Interaction of intense laser pulses with atomic clusters at different gas densities

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The interaction of intense femtosecond laser pulses with rare gas clusters was studied experimentally, the time-of-flight spectra of ions from exploding clusters at different gas densities have been measured. It is found that while the relative components of ions in low and high energy of the ion energy spectrum decrease with the increase of the gas density, the average ion energies are the same for different gas densities, which indicates that the effect of gas density on laser-cluster interaction is not important under our experimental conditions.

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The interaction of high-intensity femtosecond laser pulses with gases of atomic clusters has been a topic of considerable interest in the past few years. Rare gas atomic clusters are aggregates of atoms bonded together by van der Waals forces. Compared with gaseous and solid targets, a gas of large clusters presents a unique environment for laser-matter interaction. The clusters can efficiently absorb laser light due to the local solid density of individual clusters. The laser-heated clusters explode, producing energetic particles[1,2] and keV X-ray radiation[3]. The observation of neutrons generation[4] from the interaction of intense laser pulses with a dense jet of deuterium clusters was also reported.

In all the laser-cluster interaction experiments, the cluster targets were formed by adiabatic expansion into vacuum of a high-pressure atomic gas puff produced by a pulsed valve with a nozzle. For some possible applications of the laser-cluster interactions, such as the intense X-ray generation and the neutron sources, the laser focus should be located very close to the nozzle output of a high-density cluster jet. Because the nozzle parameters and experimental conditions are different for different research groups, the gas densities of clusters are different, which may influence the experimental results and affect the comparability of these results. For example, Zweiback *et al.*[5] found that the neutron yielded from femtosecond laser-driven explosions of deuterium clusters depends strongly on the distance between gas jet nozzle and the laser propagation axis as well as the laser focal position with respect to the center of the gas jet. Krainov *et al.*[6] suggested that for a high density cluster source, a description of the evolution of the laser-heated cluster should include both electrons inside and outside the cluster. Here, the electrons inside the cluster are the ionized electrons within the cluster, and the outside electrons are those electrons that escaped from the cluster and other clusters nearby. The generation of these outer ionized electrons is mainly determined by laser parameters and the gas density. So, the influence of gas density should also be taken into account in the laser-cluster interaction. Smith *et al.*[7] showed that by holding $P_0T_0^{-1/2}$ constant when changing cluster size (P_0 and T_0 are the gas backing pressure and temperature, respectively), the mean gas density could also hold constant. This allows detailed interaction studies as a function of mean cluster size to be made without changing other density-dependent effects. But till now, in what respect, to what degree, the gas density would affect the laser-cluster interaction is still unknown. In this letter, we present experimental results from the interaction of laser pulse with rare gas clusters of different gas densities. By changing the distance between the laser focus and the orifice of the nozzle, we have measured the time-of-flight (TOF) spectra and energy spectra of ions from laser irradiated clusters at different gas densities. It is found that for high gas density, the fraction of ions at the low and high energy ends of the ion energy spectrum decreases due to the collision between ions from nearby clusters, however, the average ion energy seems not sensitive to the gas density.

In the experiment, clusters were produced by supersonic expansion of a gas into vacuum through a conical nozzle (700 μm in diameter, 5° opening angle, and 28 mm in length). The cluster size was controlled by changing the gas backing pressure P_0. The average cluster sizes were estimated according to Hagena's empirical scaling parameter[8,9]. The laser used for the experiments was a chirped-pulse-amplified Ti:sapphire laser system operating at a wavelength of 790 nm, producing about 30 mJ in 60-fs pulse with a 10-Hz repetition rate. Using an off-axis parabolic mirror with a focal length of 20 cm, the laser was focused downstream of the nozzle, and the laser intensity at the focal spot was $\sim 2\times 10^{16}$ W/cm^2 under this focal configuration. Because of the diverging cone of the gas jet, the average density of the gas at the laser focus decreased exponentially with the increase of the nozzle-focus distance[10,11]. By changing the position of the nozzle axially, the distance between the nozzle and the laser focal spot could be varied from $d = 2-12$ mm to alter the gas density at the focal spot which was located at the axis of the nozzle while the average cluster size remained unchanged. The electrons and ions expelled from the clusters with velocities perpendicular to both the cluster jet and the laser beam travelled along a 120-cm-long field-free flight tube after passing through a skimmer and were detected by a dual microchannel plate (DMCP). The front plate of the DMCP was held at -1.5 keV and the back was grounded. A grounded metal mesh was placed in 5 mm before the front plate of the DMCP ensuring that the flight-tube was field free. The signal from the DMCP was recorded using a 500-MHz, 1-GS/s digital oscilloscope (LeCroy 9350A). The ion energy was determined by TOF measurements through $E = \frac{1}{2}m(l/t)^2$ (m is the mass of ion, l is the length of the flight tube, and t is the flight time) and the energy distribution $f(E)$ of the ions was derived from the TOF spectrum $f(t)$ via $f(E) = f(t) \times (\mathrm{d}E/\mathrm{d}t)^{-1}$. The average ion energy $\bar{E}$ was calculated from $f(E)$ by $\bar{E} = \int Ef(E)\,\mathrm{d}E / \int F(E)\mathrm{d}E$.

Figure 1 shows the TOF spectra $f(t)$ which are an average of 200 laser shots and the corresponding ion energy distributions $f(E)$ for argon clusters at 10-bar gas backing pressure, and the distance between the nozzle and the laser focus are 4 and 8 mm, respectively. The first downward sharp peak in Fig. 1(a) is due to the electrons produced in the interaction and the photoelectrons created by X-ray emission. The time for the photons to pass the flight tube can be ignored, so we use the beginning of the sharp peak as the origin of the flight time of the ions. A long time scale broad feature followed is the signal of the argon ions.

From Fig. 1(b) we can see that, because of the difference of gas densities at the two positions, the ion energy distributions differ a lot in amplitude, i.e., there is much difference for the ion yielded in the two cases, but the ion energy distributions are similar. To make it more clear, Fig. 2 shows the ratio of the two ion-energy distributions. If the influence of the atomic density could be ignored, the ratio should be approximately equal to the ratio of the densities at the two positions and be a constant for different ion energies. It can be seen that the ratio at the two ends of the ion energy distribution is less than that of the medium, energy indicating that for high gas density, the relative fractions of ions at the two ends of the ion energy distribution are less than those of low gas density. It is evident that after the explosion of the clusters, the collision between ions is much more violent for higher gas density. During the collisions, the low energy ions obtain energy and those ions with high energy lose energy, resulting in the decrease of the number of these ions. Then, it is reasonable to speculate that the difference of the ion energy distribution for different gas densities is mainly caused by the collision between the ions after the clusters explode, and does not demonstrate any significant influence of the gas density upon the laser-cluster interaction.

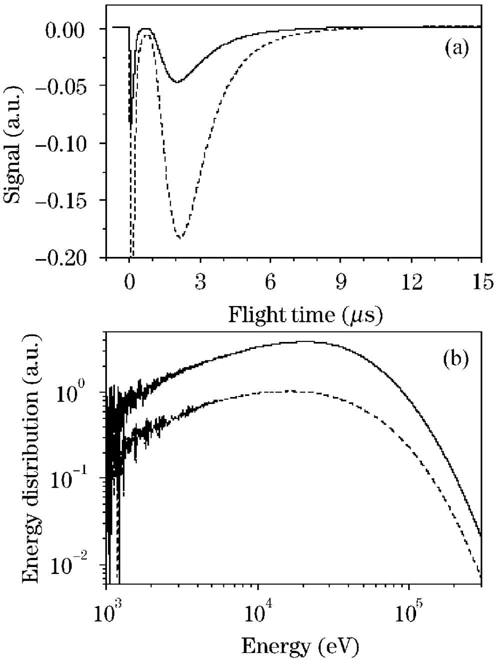

Fig. 1. Time-of-flight spectra (a) and energy spectra of ions from laser ($\sim 2 \times 10^{16}$ W/cm^2) irradiated argon clusters at 10-bar gas backing pressure, the distances between the nozzle output and the laser focus are 4 (solid line) and 8 mm (dash line), respectively.

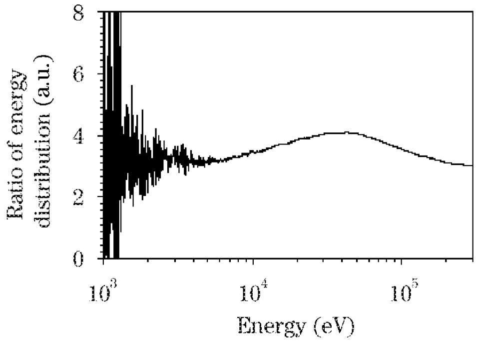

Fig. 2. Ratio of the ion energy spectrum.

We have also measured the average ion energy as a function of a variety of laser focal positions at different gas backing pressures. Figure 3 shows the average ion energy for different nozzle-focus distances at the gas backing pressures of 10 and 4 bar, respectively. For one cluster, if the electrons escaping from nearby clusters have some influence on its interaction with laser field, the direct result is that the energy absorption of the cluster to the laser field is affected, and furthermore, the ion energy and the energy distribution is affected. Because the collision between ions does not influence the total energy of the ions, the average ion energy reflects the influence of the atomic density. We can see clearly from Fig. 3 that for different nozzle-focus distances, i.e., for different gas densities, the average ion energy is basically the same. The distinction for 10- and 4-bar gas backing pressures is just the decrease of the average ion energies from ~60 to ~30 keV. These results seem to suggest that in the case of high density cluster jet, the laser-cluster interaction can be considered as the interaction of laser pulse with a single cluster, and the influence of the gas density is not important.

For a laser heated cluster, the expansion time can be approximately written as $\tau \sim \frac{r_0}{v}(n_0/n_s)^{1/3}$, where r_0 is the initial radius of the cluster, n_0 is the initial density,

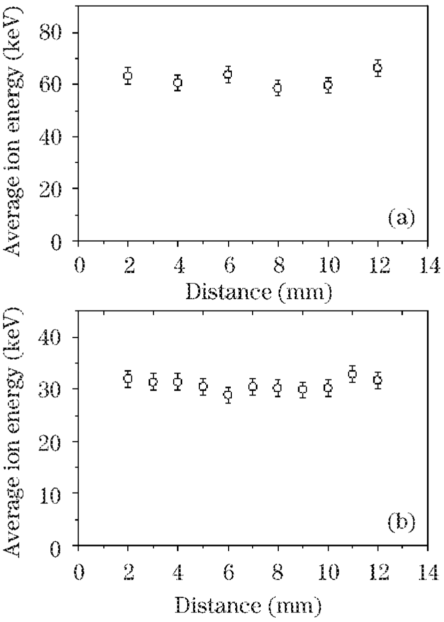

Fig. 3. Average energy of argon ions for different nozzle-focus distances at different gas backings of 10 (a) and 4 bar (b).

n_s is the density after expansion, and ν is the cluster expansion velocity. Using the average ion energy in Fig. 3, the expansion time is estimated to be about in the order of picosecond, much longer than the laser pulse duration time. It means that the laser pulse has passed long before the cluster has fully expanded. In this situation, the cluster can be taken as interacting with laser pulse singly. It is reasonable to believe that if we use a laser with long pulse duration, such as in the order of picosecond or longer, for high gas backing pressure, the effect of the gas density will emerge. In addition, the density of electrons outside is determined by both the gas density and the laser parameter, if the laser intensity increases, the electrons outside may be driven by laser field to enter into the cluster and influence its absorption to the laser energy similar to the heating mechanism proposed by Brunel[12].

There is another factor concerning gas density that should be considered during the laser-cluster interaction. Because of the diverging cone of the gas jet, increasing the nozzle-focus distance broadens the spatial extent of the gas through which the laser propagates. When the gas density is very high, part of the laser energy is absorbed or scattered before reaching its focus, and the focusing of the laser beam is affected simultaneously, at this moment, to some extent, the laser-cluster interaction will be influenced indirectly by the gas density.

In conclusion, we have measured the time-of-flight spectra of ions from exploding clusters in an intense laser field, and calculated the ion energy spectra and the average ion energy at different gas densities. The results show that under our experimental conditions (laser pulse width ~60 fs, peak intensity $\sim 2\times 10^{16}$ W/cm^2), the effect of the gas density on the laser-cluster interaction is not obvious, while the ion energy spectrum is affected due to the collision between ions after the cluster explodes, the average ion energy which truly reflects the absorption of clusters to laser energy is not influenced by the gas density.

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