Growth of deuterium clusters in a gas jet and ion energy spectrum of clusters in ultra-short laser field*

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Large deuterium clusters are generated using a cryogenic pulse valve with a cone nozzle (21 mm long, 4° open angle). Rayleigh scattering experiment is carried out to obtain the scaling relation between scattering signal S_R and backing pressure P_0. A method using the Coulomb explosion model is proposed to verify that the clusters continue to grow after their leaving the nozzle. Our experiments suggest a tentatively optimized position for laser cluster interaction.

Keywords: deuterium cluster, Rayleigh scattering, Coulomb explosion, ion energy spectrum

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1. Introduction

For tens of years, the characterization of atomic clusters has been widely investigated because of the unique position that clusters hold as an intermediate state between gas and solid.[1−7] Much effort has been made to study the ways to produce atomic clusters. For different kinds of atomic clusters, free gas expansion, laser vaporization as well as magnetron sputtering are usually used.[8−11] There are two schemes of cluster generation within the framework of the free gas expansion method. In the first case, a gaseous flux passes through a nozzle and expands into a vacuum. In the second case, a vapour is formed in a chamber which is held at high temperature before it expands into a vacuum.

In recent years, the interaction of ultra-intense ultra-short laser pulse (UULP) with atomic clusters has become an active area of research. Many results, including theoretical arguments and experimental data, have shown that laser clusters interaction is extremely energetic due to the high local density of clusters.[12−17] Energy deposition rates of 240 mW per atom have been measured.[18] A number of experiments have been performed to examine the high order harmonic generation, neutron, high energy ions jet, and fast electron and X-ray emission from van der Waals bonded clusters in UULP fields.[14,19−23] In these studies, large cluster formation and characterization are of critical importance.[7,24−28] Rayleigh scattering technique is usually employed to determine the average size of the cluster beam. However, little work deals with the growth of the clusters.

In this paper, large deuterium cluster generation is experimentally demonstrated using a cryogenic pulsed valve at 80 K. The interaction experiments are performed on the SILEX-I 20 TW laser facility and energetic ions are measured. According to the Coulomb explosion model, the observed deuterium ions indicate the growth of the clusters after their leaving the nozzle.

2. Generation of deuterium clusters

It is known that the size of the gas atomic cluster produced by free expansion depends on the atomic species, gas backing pressure, initial temperature and the geometry of the jet nozzle. Hagena and Obert,[3]

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and Örmer *et al.*[29] found an empirical parameter Γ^* which can predict the onset of clustering in gas jets and the size variation of the clusters. From Γ^*, one can facilitate the production of large clusters by increasing the backing pressure. Unlike inert gases, light gases such as H_2, D_2 and He interact weakly at room temperature. These gases must be cooled at high backing pressure for clustering.

We used a cryogenic pulse valve with a cone nozzle to generate large deuterium clusters. The cone nozzle is 21-mm long and has a 0.5-mm orifice. A time resolving Rayleigh scattering experiment was performed using the second harmonic (532 nm) of a Q-switched Nd:YAG laser. Figure 1 shows the experimental setup. The laser beam was focused to intersect the cluster jet at right angle below the cone nozzle exit. The scattered light was collected by a photomultiplier perpendicular both to the laser beam and to the cluster jet. A photon trap placed opposite to the laser beam can dramatically reduce the noise signal.

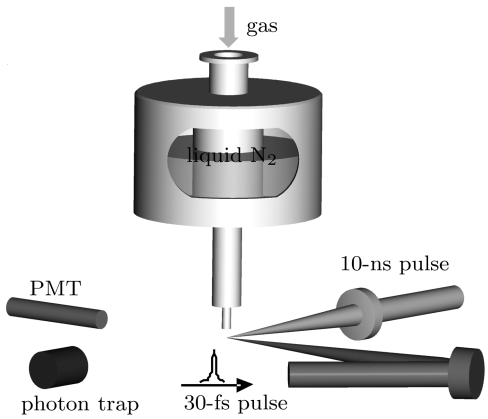


Fig. 1. Experimental setup for Rayleigh scattering measurement and UULP cluster interaction.

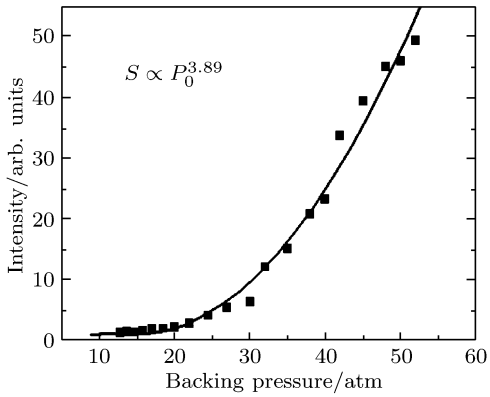


Fig. 2. Rayleigh scattering intensity versus backing pressure (1 atm = 1.01325×10^5 Pa).

According to the classical Rayleigh scattering theory and on the assumption that all atoms of the stagnation gas (backing pressure P_0) are condensed into clusters in the supersonic expansion, we have $S_{\rm R} \propto P_0 N_C$, where $S_{\rm R}$ is the intensity of the Rayleigh scattered light and $N_{\rm C}$ is the average size (number of atoms per clusters) of the clusters. Figure 2 shows the results of D_2 at 80 K. The signal intensity of Rayleigh scattered light $S_{\rm R}$ increases rapidly with P_0 in a range from 12 atm (1 atm = 1.01325×10^5 Pa) to 52 atm. Good power variance of $S_{\rm R}$ with P_0 indicates $S_{\rm R} \propto P_0^{3.89}$. Thus we have $N_C \propto P_0^{2.89}$. According to the assumption that the threshold for observing optical scattering from a cluster jet typically occurs at $N_{\rm C} = 100$, we can estimate cluster sizes at different backing pressures.[29]

3. Theory model

It is known that the process of cluster formation is a result of the attachment of free atoms to clusters, or a result of two neutral clusters in collisions. Therefore, the cluster will continue growing until the density of the expanding gas becomes low and collisions between clusters finish. As mentioned in Section 2, Rayleigh scattering experiment can give us only the average size of the cluster beam. To explore the growth of the cluster after leaving the nozzle, one can turn to the energetic ions emitted from the intense laser interaction with clusters.

Under the irradiation of the intense laser pulse, the deuterium clusters could be stripped of almost all their electrons. These electrons themselves will create an even, strong electric field and accelerate ions to a very high speed. Meanwhile, the positive charge density in the clusters is very high. So the deuterium clusters will explode and release large electrostatic energy transferring to ion kinetic energy. According to the Coulomb explosion model,[26,27] the ion energy is dependent on the size of cluster, $E \propto \langle q \rangle e^2 r^2$, where $\langle q \rangle$ is the average charge state in the cluster, e is the electron charge and r is the initial radius of the cluster. In general, the average charge state in the cluster is determined by the laser intensity. Given the laser intensity, the ion energy varies with the square of the radius of the cluster.

4. Growth of the cluster after leaving the nozzle

We performed the laser cluster interaction experiments on the SILEX-I Ti:sappire laser facility based on the chirped-pulse amplification (CPA) technique, operating at a central wavelength of 800 nm, at the Laser Fusion Research Center (LFRC), China Academy of Engineering Physics (CAEP). As shown in Fig. 1, the 30-fs laser pulse delivering 400-mJ energy was focused into the gas jet of deuterium clusters by an f/10 off-axis parabolic mirror. A spot of 20-μm diameter in full width at half maximum (FWHM) was obtained. The contrast ratio of the laser system is typically greater than 10^{-6} on the nanosecond scale.

The energetic deuterium ions expelled from the clusters jet propagated along an 80-cm flight tube were detected by an ion time of flight (TOF) spectrometer with dual micro-channel plate (DMCP). The front plate of the DMCP was held at –1.7 kV and the back one was grounded. The signal was recorded by a digital oscilloscope. To obtain the flight time of the energetic ions, we detected the scattered laser signal and found that it was synchronous with the X-ray signal. The typical raw signal is shown in Fig. 3. The ion energies are determined by flight time measurement by $E = (1/2)m(l/t)^2$, where m is the mass of the ion, l is the length of the flight tube and t is the flight time. The ion energy distribution can be obtained from the flight spectrum via $f(E) = f(t) \times (\mathrm{d}E/\mathrm{d}t)^{-1}$.

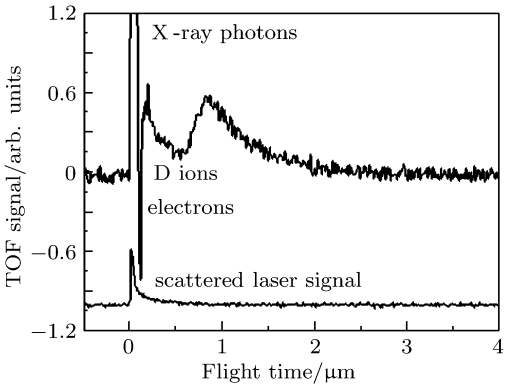


Fig. 3. Typical flight time spectrum of ions.

In our experiments, we changed the position of the laser focus while keeping other conditions unchanged. As shown in Fig. 4, the position of the laser focus under the nozzle has a remarkable influence on the ion energy spectrum. The intensity of the energetic ions increases with the laser focus deviating from the nozzle, at 0.6, 1.2 and 3 mm. These data indicate that the clusters continue to grow after their leaving the nozzle. As mentioned above, the process of cluster formation is the result of attachment of free atoms to clusters. This process does not finish as the clusters leave the nozzle because the density of the expanding gas is still high enough for free atom cluster collisions. Therefore, the clusters at 3 mm are larger than those at 1 mm.

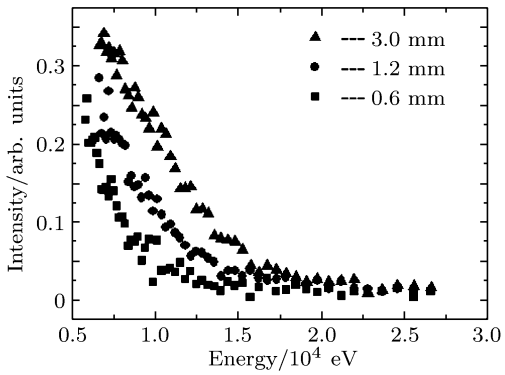


Fig. 4. Ion energy distributions from D_2 clusters obtained with different focus position below the nozzle.

On the other hand, the size of the cluster decreases rapidly if the density of the expanding gas turns lower. Cluster evapouration, instead of attachment of free atoms to clusters dominates once the gas intensity becomes low enough. In our experiments, few energetic ions were observed when the laser focus was moved to 5 mm under the nozzle.

5. Conclusion

We have shown that large deuterium clusters generate by expanding high pressure gas through a cone nozzle. The growth of the clusters after their leaving the nozzle is verified by ion energy spectra variation, which is helpful for intense laser cluster interaction experiment. Next, we will go on to explore the cluster fusion because this fusion source is of interest as an ultra-fast pulsed neutron emitter for time resolved material damage studies.

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