

Characterization of a cryogenically cooled high-pressure gas jet for laser/cluster interaction experiments

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We have developed and carried out detailed characterization of a cryogenically cooled (34–300 K), high-pressure (55 kTorr) solenoid driven pulsed valve that has been used to produce dense jets of atomic clusters for high intensity laser interaction studies. Measurements including Rayleigh scattering and short pulse interferometry show that clusters of controlled size, from a few to $>10^4$ atoms/cluster can be produced from a broad range of light and heavy gases, at average atomic densities up to 4×10^{19} atoms/cc. Continuous temperature and pressure control of the valve allows us to vary mean cluster size while keeping the average atomic density constant, and we find that many aspects of the valves behavior are consistent with ideal gas laws. However, we also show that effects including the build up of flow on milliseconds time scales, the cooling of gas flowing into the valve, and condensation of gas inside the valve body at temperatures well above the liquefaction point need to be carefully characterized in order to decouple the operation of the jet from the laser interaction physics. © 1998 American Institute of Physics. [S0034-6748(98)01011-9]

I. INTRODUCTION

Atomic clusters from sources including gas jets¹ and effusive ovens² have been studied for many years using techniques including infrared spectroscopy, low power optical probing, microwave ionization, and electron attachment. Clusters have been of particular interest in chemistry as they enable powerful techniques used in the investigation of molecular dynamics and collective effects in low temperature systems. More recently the interaction of clusters with picosecond and femtosecond lasers producing intensities $>10^{15}$ W cm⁻² has attracted considerable interest.^{3–14} This is due to the very significant changes that occur in the interaction physics when moving from monatomic gases to clusters of even a few tens of atoms^{4,11} and is in part a consequence of the high local density within the cluster greatly enhancing coupling of laser energy into both ions and electrons. In a monatomic gas heated by an intense subpicosecond laser pulse at 10^{15} – 10^{18} W cm⁻² ion temperatures of 10–20 eV and coupling efficiencies $\ll 1\%$ are quite typical. In stark contrast, the interaction of such a laser with a suitable cluster medium generates extremely hot plasmas with mean ion temperatures in the 10–50 keV range^{3,5} and peak ion temperatures of 1 MeV.³ Due to the extremely efficient ($>90\%$) absorption of intense laser light in cluster media,⁶ such experiments can now be carried out with table top laser systems.

Hot ion plasmas produced using clusters are of great interest for studies in areas including astrophysics, high energy density plasma physics,^{6,7,10} lithographic x-ray sources, and thermonuclear fusion. For these experiments and applications, a well-characterized source that produces clustered

gases of carefully controlled size, composition, and high mean density is extremely important. In addition, a broad range of valve and jet parameters including the absolute density, density profile within a few mm of the nozzle and cluster formation as a function of time, pressure, and temperature all need to be well understood if the laser/cluster interaction physics is to be properly decoupled from the operation of the jet. There is an extensive body of literature on sonic and supersonic flows through nozzles, but in practice it is extremely difficult to perform a detailed and accurate first principles calculation for a given valve, and no rigorous quantitative theories of cluster formation currently exist. For this reason, an experimental determination of a particular gas jets behavior is generally more useful. In addition, most previous work on diagnosing cluster jets has concentrated on aspects such as the beam intensity at long distances from the nozzle that are of little relevance in most current laser interaction experiments.

In this article, we report on the development and detailed characterization of a high backing pressure (>55 kTorr) pulsed solenoid valve source of van der Waals or hydrogen bonded clusters that we have used extensively in a broad range of cluster interaction^{3–10,12} and high harmonic generation^{15,16} experiments. The valve can be continuously temperature controlled between 34 and 300 K, allowing both size selection of clusters, and clustering of light gases such as H₂, D₂, and He which interact weakly at room temperature. Devices of this type have been described previously for producing molecular and cluster beams^{17,18} and for injecting fuel into fusion reactors¹⁹ and a good review of their operation is provided by.²⁰ Here we describe very high-pressure, continuous temperature-controlled operation of such a valve along with a detailed characterization of the cluster medium produced, using force transducers, time-resolved Rayleigh

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TABLE I. Hagen parameter k values for a range of gases from Ref. 23 and calculated from Ref. 25.

Gas	H ₂	D ₂	N ₂	O ₂	CO ₂	CH ₄	He	Ne	Ar	Kr	Xe
k	184 ^a	181 ^a	528 ^a	1400 ^a	3660 ^a	2360 ^b	3.85 ^b	185 ^b	1650 ^b	2890 ^b	5500 ^b

^aReference 25.^bReference 23.

scattering, and short pulse interferometry. We have concentrated on the valve and jet parameters of particular importance in laser/cluster interaction studies. For example, a complete understanding of the pressure scaling of absolute jet density in the tightly bounded flow region within ≈ 2 mm of the nozzle is extremely important in a nonlinear interaction such as high harmonic generation.^{9,15,16} We detail a device that we have used extensively,^{3-10,12,15,16} highlight some of the unexpected behavior exhibited by the valve, and set out the broad parameter range accessible with a relatively simple system as a guideline to others entering this exciting field.

We show that the valve allows broad size selection of clusters of many gases through control of the preexpansion pressure P_0 and temperature T_0 . By holding $P_0 T_0^{-1/2}$ constant when changing cluster size, the mean atomic density can also be held constant. This allows detailed interaction studies as a function of mean cluster size to be made without changing other density dependent effects such as the growth of plasma instabilities. However, we have not attempted to investigate the cluster size distribution due to the extreme difficulty of this measurement^{20,21} and because Rayleigh scattering diagnostics and the high intensity laser cluster interactions we study are typically dominated by larger clusters.

II. CLUSTER PRODUCTION IN GAS JETS

Expansion of gas through a nozzle into vacuum results in substantial cooling in the frame of the moving gas, and atoms or molecules that interact weakly at room temperature can form clusters as a result. Attractive forces between atoms can be due to van der Waals or hydrogen bonding, and the clustering effect is primarily determined by the temperature and pressure of the gas reservoir, shape and size of the nozzle, and strength of the interatomic bonds formed.^{22,23} As is the case with many complex fluid dynamics problems, there is currently no rigorous theory to predict cluster formation in a free jet expansion. However, the onset of clustering, and size of clusters produced can be described by an empirical scaling parameter Γ^* referred to as the Hagen parameter^{23,24}

$$\Gamma^* = k \frac{(d/\tan \alpha)^{0.85}}{T_0^{2.29}} P_0, \quad (1)$$

where d is the nozzle diameter (mm), α the expansion half angle ($\alpha = 45^\circ$ for sonic nozzles, $\alpha < 45^\circ$ for supersonic), P_0 the backing pressure (mbar), T_0 the pre-expansion temperature (Kelvin), and k a constant related to bond formation (see Table I).

Clustering generally begins for $\Gamma^* > 100-300$, with the number of atoms per cluster N_c scaling as $N_c \propto \Gamma^{*2.0-2.5}$.^{22,26} We can thus use the strong variation of N_c with T_0 and P_0 to

engineer a medium of arbitrary mean density and N_c , for interaction experiments. However, this requires a detailed knowledge of the valve and cluster jet behavior over a broad range of conditions. Fortunately, this is somewhat simplified by the fact that the model of corresponding jets²⁰ allows the measured properties of a jet for one gas to be usefully extrapolated to other gases.

III. THE CRYOGENIC JET

A General Valve Corporation series 99 solenoid driven pulsed gas valve with a 0.3 mm diameter cylindrical aperture was used as a basis for this work. General valves drive electronics were used but the valve was modified in several ways. The valve was surrounded by a close fitting 5 mm copper jacket wrapped with 2 mm bore copper tube (Fig. 1) carrying a coolant, either nitrogen gas forced under pressure through a separate liquid N₂ cooled heat exchanger or liquid helium. The jacket temperature was monitored with a type K thermocouple corrected for nonlinearity below 100 K and a cooled heat shield surrounded the valve to limit radiative heating when operating below 80 K. An important advantage of our cooling method compared to use of a simple reservoir attached directly to the valve¹⁹ is that by varying the coolant flow rate, we can control the valves temperature to within a few degrees over the full range 34–300 K, rather than having to work at a few discrete temperatures set by the boiling points of suitable cryogenic fluids. This is critical for detailed investigations of effects such as hot ion production as a function of cluster size where the extremely strong scaling

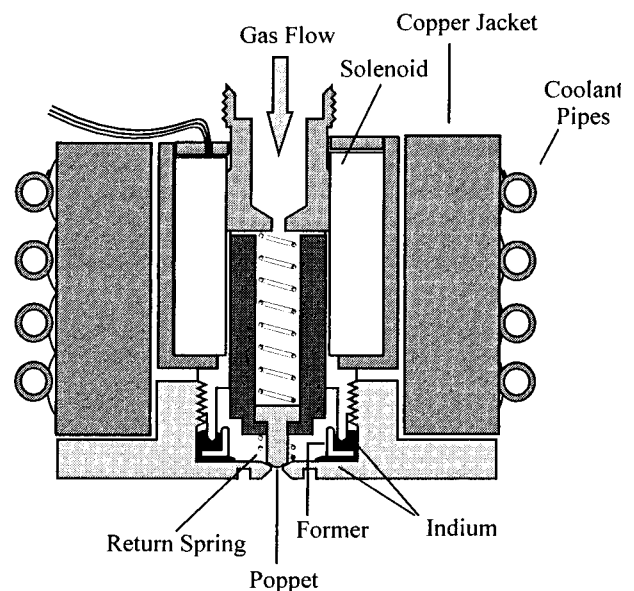


FIG. 1. Section through the cooled valve showing details of the indium seal, poppet, and solenoid assembly.

of N_c with temperature ($N_c \propto T_0^{-5}$) would otherwise result in large discontinuous jumps in mean cluster size.

For cryogenic operation, the valve was originally fitted with a copper gasket seal. However, its performance depends critically on how the valve nozzle is fixed to the body. This is extremely difficult to optimize with a hard copper gasket, and instead we used a deformable indium seal, with a stainless-steel former to prevent indium being pushed into the poppet assembly. This allows us to pressurize and fire the valve while adjusting the nozzle, and we note that the noise made by the valve during this process is an extremely useful diagnostic of its operation. Under or overtightening of the nozzle by $\approx 10^\circ$ seriously degrades its performance, for example, causing the valve to alternate between full and partial opening on successive shots.

The valve can operate at pressures >55 kTorr cooled to ≈ 70 K without substantial leakage, and has been used on systems maintained at $<2 \times 10^{-5}$ Torr background pressure. This is particularly important in fast ion and electron measurements³⁻⁵ where signal from a monatomic background must be minimized. In both experiments and characterization studies, we operate the valve in vacuum (10^{-1} – 10^{-5} Torr) such that interactions between the jet and background gas are negligible. When operating at low temperatures, care needs to be taken in purging the valve and gas lines before cooling as ice crystals can block the nozzle or prevent full closure after firing. Below 70 K it is increasingly difficult to maintain a perfect seal, however, the valve itself still performs well.

IV. FLOW CHARACTERIZATION AS A FUNCTION OF BACKING PRESSURE

Processes such as high harmonic generation are critically dependent on the density and length of the medium in which they occur,^{9,15,16} as are optical scattering diagnostics of cluster size. For this reason, a good understanding of how the jet density scales with backing pressure close to the nozzle is important, and was investigated in several ways.

A small piezoelectric transducer that produced a signal proportional to impulse was placed below the nozzle to quantify gas flow as a function of backing pressure and temperature. (This elegant device also makes a useful real-time diagnostic during experiments as it is simple and noninvasive, but gives an immediate indication of a valve failure due, for example, to poppet damage.) Adiabatic expansion through a nozzle typically produces a cluster beam with uniform axial velocity²² set by the free expansion velocity $[\gamma/(\gamma-1)]^{1/2}(k_b T_0/m)^{1/2}$ with γ the ratio of specific heats, k_b Boltzmann's constant, and m the atomic mass. Impulse to the force transducer is thus proportional to the number of gas atoms that hit it for constant T_0 and transverse jet profile.

We find an extremely linear variation of transducer signal as a function of backing pressure P_0 from 0 to 50 kTorr (Fig. 2) showing that for a fixed opening time the volume of gas ejected is constant and that as expected from ideal gas behavior the number of atoms released $N \propto P_0$. This was confirmed by measuring the number of shots required to give

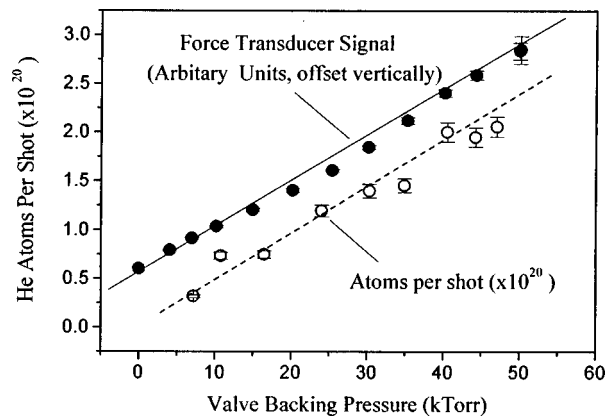


FIG. 2. Number of atoms per shot released (○) for a 40 kTorr He fill at 293 K and a $600 \mu\text{s}$ opening time and (offset vertically for clarity) force transducer signal (●) showing linear behavior.

a set jump in pressure in the closed vacuum chamber containing the valve and from this we can also calculate the number of atoms released per shot (Fig. 2).

V. FLOW CHARACTERIZATION AS A FUNCTION OF VALVE TEMPERATURE

Cooling provides a powerful method of varying mean cluster size ($N_c \propto T_0^{-5}$), but also effects the density and transverse profile of the jet, and at low temperatures the valve can exhibit unusual behavior including large scale droplet formation. Characterizations of cluster jets as a function of temperature previously reported have typically been limited to a few points set by direct use of a cryogenic coolant and consequently there are few detailed temperature scaling studies in the literature. However, our indirect cooling method allows continuous temperature variation studies to be carried out with relative ease.

Ideal gas behavior should give a preexpansion density inside the valve $\propto T_0^{-1}$, an axial jet velocity $\propto T_0^{1/2}$ and the number of atoms released per shot should thus scale as $N \propto T_0^{-1/2}$. For a constant jet profile, the average atomic density in the jet should then scale as $n_0 \propto T_0^{-1}$. In practice, we see significant departures from this behavior. Figure 3 shows the

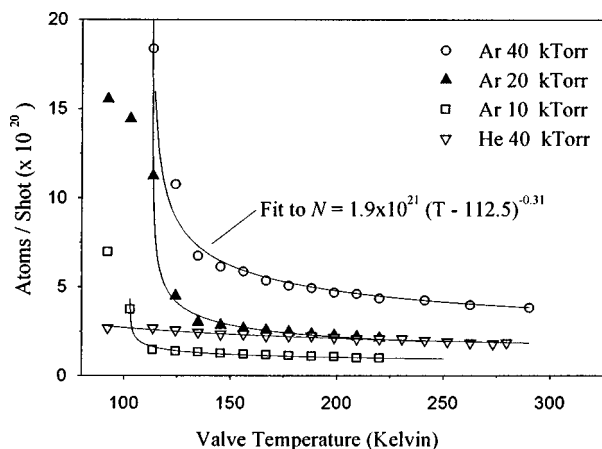


FIG. 3. Number of atoms per shot N released by the jet as a function of temperature for Ar at 40, 20, and 10 kTorr and He at 40 kTorr with fits to $N \propto (T_0 - T^*)^{-\beta}$.

number of atoms released for a range of conditions with the valve fired at 0.1 Hz to ensure complete cooling between shots. We can fit these plots to an empirical temperature scaling of the form $N \propto (T_0 - T^*)^{-\beta}$, where T^* is a temperature slightly greater than the liquefaction point ($T^* \approx 112$ K for Ar) and β a parameter that ranges between 0.2 and 0.4 (higher for higher backing pressures). We also find that the force transducer signal varies as $\approx T_0^{0.17}$ which is consistent with an ideal gas scaling of axial velocity $\propto T^{0.5}$ and the observed $N \propto T_0^{-0.3}$ scaling. These results imply a density scaling of the medium directly below the nozzle of $n_0 \propto T_0^{-0.8}$ which is broadly consistent with absolute electron density measurements detailed in Sec. IX where we see a scaling $n_0 \propto T_0^{-0.5}$.

In Ar there is a rapid rise in the number of atoms released by the valve well before the liquefaction point (87 K), which coincides with significant changes in the nature of the medium produced. The sudden pressure drop in the valve body during firing causes rapid condensation of liquid Ar inside the valve, which is then expelled as a spray of ≈ 1 μm diameter droplets rather than a clustering gas stream. At this point, the jet is actually visible by eye under vacuum, hence our assumption of the micron size distribution. The point above the liquefaction temperature at which this effect occurs for Ar is pressure dependent, as shown in Fig. 3 and as expected we do not see a similar effect in He with its much lower boiling point. We have not investigated this effect further, but highlight it here as sudden changes in mass flow rate and cluster size of this type are not predicted by ideal gas behavior or Hagen's scaling laws and will have a significant impact on an interaction experiment. The interaction length for a laser focus in the jet remains constant as a function of temperature, but the density-length product changes in a way not predicted by assuming ideal gas behavior and a scaling $n_0 \propto T_0^{-0.5}$ rather than $n_0 \propto T_0^{-1}$ typically best describes the average atomic density of the jet.

VI. TIME VARIATION OF CLUSTER PRODUCTION

Movement of the poppet and the subsequent build up of flow through the nozzle when the valve is fired both limit cluster formation. For room temperature operation, cluster production can eventually reach steady state, but at low temperatures heat input from the solenoid limits clustering. As a result there is a significant time during which the scaling relationship for N_c derived from Γ^* is invalid, and a clear optimum time for maximum clustering at low temperatures. Care thus needs to be taken in setting the relative timing between firing the jet and the arrival of the laser pulse in interaction experiments.

We examined these effects by time resolving Rayleigh scattering from clusters^{19,27} formed in the jet. A 526 nm cw probe beam was focused 2 mm below the nozzle using an F/20 optic, and light scattered at 90° to the probe axis collected with an F/3 lens and focused into a photomultiplier outside the vacuum chamber. Total scattered signal S_{RS} is proportional to the product of the scattering cross section σ and cluster number density n_c , with the classical Rayleigh cross section²⁸ given by

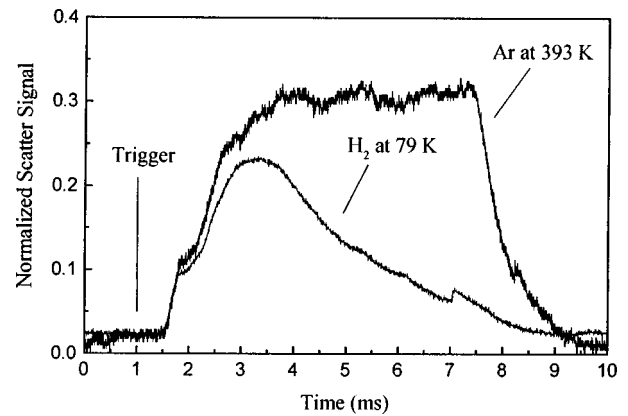


FIG. 4. Time-resolved 90° scattered light signal for 50 kTorr Ar at 293 K, and H₂ at 79 K (scaled to similar peak values for clarity) for 6 ms valve opening times, showing the build up and decay of cluster formation.

$$\sigma = \frac{8\pi r^6}{3\lambda^4} \left(\frac{n^2 - 1}{n^2 + 2} \right)^2, \quad (2)$$

with r the cluster radius, λ the optical wavelength, and n the refractive index of the scattering medium.

Figure 4 shows the time history of the scatter signal from 50 kTorr fills of Ar at 293 K and H₂ at 79 K for a long (6 ms) valve opening time. The two scatter signals have been scaled to give similar peak values for clarity. The valve was triggered at $t = 1.0$ ms and the scattered light signal (and thus both gas flow and cluster formation) are seen to build up over ≈ 2.5 ms. The signal from Ar then reaches steady state, but in contrast the H₂ signal falls off in time (D₂ behaves similarly as one might expect). As the valve closes there is a more rapid turn off of the scattering signal and we see occasional spikes in the scattered light amplitude that we attribute to gas momentarily forced out of the nozzle by the closing poppet. For both Ar and H₂, changing the backing pressure alters the peak scattering signal as expected, but not the overall time history.

After ≈ 3 ms the preexpansion volume of gas ejected is comparable to that of the valve body, and we might expect that warm gas flowing into the valve begins to decrease the mean cluster size, and hence the scattering signal. However, cooling the pipe-work supplying the jet to 79 K does not change the fall off in cluster signal with time for H₂ or D₂. The mechanism for this “self limiting” of clustering seems to be ohmic heating of the solenoid that results in a significant temperature rise inside the valve during cryogenic operation. There is thus an optimum time after firing the jet for production of the largest D₂ and H₂ clusters of interest for thermonuclear fusion studies, and care need to be taken in establishing this time on any given cooled pulsed valve system.

VII. GAS JET REPETITION RATE

High intensity laser systems operating at 10 Hz are relatively common, and so an important parameter is the maximum frequency a device of this type can be run at before clustering is adversely effected by ohmic heating by the solenoid or a lack of time for complete cooling of gas entering

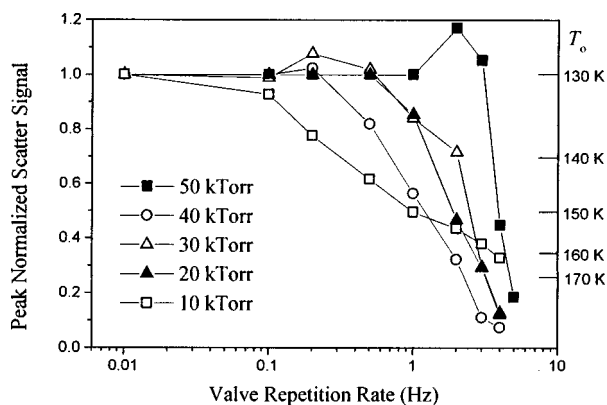


FIG. 5. Peak 90° scatter signal (normalized to 1 for low repetition rates) for Ar at 10, 20, 30, 40, and 50 kTorr at a temperature of 130 K as a function of jet repetition rate, showing a fall off in cluster signal for both higher repetition rates and lower backing pressures. Right axis gives estimated T_0 from a scaling $N_c \propto T_0^{-5}$.

the valve. This is also an issue in proposed applications of laser heated clusters including low debris x-ray sources for lithography, in which valve repetition rate may limit the available time-averaged x-ray power.

This was investigated by measuring scatter signal from the cluster medium as a function of jet repetition rate. Figure 5 shows peak scattered signal from Ar at 130 K for several backing pressures, normalized to unity for low (0.01 Hz) repetition rates. At >0.1 Hz the scatter signal begins to fall off with increasing repetition rate and we can use the scaling of N_c with T_0 to estimate the temperature inside the valve (right axis of Fig. 5). It is interesting to note that this is less of a problem for higher backing pressures, even though the thermal conductivity of a gas is typically constant in pressure (the conductivity of Ar actually increases by $\approx 5\%$ from 10 to 50 kTorr). For higher pressures, a larger mass of gas flows into the valve between shots, and it seems likely that the precooling of gas in the pipe work above the valve results in a slightly warmer valve body having less effect on the gas reservoir temperature reached before firing. The cooled jet can thus be run at higher frequencies when operating at high pressure without adversely affecting the preexpansion temperature of the gas. However, it is clear that continuous low temperature, high frequency (≈ 10 Hz) operation of devices of this type will require an improved cooling system and the possible addition of insulation within the valve to isolate the solenoid from the gas reservoir.

VIII. CLUSTER SIZE DETERMINATION

Changes of several orders of magnitude occur in the peak ion energy and absorption efficiency of laser light in a cluster gas as the cluster size N_c is increased from a few tens to a few thousands of atoms^{3,6} and so a good estimate of N_c is critical in interpreting interaction experiments. While the size of clusters produced in a free jet expansion is difficult to measure directly, it can be estimated using Rayleigh scattering. The threshold for observing optical scattering from a cluster jet typically occurs for $N_c \approx 100$ (Ref. 23) and the pressure scaling of scattering after this point allows us to estimate the average cluster size (provided the mean density

as a function of pressure is also well known). However, it is important to note that this method tells us little about the cluster size distribution.²¹

As the majority of atoms in the jet condense into clusters for $\Gamma^* > 100$, it follows that the cluster number density n_c will be given by the monomer density before clustering, n_0 divided by N_c hence $n_c \approx n_0/N_c$. For spherical clusters, the cluster radius $R_c \propto N_c^{1/3}$ and the scattering signal S_{RS} proportional to $n_0 N_c$. We know that the monomer density before clustering is exactly proportional to backing pressure P_0 , and so have $S_{RS} \approx P_0 N_c$. Finally, the Hagen parameter shows that $N_c \propto P_0^{2-2.5}$ and so the scattered light signal S_{RS} should vary as $P_0^{3-3.5}$.

Experimentally, we find that the scattered signal does typically vary as $P_0^{3-3.5}$ for a wide range of conditions, consistent with published scalings of cluster size with backing pressure. The average cluster size at a particular backing pressure can then be estimated from scattering data of the type shown in Fig. 6. Here we assume the observed onset of optical scattering corresponds to $N_c \approx 100$ (Ref. 23) and use the measured increase in scattered signal to calculate the cluster size as a function of backing pressure.

For H_2 at 79 K and Ar at 293 K, Rayleigh scattering signals scale as $\approx P_0^{3.8}$ and $P_0^{3.6}$, respectively [Figs. 6(a) and 6(b)] giving peak cluster sizes N_c of ≈ 690 and 1160. However, for cooled Ar at 147 K [Fig. 6(c)], we find an initial pressure scaling of $P_0^{3.8}$ below 35 kTorr and a more rapid scaling of $\approx P_0^{4.3}$ at higher pressures. This case approaches the conditions outlined in V where liquid rather than gas is expelled from the valve. It is important to note that even a small number of large droplets can greatly enhance the scatter signal due to the strong size dependence of the Rayleigh cross section. At the break point in the curve at 33 kTorr we estimate a cluster size of ≈ 2900 atoms. If our scaling argument holds at the peak of the curve this implies a cluster size of $\approx 3.5 \times 10^4$ atoms, but it is unclear whether this is actually so. It is, however, clear that in some cases care needs to be taken when estimating large cluster sizes purely from Hagen's scaling laws, and that such estimates should be confirmed using a suitable scattering technique for a given valve if they are to be useful in interpreting interaction experiments.

Production of large clusters ($N_c > 1000$) of light atoms such as D_2 (of interest in fusion related studies) requires cooling well below 80 K. Figure 7 shows the D_2 scatter signal as a function of temperature for liquid He cooling of the jet down to 40 K. From a consideration of Γ^* , the cluster size, number density, and Rayleigh cross section we should expect both N_c and S_{RS} to scale as $T_0^{-4.6-5.7}$. For the D_2 data presented in Fig. 7 we do indeed see a good fit to $S_{RS} \propto T_0^{-5 \pm 0.3}$ and can use this to estimate cluster size by assuming $N_c \approx 100$ when we first see scattered light signal above the noise, giving a peak value of $N_c \approx 2.4 \times 10^4$ for $T_0 = 45$ K.

IX. ABSOLUTE MEASUREMENTS OF CLUSTER GAS DENSITY

Absolute measurements of average atomic density and density profiles were made using a novel short pulse inter-

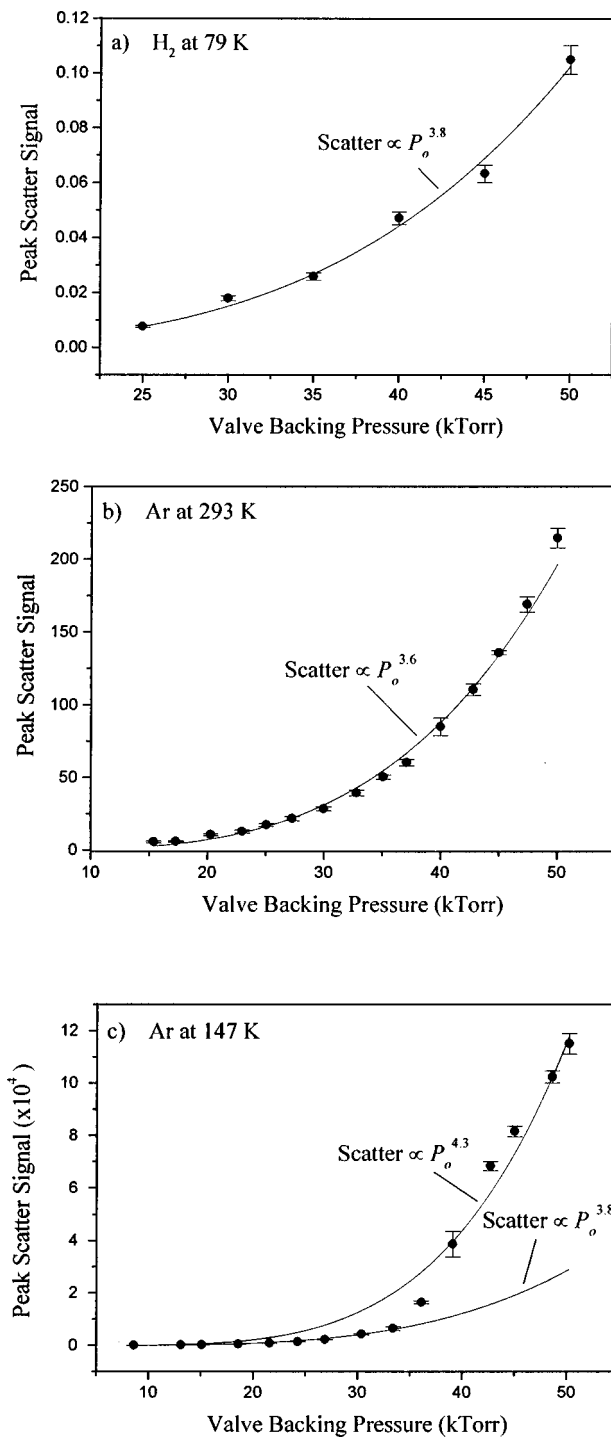


FIG. 6. Peak 90° scattered light signal as a function of pressure for (a) H_2 at 79 K and (b) Ar at 293 K showing the onset of scattering and a near cubic dependence on backing pressure, and (c) Ar at 147 K showing near cubic scaling at lower pressure, and more rapid scaling for higher pressure.

ferometry technique¹² that we have also used for ultrafast electron transport⁷ and channel formation⁸ studies in cluster media. An intense 2 ps heating pulse of 526 nm laser light was focused into a D_2 gas jet at an intensity of $\approx 5 \times 10^{16}$ $W\text{ cm}^{-2}$ causing rapid and complete ionization in the focal volume. A second 2 ps probe pulse at 620 nm illuminated this ionized region at 90° to the heating pulse axis ≈ 10 ps after its arrival. An interferogram of the probe was recorded using a Michelson arrangement, Abel inverted to recover

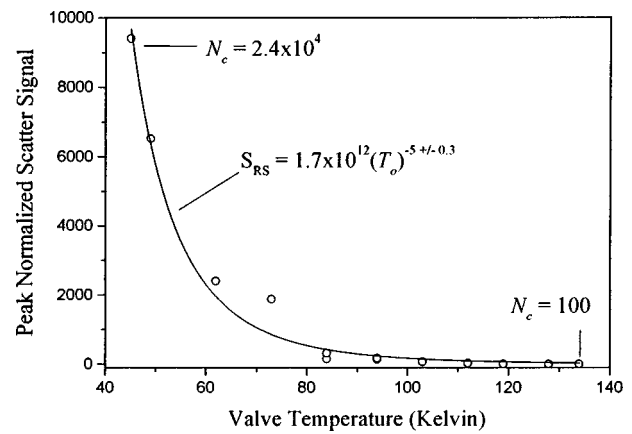


FIG. 7. Peak 90° scattered light signal from D_2 clusters as a function of valve temperature and fit to a T_o^{-5} scaling of initial temperature. Maximum cluster size N_c is $\approx 2.4 \times 10^4$ for $T_o = 40$ K.

phase information, and an electron density profile calculated. The measured electron density for D_2 at 293 and 89 K scales linearly as a function of backing pressure.¹² We know the number of electrons liberated per molecule and can thus obtain the atomic density. Peak atomic densities of 4×10^{19} atoms/cc were found, consistent with those expected from measurements of the number of atoms released by the valve, its opening time, and the axial expansion velocity.

An important advantage of this method is that it gives absolute density without reference to a calibration cell, and due to the short time scales involved, no significant hydrodynamic motion occurs during the measurement. It also removes some uncertainties concerning the refractive index of clustered media in the interferometry of cold cluster jets. Varying the position of the heating beam lets us map the density profile of the jet. Figure 8 shows measured electron density as a function of distance below the nozzle for D_2 at 79 and 293 K which we fit to Gaussian profiles. The $\approx T_o^{0.5}$ scaling between the two plots is accounted for by the additional gas released when the is valve cooled. However, there is little change in the profile of the jet when the valve is cooled, as expected from Sec. V and so the interaction length for an intense laser remains essentially unchanged.

By holding $P_o T_o^{-1/2}$ constant when changing cluster size, the mean atomic density can also held nearly constant. This makes accounting for density dependent effects in the inter-

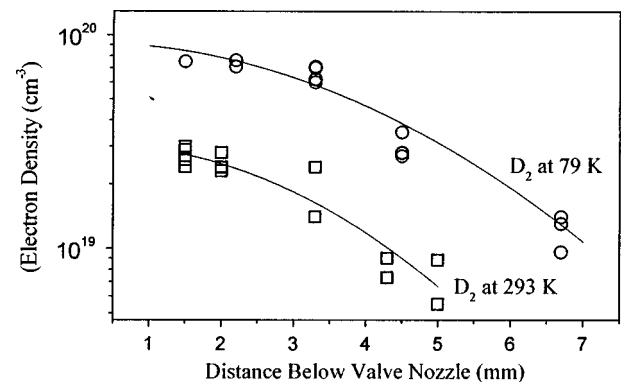


FIG. 8. Measured electron density as a function of distance below gas jet nozzle for D_2 at 79 and 293 K with Gaussian fits to the profile.

action physics simpler, and avoids the unexpected turn on of processes such as stimulated Brillouin scattering which grows nonlinearly with density. However, in experiments where cluster size is varied through temperature control it is often more convenient to work at constant backing pressure and account for the density change in the analysis, provided the interaction physics is sufficiently well understood.

X. DISCUSSION

The interaction of intense lasers with clusters is a new and rapidly expanding field that continues to produce exciting results. However, this work requires well-characterized high density cluster sources, and issues such as the scaling of density and cluster size with initial conditions become increasingly important due to the highly nonlinear nature of the laser/cluster interaction. Hagena's empirical relations, for example, give a very good general guide to cluster formation, but may not be sufficiently exact to properly extract detailed scalings as a function of cluster size from some interaction experiments. We emphasize that a good understanding of the valve and the cluster medium produced is vital if results from cluster interaction experiments are to be interpreted correctly. Pulsed valves of similar designs can produce distinctly different cluster media and we highlight the need for good diagnostics of individual devices and the care that needs to be taken in comparing results from different experiments.

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¹O. F. Hagen, Rev. Sci. Instrum. **63**, 2374 (1992).

²L. Bewig, U. Buck, and C. Mehlmann, Rev. Sci. Instrum. **63**, 3936 (1992).

³T. Ditmire, J. W. G. Tisch, E. Springate, M. B. Mason, N. Hay, R. A. Smith, J. Marangos, and M. H. R. Hutchinson, Nature (London) **386**, 54 (1997).

⁴Y. L. Shao, T. Ditmire, J. W. G. Tisch, E. Springate, J. P. Marangos, and M. H. R. Hutchinson, Phys. Rev. Lett. **77**, 3343 (1996).

⁵T. Ditmire, J. W. G. Tisch, E. Springate, M. B. Mason, N. Hay, J. P. Marangos, and M. H. R. Hutchinson, Phys. Rev. Lett. **78**, 2732 (1997).

⁶T. Ditmire, R. A. Smith, J. W. G. Tisch, and M. H. R. Hutchinson, Phys. Rev. Lett. **78**, 3121 (1997).

⁷T. Ditmire, E. T. Gumbrell, R. A. Smith, A. Djaoui, and M. H. R. Hutchinson, Phys. Rev. Lett. **80**, 720 (1998).

⁸T. Ditmire, R. A. Smith, and M. H. R. Hutchinson, Opt. Lett. **23**, 322 (1998).

⁹J. W. G. Tisch, T. Ditmire, D. J. Fraser, N. Hay, M. B. Mason, E. Springate, J. P. Marangos, and M. H. R. Hutchinson, J. Phys. B **30**, L709 (1997).

¹⁰T. Ditmire, P. K. Patel, R. A. Smith, J. S. Wark, S. J. Rose, D. Mithianaki, R. S. Marjoribanks, and M. H. R. Hutchinson, J. Phys. B **31**, 2825 (1998).

¹¹T. Ditmire, Phys. Rev. A **57**, R4094 (1998).

¹²T. Ditmire and R. A. Smith, Opt. Lett. **23**, 618 (1998).

¹³T. D. Donnelly, T. Ditmire, K. Neuman, M. D. Perry, and R. W. Falcone, Phys. Rev. Lett. **76**, 2472 (1996).

¹⁴A. McPherson, B. D. Thompson, A. B. Borisov, K. Boyer, and C. K. Rhodes, Nature (London) **370**, 631 (1994).

¹⁵J. W. G. Tisch, D. D. Meyerhofer, T. Ditmire, N. Hay, M. B. Mason, and M. H. R. Hutchinson, Phys. Rev. Lett. **80**, 1204 (1998).

¹⁶T. Ditmire, E. T. Gumbrell, R. A. Smith, J. W. G. Tisch, D. D. Meyerhofer, and M. H. R. Hutchinson, Phys. Rev. Lett. **77**, 4756 (1996).

¹⁷O. F. Hagen, Rev. Sci. Instrum. **62**, 2038 (1991).

¹⁸J. P. Bucher, D. C. Douglass, P. Xia, and L. A. Bloomfield, Rev. Sci. Instrum. **61**, 2374 (1990).

¹⁹R. Klingelhöfer and M. H. Moser, J. Appl. Phys. **43**, 4575 (1972).

²⁰O. F. Hagen, in *Molecular Beams and Low Density Gasdynamics*, edited by P. P. Wegener (Marcel Dekker, New York, 1974).

²¹M. Lewerenz, B. Schilling, and J. P. Toennies, Chem. Phys. Lett. **206**, 381 (1993).

²²O. F. Hagen and W. Obert, J. Chem. Phys. **56**, 1793 (1972).

²³J. Wörmer, V. Guzielski, J. Stapelfeldt, and T. Möller, Chem. Phys. Lett. **159**, 321 (1989).

²⁴O. F. Hagen, Z. Phys. D **4**, 291 (1987).

²⁵J. Arno and J. W. Bevan, *Jet Spectroscopy and Molecular Dynamics*, edited by J. M. Hollas and D. Phillips (Blackie, London, 1995).

²⁶J. Farges, M. F. de Feraudy, B. Raoult, and G. Torchet, J. Chem. Phys. **84**, 3491 (1986).

²⁷O. Abraham, S. S. Kim, and G. D. Stein, J. Chem. Phys. **75**, 402 (1981).

²⁸J. D. Jackson, *Classical Electrodynamics*, 2nd ed. (Wiley, New York, 1975).