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Understanding of cluster size deviation by measuring the dimensions of cluster jet from conical nozzles

Guanglong Chen,[1] Xiaotao Geng,[2] Hongxia Xu,[1] Yiming Mi,[1] Xiuli Zhang,[1] Lili Wang,[1] and Dong Eon Kim[2,3]
[1]*School of fundamental studies, Shanghai University of Engineering Science, Shanghai 201620, China*
[2]*Department of Physics and Center for Attosecond Science and Technology (CASTECH), Pohang University of Science and Technology (POTECH), Pohang, Kyungbuk 790-784, Korea*
[3]*Max Planck Center for Attosecond Science, Pohang, Kyungbuk 790-784, Korea*

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This work aims to understand the cluster size deviation from the prediction by an existing scaling law for conical nozzles. The dimensions of cluster jet at different heights above a nozzle along the direction of gas flow are measured. This study indicates that the dimension of cluster jet is underestimated in the existing scaling law and this under-estimation leads to the over-estimation of the equivalent diameter of conical nozzle. Thus the underestimation of the dimension of cluster jet may be one of possible factors responsible for the cluster size deviation (the degree of the deviation depends on details of cluster jet). *Copyright 2013 Author(s). This article is distributed under a Creative Commons Attribution 3.0 Unported License.* [http://dx.doi.org/10.1063/1.4796187]

In the study of laser-matter interaction, a clustered gas jet has severed as a good target sample in many areas of research interest,[1] such as the deuterium-deuterium nuclear fusion,[2,3] plasma waveguide generation,[4–6] laser acceleration,[7,8] pulsed x-ray,[9] harmonic generation,[10] and so on. Recently the interaction of clusters with intense X-ray pulse also attracts interest.[11] Because the cluster size information is important in understanding of laser-cluster interaction, many works have been done to characterize the cluster jet in terms of cluster size.[12–23] It is well known that the average cluster size N_c (the number of atoms per cluster) in a cluster jet can be predicted by Hagena's scaling law $N_c = 33^* (\Gamma^*/1000)^{2.35}$.[12,13] Γ^* is called as Hagena's empirical parameter and expressed as $K d_{eq}{}^{0.85} P_0/T_0{}^{2.29}$, where K is a constant related to the property of a gas species (e.g. $K = 1646$ for argon gas), d_{eq} is the equivalent diameter, and P_0, T_0 are a initial gas backing pressure in mbar and a gas temperature in Kelvin before expansion, respectively. If the cluster jet is produced through a sonic nozzle, d_{eq} is the throat diameter of sonic nozzle d_s [Fig. 1(a)]. In this case the cluster jet is called as a free jet. If a conical nozzle is used, d_{eq} is replaced by 0.74 $d/\tan\alpha$ for rare gas, where d is its throat diameter and α the half opening angle [Fig. 1(b)]. Obviously for a conical nozzle with a small half opening angle α, d_{eq} becomes much larger than its throat diameter d. Thus a conical nozzle is quite often employed in experiments where large-size clusters are needed because the cluster size N_c is proportional to $(d_{eq}{}^{0.85})^{2.35}$ for a given K, P_0 and T_0 based on Hagena's scaling law.[3]

However it have been noticed that the cluster size deviates from the prediction by the scaling law, especially in the case of high backing pressure and small opening angle of a conical nozzle.[20–22] Our study aims to understand the causes for such a deviation.

To understand the cluster size deviation, it is necessary to further interpret d_{eq}. For the axisymmetric gas expansion of a free gas jet produced through a sonic nozzle (d_s), it is known that the flow field is similar to that of a point source close to the nozzle throat, as shown in Fig. 1(a), and the free jet expansion is two dimensional.[13,14] For the free gas jet, elementary gas dynamics yields the basic relationship between the local atom number density n, temperature T, and the flow velocity w at the centerline of jet and the respective values n_0, T_0, and the most probable thermal velocity v_0 at the gas source. At a few d_s downstream, the flow velocity w approaches its final value

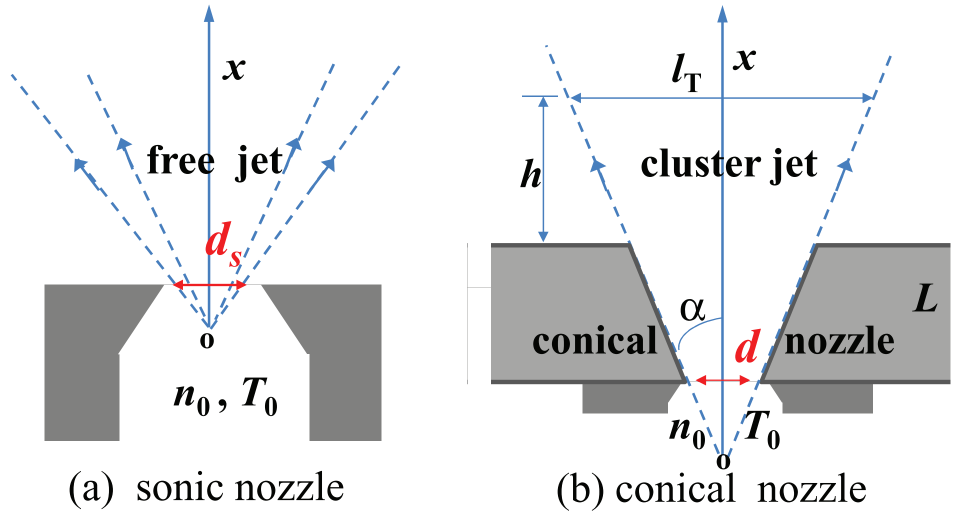

FIG. 1. The schematic diagrams for streamline of gas jet into vacuum from a sonic nozzle (a) and a conical nozzle (b) based on the idealized straight streamline model.

$w_{max} = (\gamma/(\gamma\text{-}1))^{1/2}v_0$ (where γ is the ratio of specific heats of gas), and the density ratio can be given by $n/n_0 = (M^2\ (\gamma\text{-}1)/2)^{\text{-}1/(\gamma\text{-}1)}$ (where $M = 3.26(x/d_s)^{(\gamma\text{-}1)}$ is the flow Mach number.[13,14] Then the density ratio for the monatomic gas ($\gamma = 5/3$) is then written by

$$n/n_0 = 0.15(x/d_s)^{-2}. \tag{1}$$

When the gas expands into vacuum through a conical nozzle (d, α), the gas jet is restricted by the conical nozzle wall and is not a free jet; hence, the one-dimensional flow theory is used to describe it. Under the idealized straight streamline model in scaling law, the expansion angle of gas jet is the same as the opening angle of nozzle, as shown in Fig. 1(b).[13] From Fig. 1(b), the ratio of a cross section area at a distance x in gas jet A_T ($= \pi l_T{}^2/4$) to the area of nozzle throat A^* ($= \pi d^2/4$) can be given by

$$A_T/A^* = 4(x/(d/\tan\alpha))^2 \tag{2}$$

Applying the continuity equation to a cross section A_T in the gas jet ($n^*w^*A^* = nw_{max}A_T$, where $n^* = (2/(\gamma+1))^{1/(\gamma\text{-}1)}n_0$ and $w^* = (\gamma/(\gamma+1))^{1/2}v_0$ are the atom density and the flow velocity at the throat of the conical nozzle, respectively),[13,14] the density ratio for the monatomic gas ($\gamma = 5/3$) is obtained as

$$n/n_0 = 0.325\ A^*/A_T. \tag{3}$$

By using Eqs. (2) and (3), the density ratio is given as

$$n/n_0 = 0.15(\mathrm{x}/(0.74\,d/\tan\alpha))^{-2}. \tag{4}$$

If we compare Eq. (1) with Eq.(4), it is noted that at the same distance downstream in a gas jet under the same source condition, a conical nozzle (d, α) produces the same density as a sonic nozzle with a throat diameter of $d_s = 0.74\ d/\tan\alpha$. In other words, if $\tan\alpha < 0.74$, a conical nozzle of throat diameter d can produce a gas jet with higher atom density than that produced through a sonic nozzle of the same throat diameter ($d_s = d$). In Hagena's scaling law, $0.74\ d/\tan\alpha$ is denoted by d_{eq} Obviously d_{eq} is introduced to represent an effective throat diameter of a conical nozzle from the viewpoint of atom density in a gas jet. From the discussion above, it is easily noticed that d_{eq} is closely related to the atom density. Because the atom density depends on the cross section area of gas flow, the estimation of the expansion size of gas flow is important for the estimation of d_{eq}. Considering d_{eq} is independent on the gas species, this work selects the argon jet as an example,

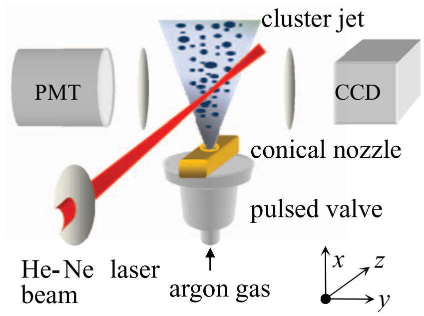

FIG. 2. The schematic diagram of the experimental setup.

and aims to experimentally investigate the dimensions of a gas jet and d_{eq}, and then understand the cluster size deviation.

Figure 2 shows the schematic diagram of the experimental setup.[22] A cluster jet was produced by the adiabatic expansion of a high pressure argon gas into vacuum through a conical nozzle on a pulsed valve. The gas backing pressure was kept at 50 bars, and three conical nozzles were used, which have the same throat diameter ($d = 500$ μm) and nozzle length ($L = 5$ mm), but different half opening angle ($\alpha = 8.5^o$, 14.0^o and 19.3^o, i.e, the corresponding dimensions of a nozzle exit are 2 mm, 3 mm and 4 mm). A He-Ne laser beam was focused into the center of the cluster jet by a lens ($f = 20$ cm). A lens ($f = 10$ cm) was used to image the 90° Rayleigh scattered light from the cluster jet onto a CCD camera placed about 42 cm away from the laser beam. To monitor the cluster formation using Rayleigh scattering measurement in the gas jet, another 2-inch. dia. lens ($f = 7.5$ cm) was placed at the opposite side of CCD about 19 cm away from the laser beam to collect the 90° Rayleigh scattered light onto a head-on type PMT. The output signal from the PMT was recorded by a 1 GHz bandwidth digital oscilloscope. An encoded motorized stage was used to adjust the position of the pulsed valve and the nozzle along the direction of gas flow which is perpendicular to the laser beam (i.e., x-direction in Fig. 2). Thus the height of the laser beam above the conical nozzle was changed.

As mentioned in Ref. 22, the signals from PMT indicate that Rayleigh scattering signals reach the steady state and have the flat top profiles when the pulsed valve opening time is longer than 3 ms. Our experimental results are obtained at the valve opening time of 3 ms. The typical CCD images of scattered light in case of the conical nozzle with $\alpha = 14.0^o$ are shown in Fig. 3 (the negative images of CCD images). To clearly illustrate the evolution of gas flow, the images at different heights were put together. The corresponding heights are from 1.8 mm (bottom) to 4.8 mm (top) by a step of 0.2 mm. The laser beam propagated from left to right (z-direction in Fig. 2). From Fig. 3, it is clear that the dimension of scattering region gets larger and the intensity of scattered light becomes weaker with the increase of height.

To more clearly show the changes in the dimension of gas flow and the scattered light intensity, the profiles corresponding to the heights of 1.8 mm, 3.2 mm and 4.8 mm were plotted together in Fig. 4. Note that all the profiles were plotted as waterfall together and inserted in Fig. 4. The gas flow evolution is clearly seen. Obviously the cluster jet has the expansion angle and the area of cross section of the gas flow becomes larger gradually. Based on the continuity equation or Eq. (3), the atom density will decrease along the gas flow. Thus the decrease of scattered light intensity should be related to the decrease of atom density because scattered light intensity depends on the atom density and the cluster size in cluster jet. It is interesting to note that the intensity at the center of gas jet is a little weak, i.e., the intensity distribution of scattered light does not have a flat top. This non-uniform distribution of intensity has also been observed for other conical nozzles in our study.

To quantitatively demonstrate the change in the dimension of gas flow, we define the dimension l as the length of scattering region where the scattered light intensity is higher than 3% of the maximum intensity. This definition is taken, based on the fact that the dimension of gas flow should be equal to the dimension of a zero-opening-angle-nozzle exit, as discussed in Ref. 22. The results are shown

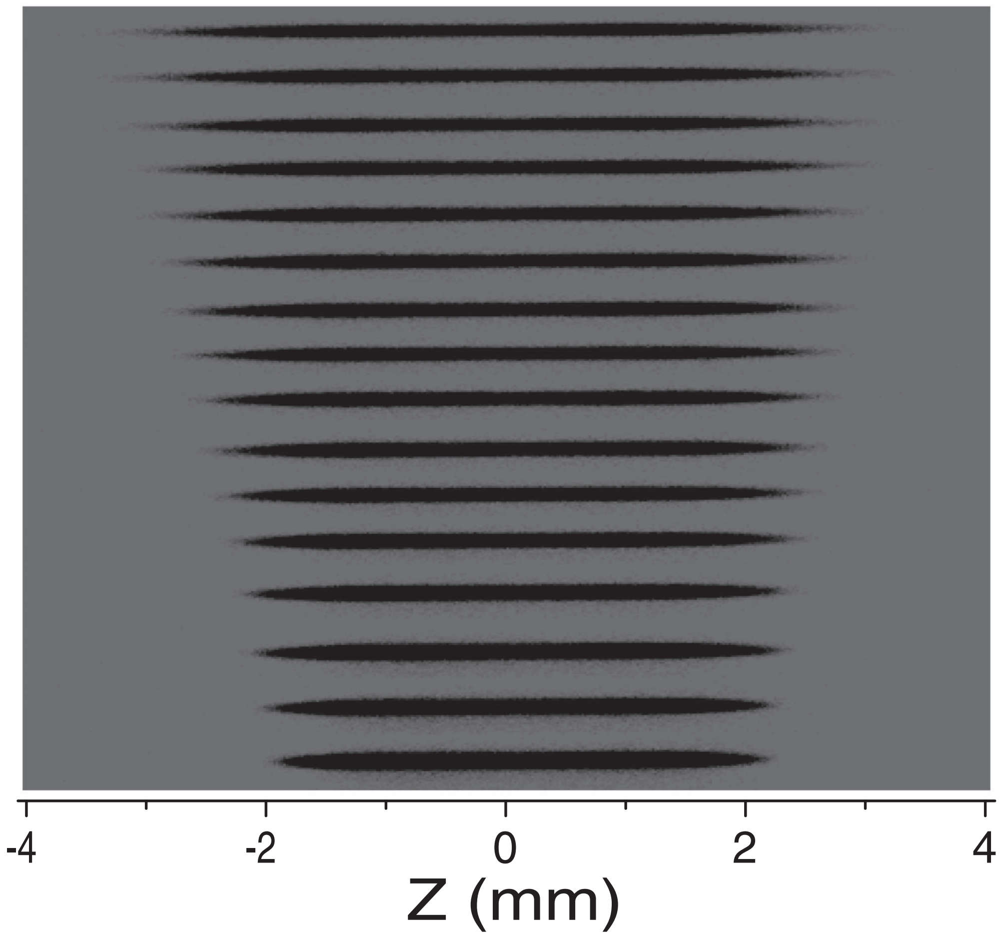

FIG. 3. CCD images at different heights for the conical nozzle of $\alpha = 14.0^{o}$. The images from top to bottom correspond to the heights above nozzle which are 4.8 mm to 1.8 mm by a step of 0.2 mm. The laser beam propagates from left to right.

in Fig. 5 (solid symbols). To compare the experimental dimensions with theoretical dimensions l_T in the idealized straight streamline model, the theoretical dimension l_T of gas flow from a conical nozzle were also calculated and plotted together in Fig. 5 (open symbols). It is clear that as the height increases, the dimension of gas flow do increases, which is in agreement with the theoretical dimensions l_T. However it is found that the experimental dimension of gas flow is always larger than the theoretical one at every height. That is to say, the real dimension of gas flow is different from that predicted by the idealized straight streamline model. The similar result can be found in Fig. 2 of Ref. 18.

For the further discussion of this deviation, firstly we define (1) the dimension ratio of l to l_T by $\eta = l / l_T$, (2) the experimental area of cross section of gas flow by $A(=\pi l^2/4)$ and (3) the theoretical area of cross section of gas flow by $A_T(=\pi l_T{}^2/4)$ based on the idealized straight streamline model. In our case, the throat area A^* is kept constant. Clearly the dimension ratio η reflects the dimension deviation, and the area ratio A/A_T is η^2. From Eq. (3), we can obtain the atom density ratio for a conical nozzle by:

$$n/n_0 = 0.325\, A^*/A = 0.325\eta^{-2}\, A^*/AT. \tag{5}$$

Combining Eqs. (2) and (5), the atomic density ratio is given as

$$\begin{aligned} n/n_0 &= 0.325\eta^{-2} \quad (x/(d/\tan\alpha))^{-2}/4 \\ &= 0.15(\mathrm{x}/(0.736\eta^{-1}d/\tan\alpha))^{-2}. \end{aligned} \tag{6}$$

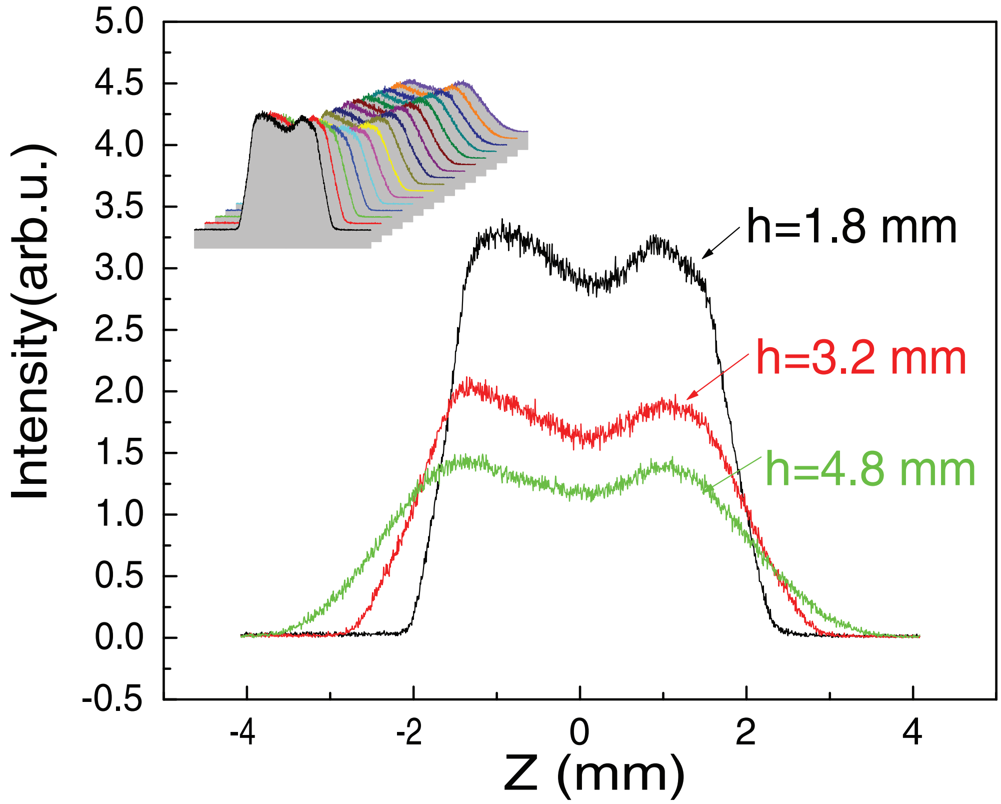

FIG. 4. The profiles of CCD images corresponding to the heights of 1.8 mm, 3.2 mm and 4.8 mm for the conical nozzle of $\alpha = 14.0^o$. The profiles at all heights were inserted in the top left corner.

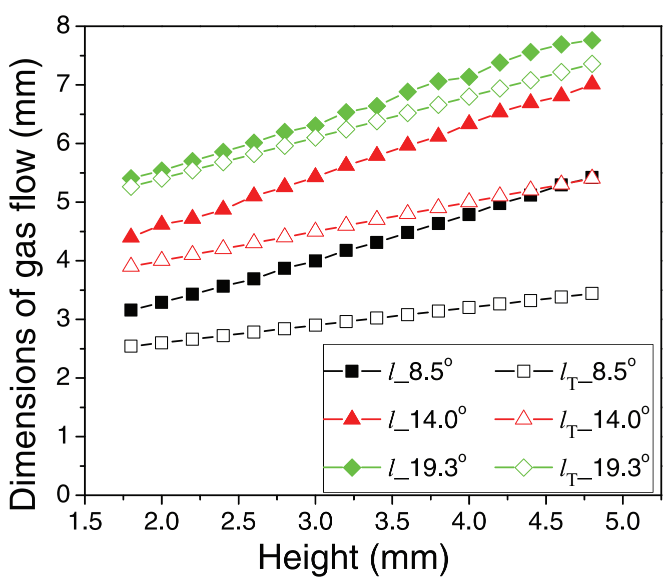

FIG. 5. The comparisons of experimental dimensions (solid symbols) with those predicted by the straight streamline model (open symbols) at different heights for three conical nozzles of $\alpha = 8.5^o$, 14.0^o and 19.3^o.

TABLE I. The dimension ratios η at different heights above nozzle for three conical nozzles.

	Height (mm)															
	1.8	2.0	2.2	2.4	2.6	2.8	3.0	3.2	3.4	3.6	3.8	4.0	4.2	4.4	4.6	4.8
$\alpha = 8.5^{o}$,	1.24	1.26	1.29	1.31	1.33	1.36	1.38	1.41	1.43	1.46	1.48	1.50	1.53	1.54	1.57	1.58
$\alpha = 14.0^{o}$	1.13	1.15	1.15	1.16	1.19	1.19	1.21	1.22	1.23	1.24	1.25	1.27	1.29	1.30	1.28	1.30
$\alpha = 19.3^{o}$	1.03	1.03	1.03	1.03	1.03	1.04	1.03	1.05	1.04	1.06	1.06	1.05	1.06	1.07	1.07	1.05

Comparing Eqs. (6) with (1), we can finally obtain the experimental equivalent diameter of a conical nozzle as

$$d_{eq}^{e} = 0.74\eta^{-1} d/\tan\alpha. \quad (7)$$

Clearly the experimental $d_{eq}{}^{e}$ is smaller than the theoretical d_{eq} (0.74 d/tan α) due to $\eta > 1$, and thus the estimated cluster size will be smaller than the theoretical size using $d_{eq} = 0.74\ d/\tan\alpha$ based on Hagena's scaling law ($N_c \sim d_{eq}{}^{0.85\times 2.35}$), i.e., the scaling law overestimates the cluster size. Using the dimension ratio η, the relation between experimental cluster size $N_c{}^{e}$ and theoretical N_c can be expressed by

$$N_c^{e} = N_c \eta^{-2.0}. \quad (8)$$

Thus the dimension ratio η reflects the deviation of not only the equivalent diameter of conical nozzle but also the cluster size. The dimension ratio can be used to estimate the cluster size deviation based on Eq. (8). The dimension ratios at different heights were calculated and listed in Table I (the second row). It is found that η generally become larger as the height increasing, i.e., η is related to the height. This means that the deviation in cluster size becomes larger as the height increases.

We also measured the dimensions l at different heights for other two conical nozzles of $\alpha = 8.5^{o}$ and 19.3^{o}, and then compared them with the theoretical dimensions l_T, as shown in Fig. 5. The similar results were obtained, i.e., the experimental dimensions of gas flow are always larger than theoretical ones. Moreover, the dimension deviation, i.e., the dimension ratio η becomes larger as the height increases. The dimension ratio η at different heights were also listed in Table I (the first and the third rows). Comparing the ratio η among these nozzles, it is not difficult to find that not only is the dimension ratio η related to the height, but also η decreases with the increase of the opening angle of conical nozzle, which means that the cluster size deviations decrease with the increase of the opening angle of conical nozzle. This result is in agreement with that discussed in Ref. 21 and 22. Considering that the gas flow close to the nozzle outlet experiences only the nozzle expansion, the actual gas flow dimension l should be the same as the theoretical dimension l_T. And then the corresponding experimental equivalent diameter $d_{eq}{}^{e}$ should be equal to the theoretical value d_{eq} (0.74 d/tan α). So we can quantitatively evaluate the cluster size in the gas flow close to the nozzle outlet using Hagena scaling law (i.e., $N_c = 33^* (\Gamma^*/1000)^{2.35}$). For example, the corresponding cluster sizes for the conical nozzles of $\alpha = 8.5^{o}$, 14.0^{o} and 19.3^{o} are 3.3×10^6, 1.2×10^6 and 6.0×10^5, respectively. Using these cluster sizes close to the nozzle outlet and the dimension ratios η listed in Table I, the cluster sizes at different heights for the three conical nozzles can be quantitatively evaluated based on Eq. (8). From the discussion above, it could be concluded that one of reasons for the fact that the cluster size for a conical nozzle deviates from that predicted by the scaling law is the underestimation of the dimensions of gas flow. This can be understood as follows: based on the continuity equation, the atom density is determined by the cross-section area of gas flow. Thus the underestimation of dimension of gas flow results in the overestimation of the atom density, while the high atom density is helpful for the formation of large-size clusters. In a sense, the straight streamline model could overestimate the atom density, and thus overestimate the cluster size. However, it is difficult to give a general quantitative relation between the experimental $d_{eq}{}^{e}$ ($N_c{}^{e}$) and the theoretical d_{eq} (N_c) because the ratio η depends on the opening angle of conical nozzle and the height above nozzle for a given conical nozzle. The proper simulation would be of great help in our better understanding of this issue.

In conclusion, we measured the dimensions of the argon jet at different heights along the direction of gas flow using a CCD camera by imaging Rayleigh scattered light from the cluster jet, and compared them with those predicted by the idealized straight streamline model for three conical nozzles of different opening angles. The results indicate that the cluster size deviation for a conical nozzle could result from the underestimation of dimension of gas flow, which is equivalent to the overestimation of equivalent diameter of conical nozzle. It is noted that the cluster size deviation depends on the height above a nozzle and the opening angle of a conical nozzle.

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