Size estimates of nobel gas clusters by Rayleigh scattering experiments

Pinpin Zhu (朱频频), Guoquan Ni (倪国权), and Zhizhan Xu (徐至展)

Laboratory for High Intensity Optics, Shanghai Institute of Optics and Fine Mechanics,
Chinese Academy of Sciences, Shanghai 201800

Received April 10, 2003

Noble gases (argon, krypton, and xenon) are puffed into vacuum through a nozzle to produce clusters for studying laser-cluster interactions. Good estimates of the average size of the argon, krypton and xenon clusters are made by carrying out a series of Rayleigh scattering experiments. In the experiments, we have found that the scattered signal intensity varied greatly with the opening area of the pulsed valve. A new method is put forward to choose the appropriate scattered signal and measure the size of Kr cluster.

OCIS codes: 140.0140, 290.5820.

Atomic cluster, consisting of a few to even millions of atoms, is a unique combination of both gas- and solid-phase components. Though a cluster medium has a unitary gas-like density, every cluster member has its own solid-like local density. Recently, many interests have been attracted on studying the interaction of atomic clusters with intense laser radiation[1−9]. It has been shown that large-size clusters absorb the laser energy very efficiently when they are irradiated[1]. In these interactions, some research groups have observed keV electrons[2], MeV ions[3−5] and X-rays in keV range[6−8], etc. And it is very exciting that when Ditmire *et al.* drove explosions in deuterium clusters with a 35-fs laser pulse, D-D nuclear fusion was produced[9]. Going with these experiments, theoretical studies have also been developed. Several models have been established to explain the experimental results[10−13]. Both experimental and theoretical studies have shown that the intense laser-cluster interaction processes have great correlation with the cluster size. So a good estimate of the cluster size is critical in interpreting the interaction experiments.

Noble gas clusters are the most commonly used clusters in the interaction studies. They can be produced in the adiabatic expansion of noble gases into vacuum through a nozzle. Because of many complex fluid dynamics problems in the expansion, there is currently no rigorous theory to predict cluster formation under these conditions. However, Hagena's empirical formula[14] can offer us a general guide to cluster formation

$$\Gamma^* = k\frac{(d/\mathrm{tg}\alpha)^{0.85}}{T_0^{2.29}}p_0, \tag{1}$$

where k is an empirical constant which depends on the gas species (1650 for Ar, 2900 for Kr, and 5500 for Xe), d is the diameter of the jet throat in units of μm, α is the jet expansion half angle, T_0 is the stagnation gas temperature in absolute scale and p_0 is the backing pressure in mbar. Many Studies on Hagena's parameter[15,16] show that the gas clustering generally begins for $\Gamma^* > 100 - 300$, with N_c (the number of atoms per cluster) scaling as

$$N_c \propto \Gamma^{*2-2.5}. \tag{2}$$

Thus we can use the strong variation of N_c with p_0 to estimate the mean N_c of a medium of clusters.

In this paper, we report a series of Rayleigh scattering experiments to measure the average cluster size of Ar_n, Kr_n, and Xe_n clusters.

The classical Rayleigh cross section is given by

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

where r is the radius of cluster, λ is the input light wavelength, and n is the refractive index of the scattering medium. If assuming all the atoms have condensed into spherical clusters, we can have $S_{\mathrm{RS}} \sim p_0 N_c$[3]. In our experiments, we have found that S_{RS} scales with p_0 as

$$S_{\mathrm{RS}} \sim p_0^{\beta}, \tag{4}$$

where β should vary from 2.5 to 3.5 inferred from expression (2). Then we can use the scaling

$$N_c \sim p_0^{\beta-1} \tag{5}$$

to estimate the average size of clusters. It must be mentioned that this scattering technique can only give information on the average size of the clusters, but no more information about the size distribution of the clusters can be obtained.

The experiments are carried out in the Laboratory for High Intensity Optics at Shanghai Institute of Optics and Fine Mechanics. The apparatus for the experiment is shown in Fig. 1.

As shown in Fig. 1, the scattering chamber is evacuated by a 3000-l/min oil diffusion pump to a base pressure of about 6×10^{-4} Pa. The noble gases (Ar, Kr and Xe) are puffed into the chamber via a supersonic nozzle, the nozzle has a diameter of 300 μm and half angle of 5°. With cool expansion, the nobel gases nucleate into clusters. After several microseconds, the clusters are irradiated by a pulsed laser beam (15 ns) from a frequency-doubled Nd:YAG laser in focused with a 50-cm focal length lens and intersect perpendicularly with the gas flow at 3-mm downstream from the nozzle. Because high-energy laser pulse can break up the clusters and cause incorrect information, the energy of the irradiating laser pulse must

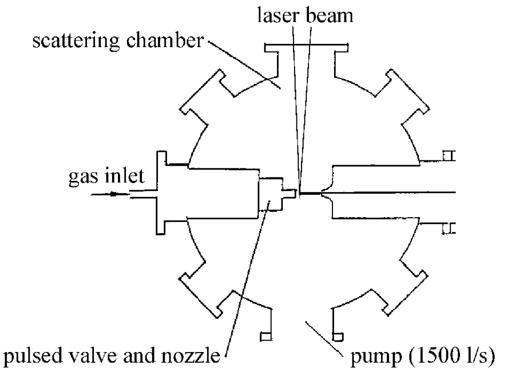

Fig. 1. Schematic of the experimental apparatus for our Rayleigh scattering experiment. It is part of our entire apparatus for laser-cluster interaction experiments.

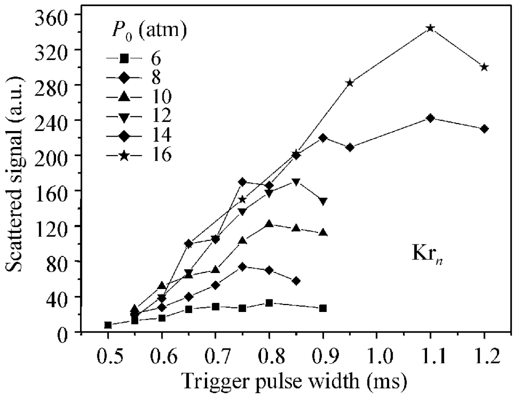

Fig. 2. Scattered signal intensity as a function of the trigger pulse width to the pulsed valve with the krypton gas backing pressure P_0 being a parameter.

not be very high. In our experiments, it is less than 0.3 mJ. The 90° scattered light is collected with a 7-cm focal length lens and imaged on a photomultiplier tube. The high voltage of the photomultiplier is above 1000 V. A 500-MHz bandwidth digital oscilloscope (Lecroy 9350AL) is used to score the electric signal.

More details about the valve must be noted, because a good understanding of the valve is vital to the correct interpretation of the results[17]. In our experiments, a dual-solenoid pulsed valve is used to produce gas atomic clusters. The valve can operate at high backing pressures (at up to 45 atm) without substantial leakage and it can maintain a vacuum system at $< 5 \times 10^{-4}$ Pa. Its operation is triggered by a square pulse signal whose width varies from 0.5 to 1.2 ms. The amount of puffed gas is determined by the effective opening area of the valve, which is proportioned to the trigger pulse width. But the proportion is not unique for different backing pressures. It is harder to open the valve at higher backing pressure.

We found that even at the same backing pressure, the scattered signal is variable for different opening area of the valve. So how to choose the appropriate scattered signal is a complicated thing. In our former works[18], we chose the scattered signals under the same effective opening area of the valve. And in order to do that, we adjusted the worst vacuum to increase proportionally with the backing pressure. But that was an approximate method and it cannot give us sufficient explanation to do so. In our current experiments, we have studied the relationship between the scattered signal and the trigger pulse width. The results of Kr clusters at backing pressures from 6 to 16 atm are shown in Fig. 2. We can see that at every backing pressure, the scattered signals of Kr clusters increase with the trigger pulse width and get saturated at a particular trigger pulse width. The saturated values of the scattered signal at different backing pressures may correspond to the state that the gases are puffed into vacuum with full expansion. So we can choose these saturated values as the appropriate scattered signals.

In our experiments, the former method is used for Ar and Xe clusters while the new method is used for Kr cluster.

By assuming that the observed onset of clustering corresponds to $N_c \approx 100$, we can use the increase in scattered signal to calculate the cluster size as a function of

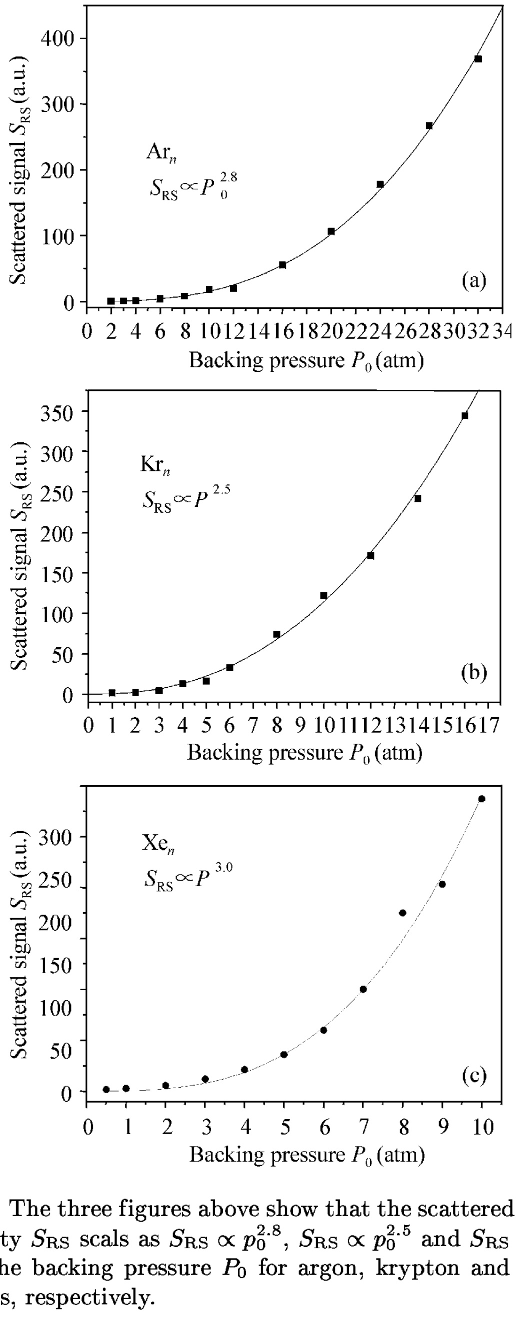

Fig. 3. The three figures above show that the scattered signal intensity S_{RS} scals as $S_{RS} \propto p_0^{2.8}$, $S_{RS} \propto p_0^{2.5}$ and $S_{RS} \propto p_0^{3.0}$ with the backing pressure P_0 for argon, krypton and xenon clusters, respectively.

Table 1. Estimated Sizes of Ar, Kr and Xe Clusters at Different Backing Pressure

	Ar Cluster		Kr Cluster		Xe Cluster	
Backing Pressure (atm)	N_c	Radius R_c (nm)	N_c	Radius R_c (nm)	N_c	Radius R_c (nm)
1	30	0.6	100	1.0	400	1.7
2	100	0.9	280	1.4	1.6×10^3	2.8
3	210	1.1	520	1.7	3.6×10^3	3.6
4	350	1.3	800	2.0	6.4×10^3	4.4
5	520	1.5	1120	2.2	1.0×10^4	5.1
6	720	1.7	1470	2.4	1.44×10^4	5.8
8	1210	2.0	2260	2.8	2.56×10^4	7.0
10	1810	2.3	3160	3.1	4.0×10^4	8.1
14	3320	2.8	5240	3.7	7.8×10^4	10.1

backing pressure. Figures 3(a), (b) and (c) show the scattered signal $S_{\rm RS}$ as the function of backing pressure p_0 for Ar, Kr and Xe clusters ($S_{\rm RS} \sim p_0^\beta$). The values of parameter β are 2.8, 2.5 and 3.0, respectively. Using these data we have estimated the average cluster size of Ar, Kr and Xe clusters, the results are shown in Table 1. We can infer that, at $p_0 = 10$ atm, an average Ar cluster contains about 1800 atoms and has a radius $R_c \sim 2$ nm, while Kr cluster contains about 3200 atoms and has a radius $R_c \sim 3$ nm, Xe cluster contains about 4.0×10^4 atoms and has a radius $R_c \sim 8$ nm.

It is worth noting that the scaling parameter β of Kr is 2.5. This value is smaller than that of Xe, even smaller than that of Ar. It does not mean that the average size of Kr is smaller than that of Ar, because krypton gas begins clustering at lower backing pressure. From Table 1, we can see that clearly.

In conclusion, we carried out a series of Rayleigh scattering experiments to measure the cluster size of noble gas clusters and got fairly good results. The formation of clusters is so complicated that it cannot be exactly explained with Hagena's empirical relations. From our experiments we can see that the scattering signal greatly depends not only on the backing pressure but also on the opening area of the valve. We put forward a new method to choose the appropriate scattered signal and use it on measuring the size of Kr cluster. We can see that the scattered signal increases with backing pressure, which shows a good fitting to power function.

P. Zhu's e-mail address is purple@siom.ac.cn.

References

1. T. Ditmire, R. A. Smith, J. W. G. Tisch, and M. H. R. Hutchinson, Phys. Rev. Lett. **78,** 3121 (1997).
2. Y. L. Shao, T. Ditmire, J. W. G. Tisch, E. Springate, J. P. Marangos, and M. H. R. Hutchinson, Phys. Rev. Lett. **77,** 3343 (1996).
3. T. Ditmire, E. Springate, J. W. G. Tisch, Y. L. Shao, M. B. Mason, N. Hay, J. P. Marangos, and M. H. R. Hutchinson, Phys. Rev. A **57,** 369 (1998).
4. T. Ditmire, J. W. G. Tisch, E. Springate, M. B. Mason, N. Hay, R. A. Smith, J. Marangos, and M. H. R. Hutchinson, Nature **386,** 54 (1997).
5. M. Lezius, S. Dobosz, D. Normand, and M. Schmidt, Phys. Rev. Lett. **80,** 261 (1998).
6. A. McPherson, T. S. Luk, B. D. Thompson, K. Boyer, and C. K. Rhodes, Appl. Phys. B **57,** 337 (1993).
7. T. Ditmire, T. Donnelly, R. W. Falcone, and M. D. Perry, Phys. Rev. Lett. **75,** 3122 (1995).
8. S. Dobosz, M. Lezius, M. Schmidt, P. Meynadier, M. Perdrix, and D. Normand, Phys. Rev. A **56,** R2526 (1997).
9. J. Zweiback, R. A. Smith, T. E. Cowan, G. Hays, K. B. Wharton, V. P. Yanovsky, and T. Ditmire, Phys. Rev. Lett. **84,** 2634 (2000).
10. C. Rose-Petruck, K. J. Schafer, K. R. Wilson, and C. P. J. Barty, Phys. Rev. A **55,** 1182 (1997).
11. T. Ditmire, T. Donnelly, A. M. Rubenchik, R. W. Falcone, and M. D. Perry, Phys. Rev. A **53,** 13 (1996).
12. I. Last and J. Jortner, Phys. Rev. A **62,** 14 (2000).
13. S. X. Hu and Z. Z. Xu, Phys. Rev. A **56,** 3916 (1997).
14. O. F. Hagena, Z. Phys. D **4,** 291 (1987).
15. O. F. Hagena and W. Obert, J. Chem. Phys. **56,** 1793 (1972).
16. J. Farges, M. F. de Feraudy, B. Raoult, and G. Torchet, J. Chem. Phys. **84,** 3491 (1986).
17. R. A. Smith, T. Ditmire, and J. W. G. Tisch, Rev. Sci. Instrum. **69,** 3798 (1998).
18. A. L. Lei, H. J. Zhai, B. C. Liu, Z. Li, G. Q. Ni, and Z. Z. Xu, Chin. Phys. Lett. **17,** 661 (2000).