasma channel is approximately flat-topped, confirming that the ionization is completely saturated in the D$_2$ gas. Using a wider field of view than that used in our experiments would permit a determination of the gas density as a function of radius in the gas plume as well. This information can be simply derived by an Abel inversion of the electron density at different points along laser propagation axis.

The measured density of the gas jet 1.0 mm below the nozzle as a function of backing pressure is shown in Fig. 2. This figure shows the density scaling of the gas jet with backing pressure of 10–65 bars for two gas reservoir temperatures, 290 and 105 K. From these data we see that the gas jet density scales linearly with increasing backing pressure for both temperatures. (Other reservoir temperatures of 290–105 K were also investigated and yielded similar linear behavior.) This linear scaling continues with backing pressure up to 65 bars, near the maximum operating pressure of this gas jet. The gas density of the cooled jet is substantially higher (2.4 times) than the gas density of the jet at room temperature. This scaling is consistent with an ideal gas treatment of the jet expansion, where $n(T_1)/n(T_2) = T_2/T_1$.

These results are of interest because they confirm that, for experiments that require high density, it is possible to cool the gas reservoir to increase the gas density, even when the jet is used near is maximum operating pressure. We also note that the densities observed in this jet are quite high, with an electron density of nearly 10% of the critical density for a 1-μm-wavelength laser pulse. Thus, by using atomic species with a greater number of electrons than hydrogen molecules, it is possible to create plasmas with density above the critical density with such a cooled jet.

We have also examined the gas density as a function of position below the gas jet nozzle. These measurements are shown in Fig. 3. Here the peak gas den-

![Laser In](image1)

![Laser Out](image2)

**Fig. 1.** Typical interferogram in a D$_2$ gas jet 1.0 mm below the gas jet nozzle. The gas jet has been cooled to a temperature of 105 K and backed with a pressure of 55 bars. The plot is the deconvolved electron density as a function of radius in the center of the image.

![Graph](image3)

**Fig. 2.** Measured gas density 1.0 mm below the gas jet nozzle as a function of backing pressure for two gas reservoir temperatures. The solid lines are linear fits to the data.
Fig. 3. Measured gas density as a function of position below the gas jet nozzle when the jet is backed with a pressure of 60 bars at two different reservoir temperatures. The solid curves are fits of Eq. (1) to the data.

distance from the nozzle is similar at both temperatures, with the lower temperature exhibiting higher density over the entire distance range studied.

We find that the scaling of the density with $y$, the distance below the gas nozzle orifice, is consistent with the expected scaling of $1/y^2$ far from the nozzle for a conically expanding gas. Near the nozzle the jet plume will be nearly collimated, and the density will be constant over this distance, a trend manifested in the data. It can be shown that the gas density on the axis of a sonic expansion, $n(y)$, will vary as

$$n(y) = \frac{n_0}{1 + \alpha y^2},$$

(1)

where $n_0$ is the gas density near the nozzle and $\alpha$ is parameter that depends on the geometry and backing pressure of the jet. We conducted a least-squares fitting of Eq. (1), using $n_0$ and $\alpha$ as fitting parameters for our data at both temperatures. These fits are shown in Fig. 3 and indicate that our gas jet at least approximately follows this scaling for both reservoir temperatures.

In conclusion, we have applied a picosecond resolved interferometric technique to the characterization of a cryogenically cooled high-pressure pulsed gas jet. We have shown that by use of picosecond laser pulse ionization of the constituent gas it is possible to determine accurately the initial gas density by measuring the electron density within a few picoseconds after the ionization and thereby avoid any plasma expansion or recombination. Using this technique, we found that a pulsed gas jet modified for low-temperature high-pressure operation can produce gas densities of nearly $5 \times 10^{19}$ atoms/cm$^3$ near the jet nozzle. This gas jet exhibits a linear gas density dependence over a wide backing pressure range. We also confirm that the gas density increases inversely with the gas reservoir temperature. Finally, we found that the density on the gas plume axis follows the $1/y^2$ scaling expected for a simple geometric gas expansion.

References