Vol 12 No 8, August 2003
1009-1963/2003/12(08)/0856-05

Chinese Physics

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Investigation of the time characteristics of a pulsed flow of large rare gas clusters*

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(Received 14 January 2003; revised manuscript received 17 February 2003)

The time characteristics of a flow of large rare gas atomic clusters formed by supersonic expansion of a gas into vacuum through conic nozzles are studied using the time resolving Rayleigh scattering method. The results show a two-region structure of the time resolved spectrum of the scattered light from the clusters. The average cluster size in the second region is much, even orders of magnitude, larger than that in the first one. Three designs of the valve-nozzle assembly have been used to investigate the origin of this finding. Finally, the potential application of the observation is proposed.

Keywords: atomic clusters, Rayleigh scattering, two-region structure
PACC: 3640, 5250

1. Introduction

The formation of clusters was first reported in 1956[1] and since then the characterization of the atomic cluster flow has been widely studied[2−6] because of the unique position that clusters hold as an intermediate state between molecules and solids. In recent years, the interaction of the femtosecond intense lasers with rare gas clusters attracts great interest for fundamental and practical reasons.[7−11] To make the study more realistic for cluster related experiments, the characterization of a cluster flow used is usually required. Many methods have been developed to estimate the cluster size and determine the size distribution in a cluster flow,[2−5], however, little has been done to characterize the time history of a pulsed cluster flow.

van der Waals force bonded rare gas clusters can be formed in the expansion of a gas through a nozzle into vacuum. The cooling associated with the supersonic adiabatic expansion causes the gas to supersaturate and the atoms will nucleate into clusters. Due to the complexity of the condensation process, sometimes the characteristics of individual cluster sources of similar design may be distinctly different.[6] Rayleigh scattering of a visible radiation has been used as a means to determine the onset of nucleation in cluster beams and the average cluster size.[6,12,13] The Rayleigh scattering signal, arising from the scattering of an incident radiation of intensity $I(\lambda)$ from N spherical particles can be written as

$$S_{\mathrm{R}} = \alpha I(\lambda) N \sigma, \tag{1}$$

where α, the detection efficiency, is determined by the detection geometry and the sensitivity of the photomultiplier used, and σ is the Rayleigh scattering cross section. Since it is impossible to determine α accurately, the size of the clusters can only be known relatively. However, the size calibration can be done by assuming that the cluster size is roughly 100 atoms when a scattered signal becomes detectable, which is a sign of onset of clustering.[8,14−16]

The number of scattering particles (clusters) is proportional to the mass flux of a flow which is proportional to the pressure P_0 of the stagnation gas. Assuming all the atoms in the flow are nucleated into

*Project supported by the National Natural Science Foundation of China (Grant No 29890210), the National Basic Research Special Foundation (Grant No G1999075200), Shanghai Foundation of Science & Technology (Grant No 0159nm022), and the Department of Physics, Shantou University, Shantou 515063.

clusters and the average number of atoms in a cluster is $\overline{n_c}$, the number of the scattering clusters is given by

$$N \propto \frac{P_0}{\overline{n_c}}. \tag{2}$$

The Rayleigh scattering cross section σ, for spherical clusters of radius r, is given by[13]

$$\sigma = \frac{128\pi^5 r^6}{3\lambda^4}\left(\frac{i^2-1}{i^2+2}\right), \tag{3}$$

where i is the refractive index of the medium and λ is the wavelength of the probe light radiation. If assume that the index of refraction is constant for different cluster sizes and the clusters are spherical, the radius r is proportional to $\bar{n}_c^{1/3}$, then for a specific wavelength, we obtain,

$$\sigma \propto \overline{n_c^2}. \tag{4}$$

Assuming that the cluster distribution is not too broad, then $\overline{n_c^2} \cong \overline{n_c}^2$. Combining the above equations, we can finally obtain the approximate dependence of the Rayleigh scattering signal S_R on the backing pressure P_0 and the average size $\overline{n_c}$ of the clusters,

$$S_R \propto \overline{n_c} P_0. \tag{5}$$

In this paper, we report observation of a novel two-plateau structure of the temporal profiles of krypton and xenon cluster flows using the Rayleigh scattering method. With three valve-conical nozzle devices, we have investigated the formation and decay processes of the clusters, and the mechanism of the formation of the second plateau is analysed.

2. Experimental

The experimental apparatus has been described in detail elsewhere.[14,15] In brief, a solenoid pulsed valve was used and the clusters were produced at room temperature in supersonic expansion of high backing pressure gases into vacuum through a 300μm conical nozzle (28mm long, 5° half opening angle). Three designs of the valve-nozzle assembly were employed of which two have with each a small room(38mm^3 and 15mm^3, respectively). The room is formed by a short channel connected with the sealing plug of the valve and ended at the nozzle throat. And, in the third design, the gas expanded into vacuum directly through the nozzle without a room. The size of the clusters was controlled by changing the gas backing pressure P_0. A 532nm light beam from a frequency-doubled Q-switched Nd:YAG laser (0.2mJ in a 10ns pulse duration) was focused by a lens onto the cluster jet at right angles about 3mm from the nozzle exit. The Rayleigh scattering signal was detected by a photomultiplier in the direction perpendicular to both the laser beam and the cluster jet. A timing unit was used to provide the variable time delay of the probe laser light with respect to the trigger time of the pulsed valve.

3. Results and discussion

The dependence of the intensity of the Rayleigh scattering signal S_R on the backing pressure P_0 was measured and Fig.1 gives a typical example. The scattering signal S_R varies as P_0^β, with β being 2.8, 3.0 and 3.0 for argon, krypton and xenon, respectively. According to Eq. (5) we obtain the average cluster size $\overline{n_c} \propto P_0^{1.8}$ for argon, and $\overline{n_c} \propto P_0^2$ for krypton and xenon. This experimental result is in good agreement with the scaling reported in the literature.[8,16]

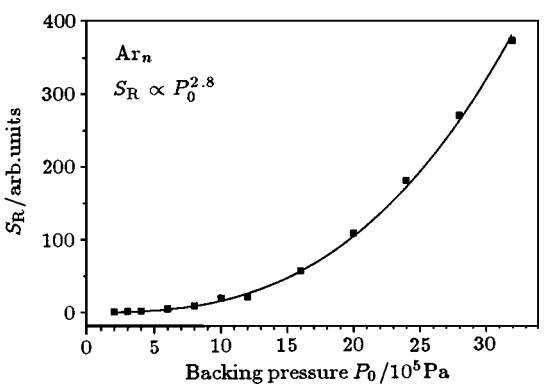


Fig.1. Rayleigh scattering signal S_R as a function of the gas backing pressure P_0 at room temperature for argon clusters.

The condensation of gas atoms into clusters is a complex process. After the gas expands into vacuum and condenses into clusters, the cluster size and the cluster density, which are of great importance for related studies, are all functions of time, so a good knowledge of the temporal profile of the cluster jet is necessary. For example, when a femtosecond intense laser interacts with gas clusters, we need to select an appropriate delay time between the trigger time of a pulsed valve and the laser incident time. For this reason, a time resolving Rayleigh scattering experiment was performed to study the time history of cluster formation and decay processes. The experimental results for argon, krypton and xenon are presented in Figs.2(a), (b) and (c), respectively. For argon, the process is similar to that in Ref.[6]. However, for krypton

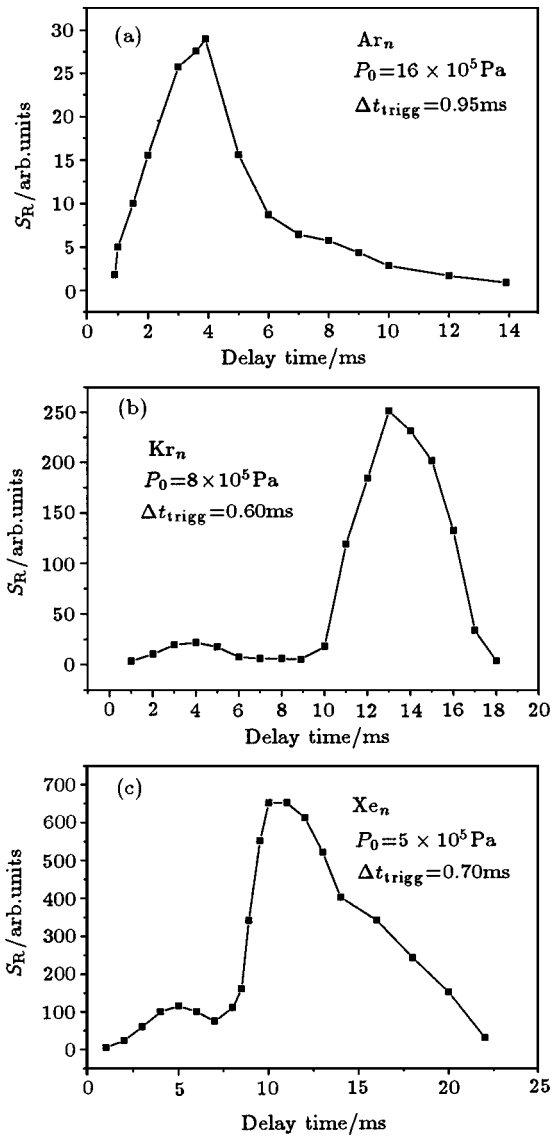


Fig.2. Time resolving Rayleigh scattering signals S_R as a function of the delay time of the probe laser light with respect to the valve's trigger time for (a) argon, (b) krypton and (c) xenon, respectively, when a 38mm^3 volume small room formed by a short channel is used.

and xenon gases, the experimental results are completely different. Unlike having a single plateau structure of the time resolved spectrum of the scattered light signal in the argon case, the time-resolved spectra of krypton and xenon have two plateaux. As indicated in Figs.2(b) and (c), after the left-hand plateau, which is similar to that in the case of argon, at about a 10ms delay time the signal increases abruptly to a much higher value, and then gradually falls down to zero. This striking phenomenon, to the best of our knowledge, had never been reported before. It did surprise us when noticing that the second plateau on the right-hand side is much more intense than the first one. Another interesting thing is that the second plateau lasts for about 10ms, especially for xenon. A careful analysis of the valve structure and the clustering process lets us believe that there are two factors that give rise to the second plateau. The first factor is related to the valve-nozzle structure. In the present valve-nozzle assembly design, the gas expands into vacuum through a 300μm nozzle after passing through a small room defined by a channel 2.5mm in diameter and 5.4mm in length. After the pulse valve was closed, residual gaseous atoms in the room would expand into vacuum slowly duo to the small throat of the nozzle, at the same time, the pressure in this small room decreased gradually. This is believed to lead to the long tail of the expansion, such as the situation seen in Fig.2(a). The duration time of the spectrum can be estimated as following. The atomic density n_a in the small room satisfies the following differential equation after the pulse valve is closed:

$$\frac{\mathrm{d}(n_a V)}{\mathrm{d}t} = -n_a v s. \tag{6}$$

Then, we can obtain:

$$n_a = n_0 \mathrm{e}^{-\frac{vs}{V}t} = n_0 \mathrm{e}^{-\frac{t}{\tau}}, \tag{7}$$

where n_0 is the atomic density of the stagnation gas, V=38mm^3 is the total volume of the small room, $v = \sqrt{\dfrac{2kT}{m}}$ is the flow velocity of the atoms, s is the area of the nozzle throat, and $\tau = V/vs$ is the residence time of the atoms in the small room. For argon, krypton and xenon, the residence times τ are 1.6, 2.2 and 2.8ms, respectively. These data can qualitatively illustrate the presence of the long tail of the Rayleigh scattering spectra of argon clusters and the first plateaux of krypton and xenon clusters as well.

The second factor is considered to be the bounce of the valve plug from the rubber O-ring when it is forced to close the valve again under the force from a spring. Due to the reopening of the valve, some gas expands into vacuum following the end of the forward main gas pulse. Compared with the forward opening, the gaseous atoms leaked are very limited. This is confirmed by a separate experiment in which the interaction of femtosecond intense laser pulses with the clusters generated by this valve was performed. From Eq. (5) we can see that the intensity of the Rayleigh scattering signal is determined by the average cluster size and the stagnation atomic density before clustering. Now, since the atomic density in the second plateau is much rarer than that in the first plateau, indicating that the cluster size in the second plateau

should be much larger than that in the first one. Then, there must be a factor that benefits the production of large clusters. When the valve reopens due to the bounce of the valve plug and some gaseous atoms leak into the short channel, those atoms and small clusters remained in the channel will act as the condensation nuclei for the leaked gas, consequently, larger clusters are formed. For krypton and xenon, this is the case. For argon, however, because the number of atoms leaked is limited and, especially, the condensation parameter of argon is small (1650) compared with krypton (2890) and xenon (5500), the clustering process would not be as violent as for krypton and xenon. As a result, a supersaturation state of the argon gas has not been reached.

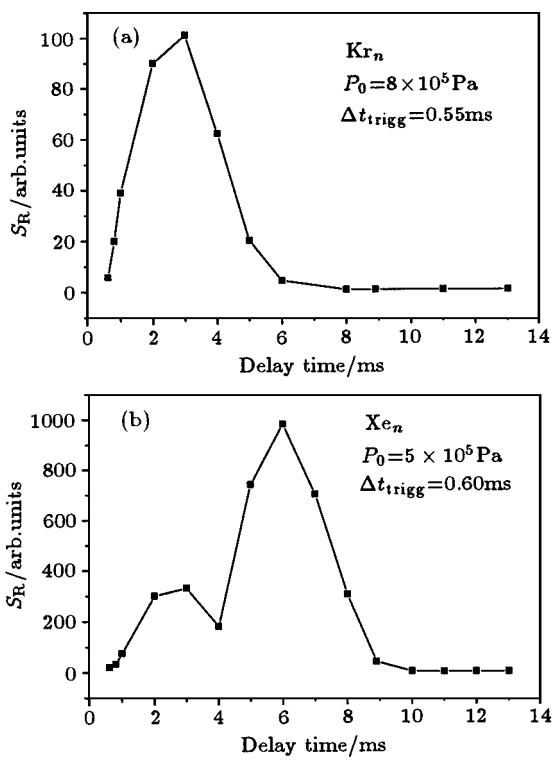


Fig.3. Time resolving Rayleigh scattering signals S_R as a function of the delay time for (a) krypton and (b) xenon when the volume of a small room is reduced to 15mm^3.

In order to verify the above judgement, two other valve-nozzle assemblies are designed. One is with a 15mm^3 room by shortening the channel length and the other is without a channel. In the later case, the gas atoms jet into vacuum directly through the nozzle when the valve is fired. Fig.3 and Fig.4 show the temporal profiles of the krypton and xenon cluster flows obtained by using these two devices. From Fig.3, when a volume reduced room is used, only xenon gas gives a second plateau of clustering. When the room of the volume is reduced, the residence time for krypton clusters is only one third of that in Fig.2 accordingly. That xenon is much more easy to form clusters compared with krypton makes it possible to create the second plateau even a reduced room is employed. The creation of the second plateau results in a longer duration time of the xenon spectrum with respect to the argon spectrum as shown in Fig.3.

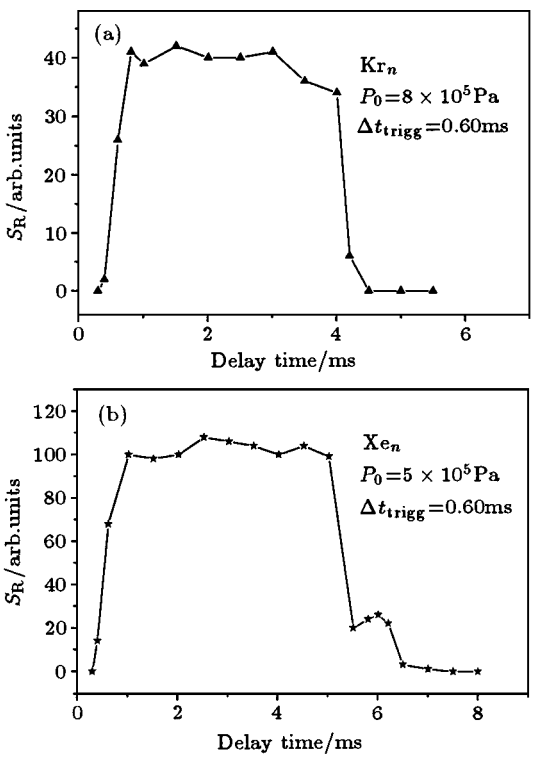


Fig.4. Time resolving Rayleigh scattering signals S_R as a function of the delay time for (a) krypton and (b) xenon without a room.

Figure 4 shows the Rayleigh scattering temporal profiles for krypton and xenon when the valve-nozzle assembly is without a small room. For krypton, just like the situation in Fig.3, we cannot observe the double plateau structure. For xenon, although the second plateau is not so strong as in Fig.3, it does exist and is very well reproduced. We propose that the second small plateau is generated by the leaked gas atoms due to the bounce of the plug. Because of the limited reopening time and the limited atom number, the second plateau is very weak in intensity and very short in duration time. Fig.4 also shows that the ascending and descending parts of the spectra are all very steep because there is not a room to slow down the gas expansion. The onset and the end of clustering are just associated with the opening and closing of the valve, in sharp contrast with the curves in Fig.2 and Fig.3.

For the spectra in Fig.2 to Fig.4, there are two things that may need to be discussed. The first is the duration time of the scattering signal. Depending on the time property of the pulsed valve used, for a 0.6ms wide trigger duration time, the real opening time of the valve is about 4ms. Actually, the real opening time of the valve changes with the variation of the gas backing pressure. Fig.5 shows the change of the duration time of the cluster flow with the trigger duration time. When the trigger duration time is changed from 0.6 to 0.7ms, the real opening time is changed from 4 to 6ms. This explains why the first plateau lasts for more than 4ms for only a 0.6ms trigger duration time. The second thing that should be concerned is the starting time of the second plateau. Fig.4(b) indicates that the second plateau arises closely following the end of the first plateau, and only lasts for about 1ms. Considering that the opening and closing of the valve will not be changed with the change of the volume of the small room, then, how to explain the several milliseconds delay of the starting of the second plateau after the end of the first plateau in Fig.2(b)? When the valve is reopened and some gaseous atoms leak into the room, clustering occurs in the room and the expansion of the cluster gas into vacuum through the nozzle takes place simultaneously. With the advance of the process, larger clusters are formed, and the Rayleigh scatter signal begins to increase. Consequently, the second plateau appears. For these large size clusters, their velocity of movement is not as large as the atoms and the small size clusters. This would lengthen their residence time in the room and result in the broadening of the second plateau.

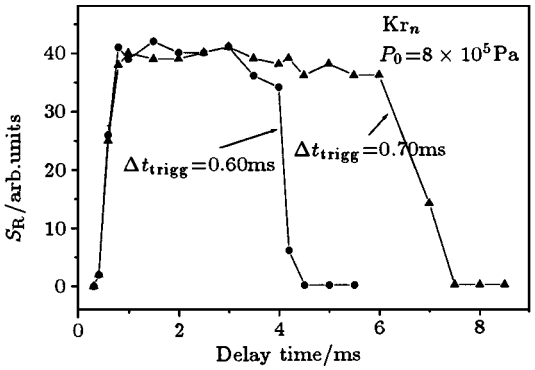


Fig.5. Time resolving Rayleigh scattering signals S_R as a function of the delay time for krypton for different trigger duration times.

This two-plateau structure of the scattered light from the clusters might provide a promising way to design a time interval adjustable double-trigger valve. By altering the time duration of the sequential double-trigger pulses and their interval deliberately, the generation of very large clusters with a moderate average density would be expected.

4. Conclusion

Using a Rayleigh scattering method, we have measured the correlation between the size of rare gas clusters and the gas backing pressure. Especially, the temporal profiles of the formation and decay processes of the clusters are investigated using three designs of the valve-nozzle assembly. A novel two-plateau structure of the temporal profiles for krypton and xenon is observed. The mechanism of their formation has been discussed. By a modification to the valve-nozzle structure, it may be promising to produce a jet of very large size clusters with a moderate average density.

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