Experimental investigation on argon cluster sizes for conical nozzles with different opening angles

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Using Rayleigh scattering measurement, we experimentally studied the effect of the opening angle of conical nozzles on the average sizes of argon clusters produced by high-pressure argon gas (up to 50 bars) expanding into vacuum. Both the scattering signal intensity and the scattering image were synchronically recorded by a photomultiplier tube and a charge-coupled device camera. These measurements allow for the comparison of average cluster sizes among conical nozzles of different opening angles. The experimental results indicate that, as expected by Hagena's scaling law, the argon cluster size is dependent on the opening angle. However, it is also found that (1) the cluster size exhibits a larger deviation from Hagena's scaling law at high backing pressure for a nozzle of a smaller opening angle and (2) the smaller the opening angle of conical nozzle gets, the weaker the pressure dependence of cluster size becomes.
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I. INTRODUCTION

In the past decade, the interaction of femtosecond intense laser pulses with gas clusters has attracted lots of research interest, resulting in important findings.[1–4] One of them is deuterium–deuterium nuclear fusion induced by a table-top femtosecond laser pulse.[1] Gas clusters have served as good samples in the laser-cluster interaction since they possess the advantage of both solid target and gas target and exhibit a high absorption of laser pulse energy.[1–4] Usually gas clusters are produced from the adiabatic expansion of a high-pressure gas into vacuum through a nozzle.[5–9] To understand basic phenomena in a laser-cluster interaction, a well-characterized cluster source is needed. Rayleigh scattering measurement has been widely used to estimate the cluster size.[10–18] With Rayleigh scattering measurement only, however, it is difficult to determine the absolute cluster size. For the axisymmetric gas expansion, the average cluster size N_c (the number of atoms per cluster) can be described by Hagena's scaling law $N_c \sim (\Gamma^*)^{2.35}$.[6–9] Hagena's empirical parameter Γ^* is expressed as $Kd_{eq}^{0.85}P_0/T_0^{2.29}$, where K is a constant related to the property of a gas species, and P_0, T_0 are a initial gas backing pressure in millibar and a gas temperature in kelvin before expansion, respectively. Γ^* takes into account all the factors affecting the average cluster size, such as the gas property, the nozzle geometry and the initial gas parameters. Here d_{eq} is a parameter related to the geometry of a nozzle in micrometer. For a sonic nozzle, d_{eq} is equal to its orifice diameter d_s. For a conical nozzle, d_{eq} is the equivalent diameter of the conical nozzle, given by $0.74d/\tan\alpha$ for rare gas, where d is its throat diameter and α the half opening angle. If we define η by the ratio of the average cluster size N_c of argon for a conical nozzle to that for a sonic nozzle under a given P_0 and T_0, η can be written as $(d_{eq}/d_s)^{0.85\times 2.35}$. When the throat diameter, d, of a conical nozzle is equal to the orifice diameter, d_s, of a sonic nozzle, the ratio η becomes equal to $(0.74/\tan\alpha)^{0.85\times 2.35}$. It is obvious from Hagena's scaling law that under the given condition of a gas source, (1) a conical nozzle (if $\tan\alpha < 0.74$) produces η times larger clusters than does a sonic nozzle whose orifice diameter d_s is the same as the throat diameter, d, of the conical nozzle and (2) among conical nozzles with the same d, the nozzle with a smaller opening angle α produces larger clusters since it corresponds to a larger η. It is noted that the Hagena's scaling law is based on the experimental data obtained at relatively low gas backing pressures. It was experimentally demonstrated that the Hagena's scaling law is also applicable to the estimation of argon cluster size even at a high backing pressure in the case of a sonic nozzle.[11,13,16] However, in usual laser-cluster interaction experiments, the high backing pressure with a conical nozzle are quite often employed to produce a high gas density with large clusters.[18] In this case, the inner boundary wall of a conical nozzle would affect the equivalent diameter d_{eq}. It is then necessary to investigate the cluster size at a high backing pressure, for a conical nozzle with a smaller opening angle. Recently, Dorchies *et al.*[18] reported a different scaling law for a conical nozzle using argon gas and found that at a high backing pressure, the cluster size exhibits the weaker dependence on a gas backing pressure than the predictions of Hagena's scaling law.

In this paper, we present the investigation of the argon cluster size for four conical nozzles with different opening angles. A photomultiplier tube (PMT) and a charge-coupled device (CCD) camera were used to detect and image scattered lights, respectively. The spatial profiles of Rayleigh scattering from CCD images were used to estimate the di-

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mension of a scattering region and the area of cross section of gas flow at the scattering region. It is shown that the average cluster size increases as the opening angle of conical nozzle becomes small, as expected by Hagena's scaling law. It is found, however, that for a conical nozzle with a small opening angle, the argon cluster size exhibits the lower power dependence on a backing pressure than, and deviates from that predicted by Hagena's scaling law, especially at high backing pressures. This could result from the fact that the boundary layer in a conical nozzle with a smaller opening angle has stronger effect on the cluster formation at high backing pressures.

II. DETERMINATION OF ARGON CLUSTER SIZE

A Rayleigh scattering signal from clusters formed in the gas jet S_{RS} is proportional to the product of the scattering cross section σ and the number of clusters n_c in a scattering region,[10]

$$S_{RS} \propto n_c \sigma. \quad (1)$$

For a spherical cluster, the average cluster radius $r \propto N_c^{1/3}$ and Rayleigh scattering cross section $\sigma \propto r^6$ (Ref. 10) so that we have

$$S_{RS} \propto n_c N_c^2. \quad (2)$$

Under the assumption that all the atoms are condensed into clusters in the gas expansion, the number of clusters in the scattering region can be expressed as

$$n_c \propto nl/N_c, \quad (3)$$

for a given laser beam, where n is the atom number density in the scattering region and l the dimension of the scattering region along the laser beam. Thus, a Rayleigh scattering signal can be rewritten as

$$S_{RS} \propto nlN_c. \quad (4)$$

For a stable gas flow, n is determined by the continuity equation $n_0 v_0 A_0 = nvA$, where n_0, v_0, and A_0, are the atom number density, the flow velocity, and the area of cross section of gas flow at the nozzle throat, respectively, and n, v, A, are the respective values in the scattering region. At a distance of a few nozzle-orifice diameters downstream, the flow velocity v reaches its final value $[\gamma/(\gamma-1)]^{1/2} v_0$, where γ is the specific heat ratio of gas.[9] Then the average cluster size for a given backing pressure P_0 can be expressed as

$$N_c \propto S_{RS} A/(n_0 A_0 l). \quad (5)$$

Because n_0 is proportional to P_0 and A_0 is the same ($A_0 = \pi d_s^2/4$) for the different nozzles, i.e., the area of the 0.5 mm diameter orifice of a pulsed valve in our case, the average cluster size can also be given by

$$N_c \propto S_{RS} A/(P_0 l). \quad (6)$$

From Eq. (6), it is possible to compare the relative average sizes of clusters N_c among different nozzles using the experimental results of the scattering signal S_{RS}, the dimension l of scattering region and the area A of gas flow at the scattering region in the cluster jet.

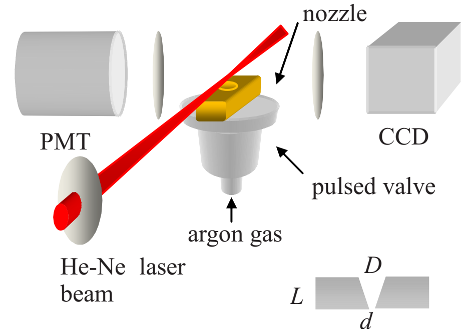

FIG. 1. (Color online) Schematic diagram of the experimental setup. The schematic side cross sectional view of a conical nozzle geometry is shown in the right low corner (d=0.5 mm, L=5.0 mm, and the sonic nozzle is made of stainless steel and the conical nozzles are made of brass).

III. EXPERIMENTS

The experimental setup is shown in Fig. 1, which was described in more detail in Ref. 17. A pulsed valve (Parker series 99) with a 0.5 mm diameter orifice was used (1/12 Hz repetition rate is set). The 0.5 mm diameter orifice itself was used as a sonic nozzle (d_s=500 μm). A conical nozzle was directly connected to the pulsed valve. In this work, four conical nozzles were used, which have the same throat diameter d of 500 μm and the nozzle length L of 5 mm but the different half opening angles α=8.5°, 14.0°, 19.3°, and 24.2° (the corresponding dimensions of a nozzle exit D=2 mm, 3 mm, 4 mm, and 5 mm, respectively). A pressure of 3×10^{-5} Torr in the chamber ($59 \times 53 \times 30$ cm^3) was maintained before the operation of the pulsed valve. Argon clusters were produced from the adiabatic expansion of a high pressure argon gas into vacuum through one of these nozzles. As shown in Fig. 1, a He–Ne laser (Uniphase 1125, 632.8 nm, and 10 mW) beam was focused into the center of argon gas flow about 2.5 mm above from the nozzle exit by a lens with a focal length of 20 cm. The laser beam diameter in the gas jet was estimated to be less than 1 mm. A 2 in. diameter lens with a focal length of 7.5 cm was mounted about 19 cm away from the laser beam to collect and image the 90° Rayleigh scattering light onto a head-on type PMT (−1 kV is applied). The output signal from the PMT was recorded by a 1 GHz bandwidth digital oscilloscope (Teketronix TDS5104). Meanwhile, a CCD camera located at the opposite side of the PMT was used to image 90° Rayleigh scattering light from the cluster jet. In our work, the collection solid angle subtended by the optical system described above was large enough to cover all the scattering regions for different nozzles. One optical setup was used for different nozzles.

IV. RESULTS AND DISCUSSION

As demonstrated in Ref. 17, the experimental data were obtained at the valve opening time of 3 ms (unless otherwise mentioned), which guarantees the buildup of steady state gas flow for these conical nozzles. First, we investigated the dependence of Rayleigh scattering signal S_{RS} on the backing pressure P_0 for four different conical nozzles. The experimental results are shown in Fig. 2(a). Every data point was

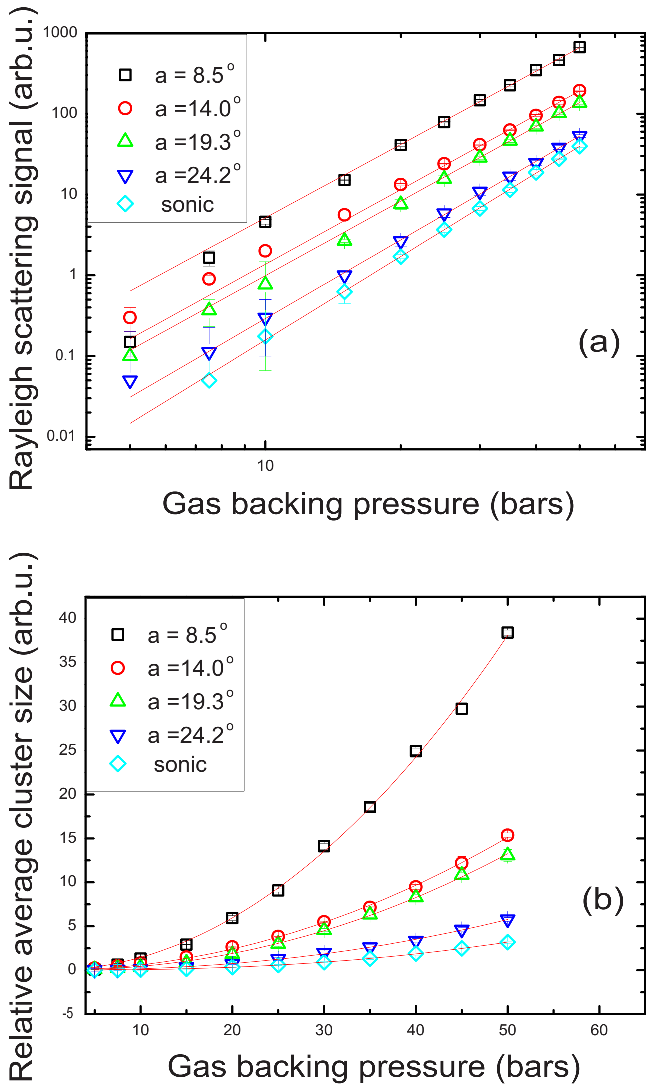

FIG. 2. (Color online) Pressure dependence of (a) Rayleigh scattering signal and (b) the relative average cluster size for different conical nozzles and a sonic nozzle. The solid lines represent the fit to the data.

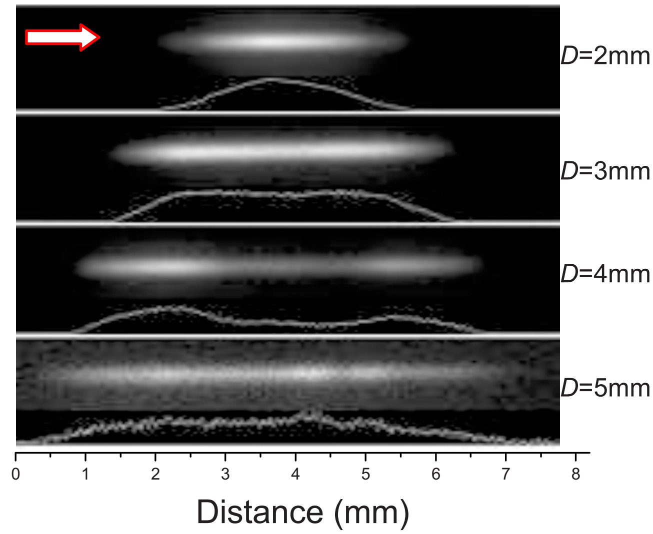

FIG. 3. (Color online) CCD images of the scattered light for conical nozzles with different half opening angles α=8.5°, 14.0°, 19.3°, and 24.2° (the corresponding dimensions of a nozzle exit D=2 mm, 3 mm, 4 mm, and 5 mm, respectively) at a backing pressure of 50 bars. The laser beam direction is shown as the arrow. The white curves show the profiles of the scattered light along the laser beam. (Note that for a D=5 mm conical nozzle, the image was obtained at the valve opening time of 5 ms due to the weaker scattering signal. The scales of signal intensity for the different nozzles are different).

obtained by averaging more than 12 gas pulses. By fitting these data points, the pressure power dependence of scattering signal $S_{RS} \sim (P_0)^\beta$ were obtained with β being 3.04 ± 0.03, 3.07 ± 0.01, 3.07 ± 0.01, and 3.23 ± 0.15 for the conical nozzles of α=8.5°, 14.0°, 19.3°, and 24.2°, respectively. It is found that there is a little difference in power exponent β among different nozzles, and β in our work is in the range of reported values by other groups.[10–18] The experimental results also show that the conical nozzle with a smaller half opening angle α produces the stronger scattering signal. It would be due to both the larger clusters and the higher atom density in the gas flow, as discussed below. It is interesting to find that for the conical nozzles, as the half opening angle decreases, the pressure dependence becomes weaker, i.e., power β gradually decreases, which will be discussed in the following.

To compare the average cluster size N_c produced from the different nozzles, the dimension l of a scattering region and the area of cross section A of gas cluster jet at the scattering region are required from Eq. (6), i.e., $N_c \propto S_{RS}A/(P_0 l)$. In our work, the CCD image from the scattered light was used to estimate the scattering dimension l and the area A. Figure 3 shows the typical CCD images at 2.5 mm away from the nozzle exit for different conical nozzles at a backing pressure of 50 bars. The white curves in Fig. 3 show the profiles along the laser beam at the center of the image, i.e., the spatial distribution of the scattered light. We defined the dimension l as the length of scattering region where the scattered light intensity is higher than 3% of the maximum intensity.[17] From the CCD images, the dimensions of the scattering region l were obtained to be 3.7 mm, 5.1 mm, 6.1 mm, and 7.0 mm for the conical nozzles of α =8.5°, 14.0°, 19.3°, and 24.2° (corresponding D=2 mm, 3 mm, 4 mm, and 5 mm), respectively. Then the area A ($\pi l^2/4$) can be obtained. Using Eq. (6), we calculated the relative average argon cluster size at a backing pressure of 50 bars for the nozzles. By A and l at a pressure of 50 bars, the relative cluster size at the lower backing pressure was obtained and shown in Fig. 2(b). Figure 2(b) shows the dependence of relative cluster size on the backing pressure. It is indicated that as the opening angle of a conical nozzle decreases, the cluster size increases. For example, at a given pressure, the cluster size is the largest for the α=8.5° nozzle, while the cluster size is the smallest for the α=24.2° nozzle. That is to say, a conical nozzle with a smaller opening angle is useful for the formation of larger clusters. This is qualitatively in agreement with that expected by Hagena's scaling law for a conical nozzle. By fitting the relative cluster sizes to the pressure power dependence $N_c \propto (P_0)^{\beta *}$, the powers β^* of 2.02 ± 0.02, 2.00 ± 0.02, 2.11 ± 0.04, and 2.23 ± 0.14 were obtained for the conical nozzles of α=8.5°, 14.0°, 19.3°, and 24.2°, respectively. Note that the power β^* is approximately equal to $\beta - 1$ based on Eq. (6). Like the pressure dependence of Rayleigh scattering signal, it is interesting to find that for

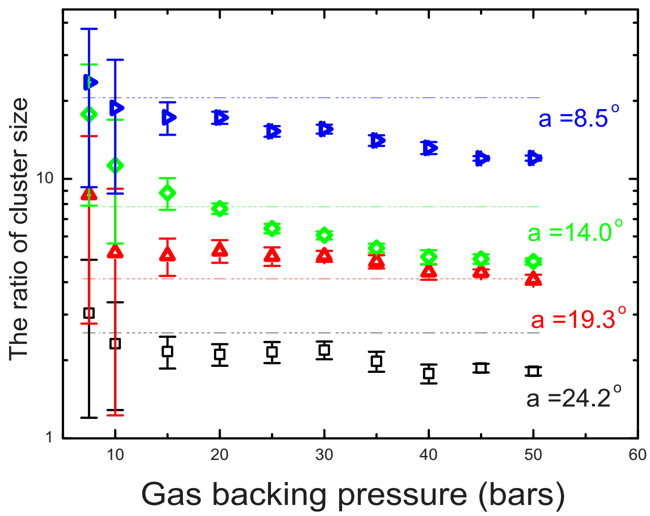

FIG. 4. (Color online) Experimental ratio η_{exp} of the relative average cluster size for different conical nozzles to that for the sonic nozzle with a 500 μm diameter orifice. The dashed lines represent the ratios η expected by Hagena's scaling law for different conical nozzles. Note that the large errors at lower backing pressure result from the fluctuation of weak Rayleigh scattering signal.

the conical nozzles, as the half opening angle decreases, the pressure dependence of the cluster size becomes weaker, i.e., power β^* shows the decreasing tendency. For example, the power β^* decreases from 2.23 to 2.02 as the α decreases from 24.2° and 8.5°, respectively. This implies that the pressure dependence of cluster size for conical nozzles is related to the opening angle, i.e., nozzle geometry. This is different from Hagena's scaling law, where the power is kept constant as 2.35.

To compare experimental cluster size quantitatively with that expected by Hagena's scaling law, we calculated the experimental ratios of the relative average cluster size for these conical nozzles to that for the a sonic nozzle from our experimental results and defined them as η_{exp}. Rayleigh scattering signal and the relative average cluster size for the sonic nozzle are shown in Figs. 2(a) and 2(b), respectively.[17] The experimental ratios η_{exp} are shown in Fig. 4. Based on Hagena's scaling law, η is kept constant at different backing pressure. As shown in Fig. 4, the corresponding η is 20.6, 7.8, 4.1, and 2.6 for the conical nozzles of α=8.5°, 14.0°, 19.3°, and 24.2°, respectively. Note that η is calculated from $(0.74/\tan \alpha)^{0.85\times 2.23}$, where our experimental power β^* (2.23) for the conical nozzle of α=24.2° is used. It is found from Fig. 4 that the experimental ratio η_{exp} exhibits a different case and varies as the backing pressure. At a lower backing pressure (<30 bars), the experimental ratio η_{exp} is roughly in agreement with η, especially for the conical nozzle of α=19.3° and 24.2°. For example, at a backing pressure of 30 bars the ratios η_{exp} are 15.6, 6.1, 5.0, and 2.2 for the conical nozzles of α=8.5°, 14.0°, 19.3°, and 24.2°, respectively. At a high backing pressure (50 bars), the ratios η_{exp} (4.1 and 1.8) is roughly in agreement with η (4.1 and 2.6) for the conical nozzles (α=19.3° and 24.2°, respectively). On the other hand for the α=8.5° and 14.0° conical nozzles, a poor agreement is observed. The experimental ratios η_{exp} are 12.0 and 4.8 for the conical nozzles of α=8.5° and 14.0° at a backing pressure of 50 bars, respectively, which is much lower than η (20.6 and 7.8). This means that Hagena's scaling law overestimates the cluster size for conical nozzles of small opening angle at high backing pressures. That is to say, the argon cluster size deviates from what is expected by Hagena's scaling law at high gas backing pressure; the deviation becomes larger, as the opening angle gets smaller. It could be related to the fact that Hagena's scaling law is based on experimental data at lower backing pressures and the idealized equivalent diameter model is used, in which the effect of the boundary layers in a conical nozzle is not taken into account. We studied the case of the high backing pressure, which is a usual situation in laser-cluster interaction experiments. At a higher backing pressure, the inside wall of a conical nozzle could has more effect on the gas flow formation, making the effective space for the cluster formation restricted, as discussed in Ref. 19. It is interesting to note that if β^*=2.0 (the experimental power for conical nozzle with α=8.5°) is used to calculate η, the ratios η will be 14.9 and 6.1, and then the difference between η and η_{exp} tends to decrease, though η is still larger than η_{exp}. At present, although it is not clear that how the inside wall a conical nozzle makes effect on the gas flow, it can be concluded that a conical nozzle of a small opening angle degrades in cluster formation at high packing pressure.

V. CONCLUSIONS

Measuring the scattered light intensity by PMT together with the scattered light image taken by CCD, the relative average size of clusters formed in argon gas jet has been investigated. A jet of argon clusters were developed from conical nozzles with the same throat diameter but the different exit diameters. The results show that the power dependence of Rayleigh scattering signal and relative cluster size on pressure is related to the opening angle of conical nozzle. It is interesting to note that the pressure dependence of cluster size becomes weaker as the opening angle decreases. The detailed comparison of cluster size between experimental results and prediction from Hagena's scaling law was performed. It is found that the average cluster size increases with the opening angle of a conical nozzle, as expected by scaling law. However the argon cluster size itself deviates from what is expected for a conical nozzle of a smaller opening angle at a high backing pressure; the performance of conical nozzle in terms of cluster formation degrades for a smaller opening angle at a high backing pressure.

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