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High-intensity femtosecond laser absorption by rare-gas clusters*

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The energy absorption efficiency of high-intensity ($\sim 10^{16}$W/cm^2) femtosecond laser pulses in a dense jet of large rare-gas clusters has been measured. Experimental results show that the energy absorption efficiency is strongly dependent on the cluster size and can be higher than 90%. The measurement of the ion energy indicates that the average ion energies of argon and xenon can be as high as 90 and 100keV, respectively. The dependence of the average energy of the ions on the cluster size is also measured. At comparatively low gas backing pressure, the average ion energies of argon and xenon increase with increasing gas backing pressure. The average ion energy of argon becomes saturated gradually with further increase of the gas backing pressure. For xenon, the average ion energy drops a little after the gas backing pressure exceeds 9 bar (3.2×10^5 atoms/cluster). The result showing the existence of a maximum average ion energy has been interpreted within the framework of the microplasma sphere model.

Keywords: atomic clusters, femtosecond intense laser pulse, high-energy ions
PACC: 3640, 5250, 3380

1. Introduction

In recent years, the interaction of high-intensity femtosecond laser pulses with matter has become an important field. In these studies, the interaction of laser pulses with the target of rare-gas clusters is of considerable interest.[1−9] Rare-gas clusters are aggregates of atoms and are bonded together by van der Waals force. Compared with gaseous and solid targets, a gas of large clusters presents a unique environment for laser–matter interaction. Though the average density of a gas containing large size clusters is low, the local density in a cluster is nearly as high as the solid. So, the cluster will efficiently absorb laser light due to rapid inverse bremsstrahlung heating. And, owing to the lack of conductive dissipation of heat into a bulk substrate, almost all the absorbed laser energy by the clusters will transfer to the kinetic energy of particles and the x-rays emitted during the explosion of the clusters. Over the past few years, energetic ions with energy up to 1MeV,[3] electrons in the keV range,[5] and high charge states of ions[6] produced in the interaction of intense laser pulses with a low density rare-gas cluster beam have been observed. The observation of keV x-ray emission[7] and neutron generation[8] from the interaction of intense laser pulses with a dense jet of cluster were also reported.

All these experimental results indicate that the energy absorption efficiency of the laser light by the plume of rare-gas clusters must be very high. Using 2ps laser pulses, Ditmire *et al* have measured the energy absorption efficiency of rare-gas clusters directly.[9] They found that with a peak laser intensity of 7×10^{16}W/cm^2, the energy absorption efficiency could reach as high as 95% for xenon clusters. Using 150fs laser pulses and at the peak intensity of 1×10^{15}W/cm^2, Lin *et al* have also measured the energy absorption efficiency of rare-gas clusters,[10] showing that the energy absorption efficiencies turned

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out to be only 45% and 33% for xenon and argon clusters, respectively.

A dense jet of clusters has been demonstrated to have very high energy absorption efficiency to an intense picosecond laser light and the maximum ion energy of 400keV was reported by Ditmire *et al*,[9], however, no information concerning the energy distribution of the ions has been given. The limited maximum ion energy was attributed to the relatively short flight path used in the time-of-flight(TOF) measurement in which the fast ions were obscured by the presence of the photoelectron spikes. In this paper, we report the laser energy absorption measurement of intense femtosecond laser pulses in a dense jet of rare-gas clusters. Compared with the 2ps laser pulses used in the experiment of Ditmire, we find that a shorter laser pulse width as 60fs in our case seems more beneficial to the energy absorption of the laser pulses. We have also measured the energy of ions produced in the laser interaction with jets of clusters. We find that the average ion energies increase with increasing the gas backing pressure for argon. For xenon clusters, however, there is a maximum average energy of the ions, and the average ion energy decreases as the cluster size is further increased.

2. Experimental

In our experiment (illustrated in Fig. 1), clusters are produced by supersonic expansion of a gas into vacuum through a conical nozzle. The aperture of the nozzle is 700μm in diameter and the opening angle is 5°. The estimation of the average cluster size has been made based on our previous work[11] while the Hagena's empirical scaling parameter is also taken into account.[12,13] The presence of large clusters in the gas jet is confirmed by Rayleigh scattering experiment. Our Rayleigh scattering measurement indicates that the average cluster size varies as (backing pressure)$^{\alpha}$ with $\alpha = 2$ for krypton and xenon and $\alpha = 1.8$ for argon. In our experiment, the average cluster size was controlled by changing the gas backing pressure.

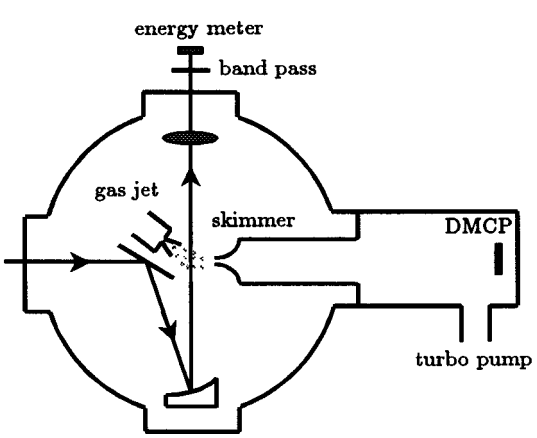


Fig.1. Experimental set-up for measuring laser energy absorption efficiency by a jet of rare-gas clusters and ion energy of exploding clusters irradiated by an intense laser pulse (not in scale).

The laser used is a chirped-pulse-amplification Ti:sapphire laser which delivers 60fs pulses at the wavelength of 790nm with a 10Hz repetition rate. In the experiments, the laser pulse energy used is up to 50mJ although it can reach a much higher value. The laser light is focused by an off-axis parabolic mirror with focal length of 20cm and the focal spot is 2mm downstream of the nozzle. With this focal configuration, the focal spot size is found to be about 50μm in diameter. The transmitted light was collected by an 8cm diameter lens to assure that any light refracted by the plasma at the focus was collected. The energy of the transmitted light was measured with an energy meter (Model Rj-7100, Laser Precision). An 800nm bandpass filter was put in front of the meter to assure that only the laser light was measured. The laser energies before and after the absorption by the clusters were measured when the pulsed valve was switched off and on. Then the energy absorbed by the cluster jet is the difference of these two energies. To overcome the influence of random fluctuation of the laser energy, every datum was the average of 50 shots. The ions expelled from the clusters with velocities perpendicular to both the cluster jet and the laser beam propagate along a 120cm field-free flight tube after a skimmer and are detected by a dual microchannel plate detector (DMCP). The front plate of the DMCP was held at −1.5kV and the back was grounded. A grounded metal mesh was placed 5mm before the DMCP to ensure that the flight-tube is field free. The signal from the DMCP is recorded by a 500MHz, 1GS/s digital oscilloscope (LeCroy Model 9350AL). The ion energies are determined by time-of-flight (TOF) measurements through $E = \frac{1}{2}m(l/t)^2$ (m is the ion mass, l is the length of the flight tube and t is the flight time) and the energy distribution $f(E)$ of the ions can be obtained by converting the TOF spectrum $f(t)$ via $f(E) = f(t) \times (\mathrm{d}E/\mathrm{d}t)^{-1}$. The average ion energy was computed from the energy distribution function $f(E)$:

$$\overline{E} = \frac{\int Ef(E)\mathrm{d}E}{\int f(E)\mathrm{d}E}. \qquad (1)$$

Due to the limited record length of the TOF spectrum, there is a low-energy cutoff, so, the average energy does not include ions with energies less than 500eV. Since most of the ions have energies greater than this value, this cut-off would not influence too much the average energy given here.

3. Results and discussion

The fraction of laser energy absorbed in an argon cluster jet as a function of gas backing pressure is shown in Fig. 2(a). The peak laser intensity is 2×10^{16}W/cm^2. Different from the data in Refs. [9] and [10], the energy absorption has reached 13% already at 2 bar gas backing pressure, and with increasing the gas backing pressure, the absorption efficiency rises to higher than 85%. Measurement of energy absorption of xenon was also performed, and the result is shown in Fig. 2(b). Compared with argon, xenon clusters exhibit higher laser absorption efficiency. At 1 bar gas backing pressure, the energy absorption efficiency is 26%, with increasing backing pressure, the energy absorption efficiency increases quickly, and reaches 80% at 7 bar. With further increase of backing pressure, the energy absorption efficiency increases slowly to 90% and gradually becomes saturated. In order to compare the difference of energy absorption efficiency of a gas of clusters and a gas of atoms, the energy absorption efficiency of a gas of He atoms was measured. At room temperature, He atoms in an expanding jet backed by a modest pressure will not nucleate and no clustering will take place. Shown in Fig. 2(c), as expected, the deposition of laser energy in the plume of He gas is less than 10% though it appears slightly higher than that in Refs. [9] and [10]. It is clear that the presence of the clusters plays a significant role in energy absorbing from the laser pulse.

Compared with that reported by Ditmire *et al*,[9] the peak laser intensity used in our experiment is a little lower, but the energy absorption efficiency at the same backing pressure is higher. This is attributed to two reasons. The first is the influence of the laser pulse width. Due to the rapid expansion of the clusters in the leading edge of the laser pulse, the atomic density in the cluster at the trailing edge of the laser pulse drops, consequently, the shorter laser pulse will be favourable to the absorption of the energy by the cluster gas plume. The second is the influence of the nozzle. The nozzle used in our experiment has a relatively large aperture diameter and a small opening angle. These features favour the production of larger clusters. In our experiment the cluster sizes are estimated to be considerably larger than theirs at the same gas backing pressure. In this paper, the average size of Xe clusters at P_0=1 bar is estimated to be about 4×10^3 atoms per cluster ($\sim$ 7nm in diameter), and for argon clusters, at P_0=2 bar for example, the average size is estimated to be about 1×10^3 atoms per cluster ($\sim$ 4nm in diameter). The cluster sizes at other gas backing pressures are deduced from the relations mentioned before.

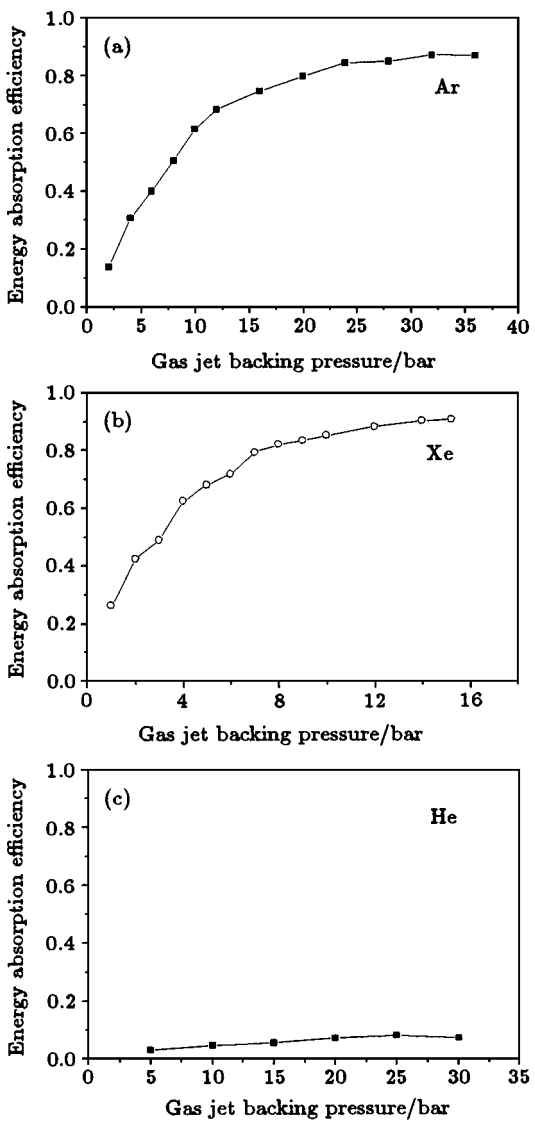


Fig.2. The energy absorption efficiency of a gas jet as a function of gas jet backing pressure with a peak laser intensity of 2×10^{16}W/cm^2 (Each point is the average of 50 shots.) (a) argon gas jet. (b) xenon gas jet. (c) helium gas jet.

It is expected that most of the laser energy deposited to the gas jet of clusters must be transferred to the kinetic energy of the ions after the explosion of clusters. To confirm it, we examined the energy of ions

using ion TOF measurement. Fig. 3(a), which represents the average of 200 laser shots, is a measured TOF spectrum of argon ions at 6 bar gas backing pressures (7×10^3 atoms/cluster and 7nm in diameter) irradiated by a peak intensity of 2×10^{16}W/cm^2 of laser pulses. The fast sharp peak in Fig. 3(a) is due to the photoelectrons from the detector caused by UV radiation emitted and by the following fast electrons from the plasma created in the laser interaction with the cluster jet. The long time scale broad signal followed after this first downgoing sharp peak is the signal of the ions. Since the time to cover the flight tube can be ignored for photons, so, we use the beginning of the first sharp peak as the onset of the ion signal (zero time) to calculate the flight time and the energy of the ions. The corresponding energy spectrum of the ions is shown in Fig. 3(b). The maximum ion energy $E_{\max}$ is 500keV and the average energy of the ions is 50keV. A striking feature of the measurement is the remarkably high ion average energy. In reference [9], Ditmire *et al* have ever estimated the laser energy deposited to one atom. At the peak intensity of 7×10^{16}W/cm^2 (0.25J energy and 2ps pulse width) and the average atomic density of 5×10^{18}cm^{-3} in the gas jet, they estimated that up to 300keV of energy is deposited per atom. In our experiment, we have not measured the plasma length and then cannot calculate the plasma volume to estimate the energy deposited per atom. Because the absorption of the laser energy is mainly determined by the peak intensity of the laser pulse, and the peak laser intensity in our experiment is 2×10^{16}W/cm^2 (20mJ energy and 60fs pulse width), which is similar to that in Ref. [9], the energy deposited per atom in our experiment should be comparable to that in Ref. [9]. Consequently, it is not surprising to find an average ion energy as high as that given above.

Figure 2 indicates that the absorption efficiency of the laser energy increases with increasing the gas backing pressure. To compare with it, we have also measured the dependence of the average energy of the argon ions on the gas backing pressure. Fig. 4 shows the average ion energy as a function of the gas backing pressure. At 2 bar backing pressure, the average energy of the ions is 20keV. With increasing the gas backing pressure, the average ion energy increases rapidly, at 10 bar, it is 72keV already. With further increase of the backing pressure the average ion energy increases slowly and gradually becomes saturated. It shows that larger clusters will be favourable to absorption of the laser energy to some extent.

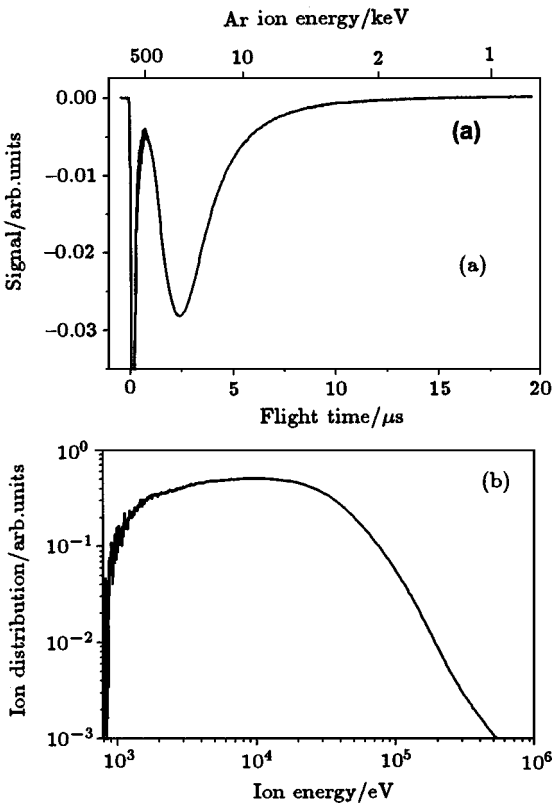


Fig.3. Time-of-flight spectrum (average of 200 shots) of ions (a) and the corresponding ion energy distribution (b) for argon clusters of 7×10^3 atoms (gas backing pressure 6 bar) irradiated by a peak intensity of 2×10^{16}W/cm^2. The maximum and average ion energies of 500keV and 50keV are obtained, respectively.

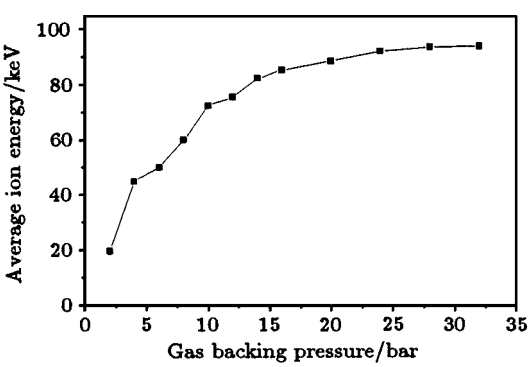


Fig.4. Dependence of the average energy of argon ions on the gas backing pressure. The laser peak intensity is 2×10^{16}W/cm^2.

As we have mentioned before that compared with argon clusters, at the same gas backing pressure, the plume of xenon clusters exhibits higher laser energy absorption efficiency. For different atomic species, the densities of atoms in a jet can be considered the same at the same backing pressure under otherwise identical conditions. So, higher energy absorption efficiency

means that more energy is deposited per atom. To verify it, we have also measured the TOF spectrum of xenon ions. Figure 5 is a measured TOF spectrum of xenon ions at 6 bar backing pressures (1.4×10^5 atoms/cluster and 23nm in diameter) irradiated by a peak intensity of 2×10^{16}W/cm^2 laser pulses. As can be seen from Fig. 5, the xenon ions exhibit very large kinetic energy. The maximum and the average ion energies are 1.1MeV and 80keV, respectively. Obviously, these values are much higher than those of the argon ions at the same backing pressure.

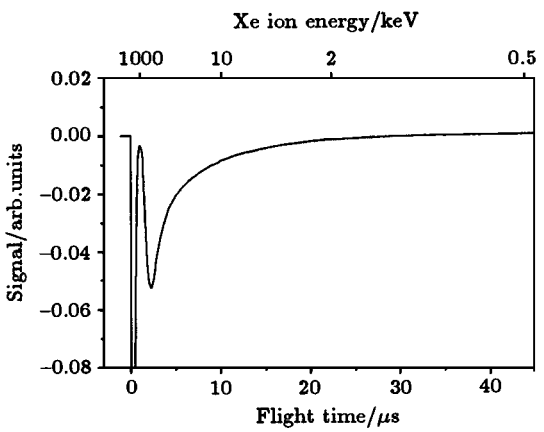


Fig.5. Time-of-flight spectrum (average of 200 shots) of ions for xenon clusters of 1.4×10^5 atoms (gas backing pressure 6bar) irradiated by a laser pulse at a peak intensity of 2×10^{16}W/cm^2.

Figure 6 shows the average energy of xenon ions as a function of the gas backing pressure. At comparatively low backing pressures, similar to argon ions, the average energy of the xenon ions increases quickly with increasing the backing pressure. But when the backing pressure is higher than 9 bar, the average ion energy has a trend of decreasing. This means that for the average ion energy, there is an optimum backing pressure. This character can be illustrated with Ditmire's microplasma sphere model.[14] In this model, a cluster is treated as a small plasma sphere when it is irradiated with an intense laser light. During the expansion process of the cluster, when the electron density within the cluster $n_e = 3n_{crit}$, where $n_{crit} = \pi c^2 m_e / e^2 \lambda^2$ is the critical electron density at which the laser frequency equals the plasma frequency, the electric field in the cluster will be much greater than that in vacuum, as a result, the energy absorption efficiency is enhanced and the electron temperature increases sharply. If this resonant point is near the peak of the laser pulse, its effect will be more remarkable. A small sized cluster expands very fast and the resonant point comes before the arrival of the peak of the laser pulse. For a too large cluster, the case is just the opposite. Therefore, it would be expected that there should exist a cluster size that fits the optimum absorption condition. Obviously, due to the existence of a wide size distribution of the large clusters in the interaction region, the effect of the resonance will be weakened. Figure 6, however, likely indicates that the influence of the resonance effect may still exist.

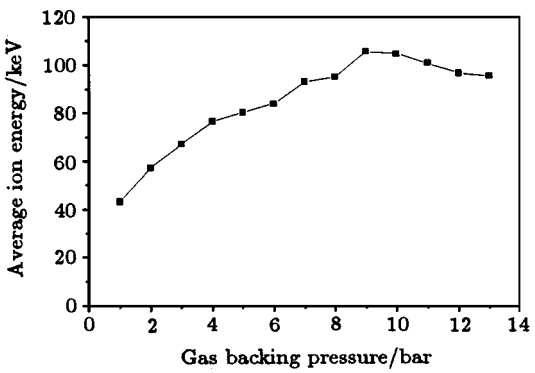


Fig.6. Dependence of the average energy of xenon ions on the gas backing pressure. The laser peak intensity is 2×10^{16}W/cm^2.

In the experiment of the intense laser pulse interaction with deuterium clusters to induce deuterium–deuterium fusion, increasing the yield of neutrons is one of the main purposes. Bearing in mind that the cross section of the deuterium–deuterium fusion reaction increases sharply for D energies beyond 5keV, and starts saturating at about 50keV,[15] increasing the average energy of the deuterium ions is of key importance. According to the results of our experiment, it could be feasible to select appropriate parameters of the laser pulse and the cluster source to increase the yield of neutrons in future laser driven nuclear fusion experiments.

4. Conclusion

We have measured the energy absorption efficiency of a dense jet of rare-gas clusters to the incident intense femtosecond laser pulses. The results show that at the comparatively low gas backing pressure, the energy absorption efficiencies increasing quickly with increasing the gas backing pressure, and at the same gas backing pressure, the energy absorption efficiency of xenon clusters is higher than that of argon. Further increase the gas backing pressure, the energy absorption efficiencies can reach as high as 90%

and 85% for large clusters of xenon and argon, respectively. The energies of ions emitted from the exploding argon and xenon clusters have also been measured. Similar to the situation in energy absorption, at comparatively low gas backing pressure, the average ion energies of argon and xenon clusters increase rapidly with increasing the gas backing pressure, and the average ion energy of xenon clusters is higher than that of argon clusters at the same gas backing pressure. The measurement of the ion energy indicates that for the average energy of the ions, there is an optimum gas backing pressure characteristic of cluster size. The mechanism of it has been analysed within the frame of the microplasma sphere model and its possible application has been discussed.

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