Volume 9, Number 6
1009-1963/2000/09(06)/0432-04
CHINESE PHYSICS
June, 2000
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MEASUREMENT OF IONIC ENERGY FROM FEMTOSECOND LASER-HEATED ARGON CLUSTERS*

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(Received 4 November 1999; revised manuscript received 27 January 2000)

The high backing pressure argon gases adiabatically expand into vacuum through a pulsed gas jet to nucleate into large clusters. The clusters were heated by a 45fs, 2.3×10^{16} Wcm^{-2} Ti: sapphire laser. The high energy of the ions produced in the cluster explosion was measured using time-of-flight spectrometry. The maximum and average kinetic energy of the ions were 0.2MeV and ∼12.5keV, respectively, indicating that the femtosecond laser interactions with argon clusters are more energetic than interactions with atoms and molecules.

PACC: 3640; 3290

I. INTRODUCTION

There have been extensive experimental and theoretical investigations of the ultrashort laser-matter interactions. The fs laser pulses based on chirped pulse amplification (CPA) have a peak intensity of 10^{14}–$10^{19}\mathrm{Wcm}^{-2}$ and pulse duration shorter than 100 fs. [1,2] The laser targets are usually the solid density bulks and atomic gases. These experiments were motivated by a desire to generate ions, electrons and photons with far higher energies than that of a single driving laser photon. Particularly, most of these experiments were involved in the inertial confinement fusion and coherent X-ray emission, such as X-ray laser and high order harmonic generation. Recently, atomic clusters have attracted much attention. Clusters are aggregates of atoms or molecules that are bonded together by Van der Waals forces. The clusters are much larger than molecules but much smaller than solid bulks. The gas phase atomic clusters have an average low density comparable to that of gases but have a local solid density. Ions with energies as high as 1MeV in the xenon clusters were produced and the average ionic energy was 45keV,[3] indicating that the fs laser-cluster interactions are more energetic than interactions with atomic gases. More recently, neutrons[4] with typical 2.45MeV energy have been generated from deuterium cluster fusion experiments and the neutron yield per joule of laser energy was found comparable to that of national-scale laser fusion experiments. In most cases, the reservoir gases used in such experiments were xenon and krypton. This paper reports the argon ionic energy spectra from fs laser-heated cluster plasmas.

II. EXPERIMENTAL SETUP

The atomic clusters are produced in the pulsed gas jet. The high backing pressure gases expand adiabatically into vacuum. This expansion cools the gases to supersaturate and nucleate into clusters. We have designed a pulsed solenoid valve gas jet which can work for gas backing pressure in excess of 5MPa and with a repetition rate of 10Hz. The sizes of noble gas atomic clusters produced in the pulsed jets depend on the atomic species, gas stagnation pressure and initial temperature, and the geometry of the jet nozzle. Hagena and Obert found an empirical parameter Γ^* that can predict the onset of clustering in gas jets and the size variation of the clusters.[5,6] The Hagena's parameter $\Gamma^*$is given by

$$\Gamma^* = k\frac{(d/\tan\alpha)^{0,85}p_0}{T_0^{2.29}}, \qquad (1)$$

where d is the jet throat diameter in μm, α is the jet expansion half angle, p_0 is the gas stagnation pressure in 10^2Pa and T_0 is the gas reservoir temperature in K, and k is a constant depending on the atomic species($k \approx 3.85$ for helium, 185 for neon, 1650 for argon, 2890 for krypton and 5500 for xenon). In our experiment, d, α and T_0 are 360 μm, 5° and 300 K, respectively, and p_0 varies between 0.4 and 4.5 MPa. A Hagena's parameter Γ^* corresponds to a mean number $\overline{N}$ of atoms in a cluster droplet, no matter how

* Project supported by the National "Climb" Program of China, the National Natural Science Foundation of China (Grant No. 29890210) and the Major Project for the Foundation Research of the Chinese Academy of Sciences (Grant No. KJ 951-B1-701).

the constituents of Γ^* in Eq.(1) vary. The larger the parameter Γ^*, the larger the number $\overline{N}$. It is obvious that the heavier noble atomic gases such as xenon, krypton and argon are easier to form clusters than helium and neon. Farges[7] found an approximate relation between $\overline{N}$ and Γ^*, i.e., $\overline{N} \approx 10^{-4}(\Gamma^*)^{2.0}$. So the sizes of the argon clusters produced in our jet were between 2.7×10^4 and 3.5×10^6 atom/cluster.

The laser used in the experiment is a 10Hz, 785nm Ti: sapphire laser, which delivers 5 up to 90mJ laser energy in 45fs. Figure 1 shows the experimental setup. The light is focused with a $f/10$ non-spherical lens into the exit of the gas jet to an intensity of $6\times10^{15}-1\times10^{17}$ Wcm^{-2} at the focus, which is 1-2mm below the nozzle output. The laser intensity is determined from the appearance intensity of He^{2+} measurement. The background pressure of the interaction chamber is 9×10^{-5}Pa. When the argon gas is allowed to enter into the chamber, the working pressure of the interaction chamber is below 6×10^{-2}Pa by adjusting the duration of the gas pulse, no matter how much the stagnation pressure of the gas is. The pressure of the detection chamber keeps 3.2×10^{-3}Pa in any case. The gas jet works with a repetition rate of 1Hz. A time-of-flight spectrometry is used to determine the ionic energy distribution. The ions pass through a 649mm field-free drift tube and strike a dual microchannel plate detector (DMCP). The DMCP output signal is capacitively coupled to an analog-digital converter(ADC) with 4ns time-resolution. The drift tube is grounded and its axis is perpendicular to both the laser beam and the gas flow. The front-end of the drift tube is 10mm apart from the laser focus. In the center of the front-end of the tube there is a 10mm aperture covered with a metal mesh.

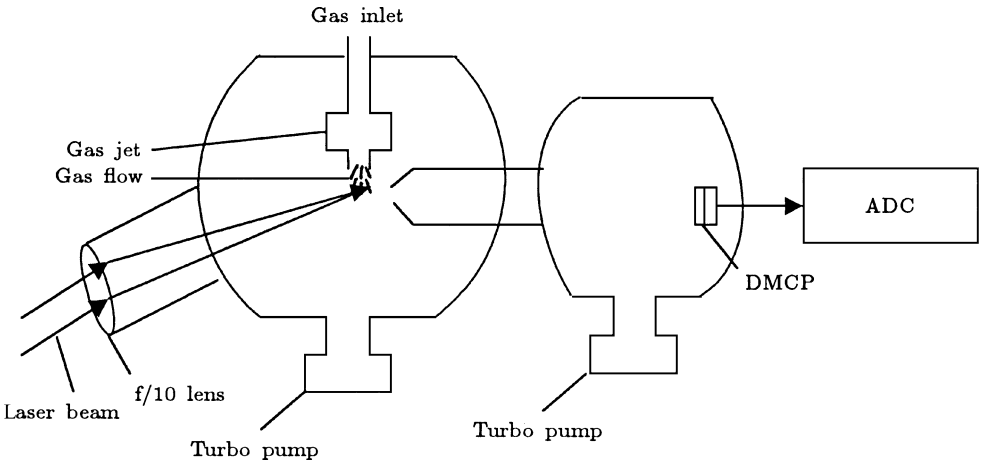


Fig.1. The experimental setup.

Ⅲ. EXPERIMENTAL RESULTS AND DISCUSSION

A typical time-of-flight trace accumulated for 500 laser shots is shown in Fig.2. The laser energy is 20mJ and the intensity evaluated is 2.3×10^{16} Wcm^{-2}. The argon gas backing pressure is 2MPa and the duration of the gas pulse is 440μs. Two peaks in the spectrum are separated clearly. The first sharp peak in order of time is from the fast electrons and the second peak extending from 1000ns is due to ions. It can be seen that the electrons do not overlap the highest energy ion signal. This makes it easy to deal with the ion signal. Since the ADC should be triggered approximately 1μs before the laser pulse, we consider the starting time of the ion signal is the time when the fast electrons arrive at DMCP. This approximation has little effect on even the highest ionic energy.

From the particle kinetic energy formula

$$E = \frac{1}{2}mv^2 = \frac{1}{2}m\frac{l^2}{t^2}, \tag{2}$$

we can obtain the ionic energy spectrum from ionic time-of-flight spectrum

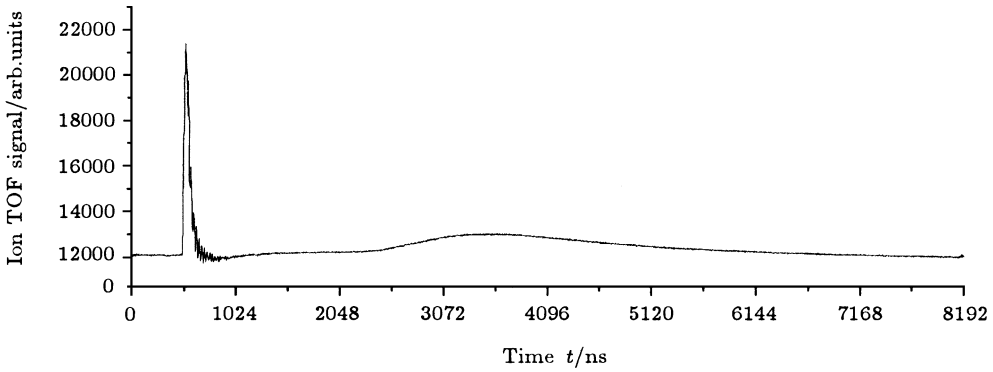


Fig.2. The ionic time-of-flight spectrum from argon cluster irradiated by 20mJ, 45fs laser pulses at a gas stagnation pressure 2MPa.

$$f(E) = \frac{\mathrm{d}N}{\mathrm{d}E} = \frac{\mathrm{d}N}{\mathrm{d}t}\frac{\mathrm{d}t}{\mathrm{d}E} = \frac{t^3}{ml^2}f(t) \propto t^3 f(t), \qquad (3)$$

where l is the length of drift tube, t is the ionic time-of-flight. Figure 3 shows the ionic energy spectrum converted from Fig.2. This energy spectrum is different from previously observed ionic energy spectrum from the intense-field fragmentation of molecules and shows that hot ions with energies up to 0.2MeV are produced. Previous study[8] of diatomic Cl_2 molecule subject to the 790nm, 130fs laser pulse with intensity (2×10^{16} Wcm^{-2})close to ours showed that the highest ionic energy obtained was below 100eV. The energy from molecular ions is due to the multielectron dissociative ionization induced by intense laser fields, which leads to charge buildup on molecular ions. This charge accumulation finally makes the molecular ions to explode by Coulomb repulsive forces. For small clusters such as C_{60} [9]subject to intense laser pulses in the intensity range of 5×10^{14} to 5×10^{15} Wcm^{-2}, the highest ionic energy is above 1keV and the ionic energy distribution is well modeled by Coulomb explosion mechanism.

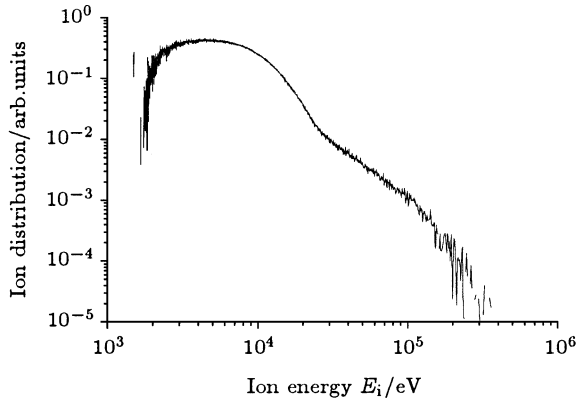


Fig.3. The argon ionic energy spectrum under the experimental condition same as Fig.2.

The highest ionic energy in our experiment was between those of small clusters and solid targets. Previous studies[10] showed that high-Z solid targets subject to laser pulse with intensity similar to ours produced characteristic ionic energy of 100-200keV.[11] The average ionic kinetic energy, defined as $< E >= \frac{\int Ef(E)\mathrm{d}E}{\int f(E)\mathrm{d}E}$, in our experiment can be obtained as high as 12.5keV.

Two mechanisms might be responsible for the high ionic energy from the atomic clusters irradiated by the fs lasers. The first is the so-called Coulomb explosion, as occurs in molecules and small clusters. The second is the hydrodynamic expansion as occurs in the solid density plasma. In the case of the Coulomb explosion, the energy of repulsion is expected to scale as the square of the ionic charge, whereas, in the case of hydrodynamic expansion, the energy is purely thermal and scales linearly as the ionic charge and the electron temperature. Ditmire *et al.*[11] assumed the cluster as a plasma ball subject to laser pulses. In their model, the cluster is initially tunnel-ionized by an intense laser field. The 'seed' electrons absorb laser energy by inverse bremsstrahlung(IB). The hot plasma then hydrodynamically expands due to thermal pressure. Although in their model Coulomb explosion mechanism due to hot electron's free streaming out of plasma ball is also considered, the Coulomb pressure is found only 10% of the thermal pressure even at the time when the IB heating rate is highest. This model can re-create the major feature of ionic kinetic energy distribution in large size xenon cluster plasmas. However, Lezius *et al.*[12] pointed out that in argon clusters that contain typically 10^5 atoms the dominant mechanism was Coulomb but in xenon clusters that contain about 10 times more atoms the hy-

drodynamic mechanism played a major role. Under our argon cluster experimental condition, we cannot determine in which process most of the ions are produced because the vacuum of the interaction chamber is so low that the highly charged state ions might recombine. The decrease of signal of ions with energies below 2keV is due to low response efficiency of DMCP to these ions.

In conclusion, we have measured the high-energy distribution of ions from femtosecond laser-heated atomic argon cluster plasmas using time-of-flight technique.Substantial numbers of ions with energies up to 0.2MeV have been produced in the laser interaction process and the average ionic energy of 12.5keV was obtained.Detailed theoretical works are needed to elucidate the experimental results.

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