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Dependence of ion kinetic energy and charge on cluster size in multi-photon ionization of xenon clusters

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ABSTRACT

Multi-photon ionization of xenon clusters created by nozzle expansion has been investigated by time-of-flight mass spectrometry. With infrared irradiation, cluster ions beyond the dimer are missing from the mass spectra, but there is a copious yield of multiply-charged atomic ions. With ultraviolet irradiation, multiple ionization is almost completely suppressed and singly ionized large cluster ions are detected. The mean cluster ion size under UV ionization shows an increasing trend with the stagnation pressure for nozzle expansion, as do the mean charge state and kinetic energy of atomic ions formed via IR irradiation. Interrogating the same cluster population by two different probes provides a corroboration between the effect of the size of the cluster in the ionization mechanism and the kinematic effects thereof. Furthermore, it provides evidence for the influence of the neutral cluster size on the formation of multiply charged ions by IR irradiation via the electron re-collision mechanism.

1. Introduction

Ionization of xenon clusters by ultrafast laser pulses yields multiply charged ions of xenon atoms in the mass spectrum. Ionic states up to Xe^{13+} has been reported by Augst et al. [1] when Xe clusters are ionized using 1 ps focused laser pulses of wavelength 1053 nm (~10^{16} W/cm^2). At higher laser intensities (~10^{17} W/cm^2, 2 ps laser pulses), Ditmire et al. [2] have observed ionic charge states up to Xe^{29+} in the mass spectrum. At similar laser intensities, it has been found that the efficiency of the production of higher charge states of xenon atoms from clusters increases with compression of the time duration of laser pulses [3–5]. Ions with charge states up to Xe^{40+} and kinetic energy 1 MeV has been observed by Ditmire et al. in the ionization of Xe clusters irradiated by fs laser pulses of intensity 2×10^{16} W/cm^2 [6,7]. Such ionization mechanism and formation of multiply charged atomic ions from clusters have been discussed in detail in the reports by Krainov et al. [8] and Saalmann et al. [9]. Earlier experiments have suggested that the interaction of high intensity laser pulse with cluster is more energetic due to the high local density in cluster than the laser-atom interaction [4,10]. It was found that electrons in cluster undergo rapid collisional heating for very short time (<ps) before the cluster break apart in the intense laser pulse [3,4].

In the nanosecond regime, the production of multiply charged atomic ions is based on the heating of quasi-free electrons by the optical field. In an experiment on Xe clusters with 532 nm irradiation at 10^{10}–10^{11} W/cm^2, Luo et al. observed the formation of Xe^{11+} [11], while Niu et al. observed the formation of Xe^{19+} and Xe^{11+} while irradiating the clusters with 1064 nm and 532 nm, respectively, at 10^{11} W/cm^2 [12]. The formation of Xe^{10+} has been reported at even lower intensities of 5×10^9 W/cm^2 [13]. The formation of highly charged atomic ions depends on the size of the neutral clusters [11,13–15]. It was observed that the maximum kinetic energy of ions increases with increasing cluster size [6,16]. A model has been developed to explain the large kinetic energies of the atomic ions thus formed [17].

Ionization of Xe clusters by ns laser pulses also results in creation of singly charged cluster ions, which are seen in the mass spectrum. Singly ionized Xe cluster ions up to Xe_{16}^{+} have been observed by Luo et al. [11]. In the majority of experiments, Xe cluster beam is generated by nozzle expansion and the size distribution of neutral clusters is inferred on the basis of Hagena's theory [18], which predicts that the average cluster size increases with the nozzle diameter and the stagnation pressure. However, no direct measurement of the cluster size is made in these experiments and the stagnation pressure of the nozzle beam is used as a proxy for the mean or most likely cluster size.

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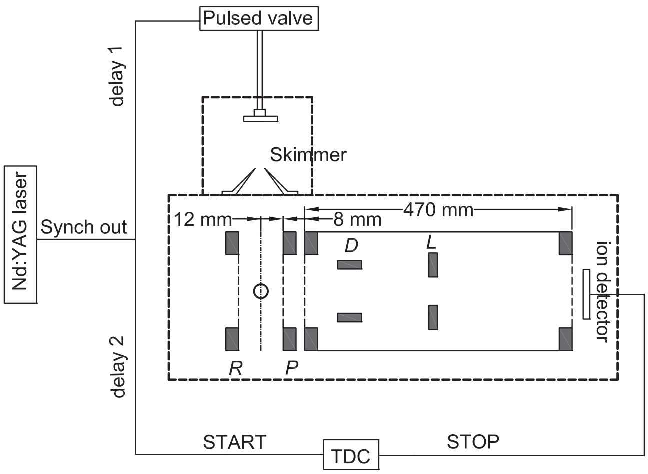

Fig. 1. A schematic diagram of the TOFMS coupled with a cluster source. Gas pulses from the valve are intercepted by Nd:YAG laser pulses. The first mesh holder *R* works as a repeller for ions whereas the second one *P* as the puller electrodes, which have high transmission meshes. The dashed line between the meshes represents the plane containing the laser beam and the nozzle beam, with the circle indicating the ionization volume. A pair of deflectors *D* and a lens *L* are used to guide the ions toward the MCP, if needed.

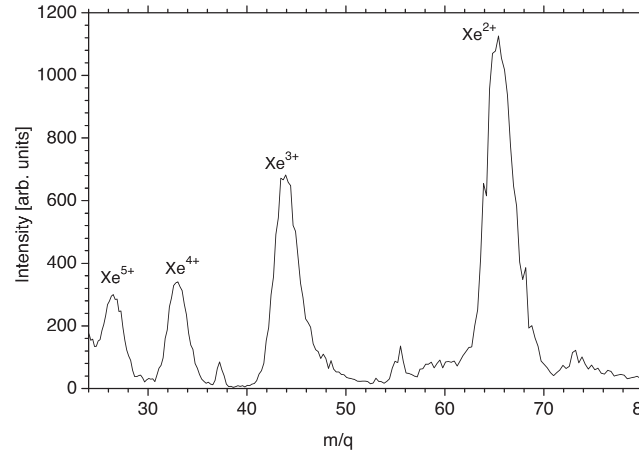

Fig. 2. Mass spectrum of multiply charged Xe atomic ions from pulsed nozzle expansion at 3 bar stagnation pressure irradiated by 1064 nm laser pulses.

In this work we examine the possibility of using a different proxy for the cluster size, based on the observed mean cluster ion size, when the nozzle beam is subjected to ionization by UV laser pulse. We have found the dependence of mean charge of the atomic ion distribution (under IR irradiation) and the mean cluster ion size (under UV irradiation) on the stagnation pressure. We have also determined the mean kinetic energy of the atomic ions based on the ion TOF distribution. The mean kinetic energies of the atomic fragments is also found to increase with the mean size of the neutral clusters.

2. Experiment

The apparatus consists of a nozzle expansion cluster source, a time-of-flight mass spectrometer and a high power Nd:YAG laser. Fig. 1 shows a schematic diagram of the experimental set-up. Xenon clusters are formed by supersonic expansion of gas from several bar pressure into vacuum through a pulsed valve (*Parker, Series 9*) having a cylindrical nozzle of diameter 1 mm. The valve open time is 300 μs and the repetition rate is 30 Hz. The expansion chamber is pumped by a combination of turbomolecular and scroll pumps. The nozzle beam is skimmed by a conical skimmer of 1.2 mm diameter opening. The skimmer separates the expansion chamber from the mass spectrometer chamber. Typical operating pressure of the cluster source is around 3 bar. The stagnation pressure and the nozzle diameter are the key parameters that determine the size distribution of the clusters in the expanding gas. High pressures strongly enhance clustering, as does an increase in the nozzle diameter, provided the vacuum conditions are preserved. The shape of the nozzle also determines the cluster size distribution; conical nozzles lead to large clusters. Determination of the actual cluster size distribution is difficult and generally theoretical estimates are relied upon using Hagena's theory [18]. For our experimental parameters, theoretical value of the expected average cluster size runs into several thousands.

The TOF spectrometer, developed in-house, is of Wiley–McLaren type [19], having two static electric field regions of extent 1.2 and 0.8 cm, and a field-free drift region of 47 cm extent. The electric field in the first region is 250 V/cm and in the second region it is 2500 V/cm. The ion detector is microchannel plate (MCP). The flight time information can be obtained by operating the spectrometer in the ejected electron and recoil ion coincidence mode (applicable only for studying positive ions) or, when a pulsed ionizing agent is available, by triggering the acquisition synchronous with the ionization pulse (used for the present study). The time measurement is carried out using a multi-hit flash TDC. The resolving power in the first mode is 340, which may be taken as the intrinsic resolving power of the spectrometer. In the pulsed ionization the resolving power is diminished. The mass spectrometer vacuum enclosure has a base pressure of 8×10^{-8} mbar, which rises to 3×10^{-6} mbar in pulsed valve operation.

A pulsed Nd:YAG laser at the fundamental wavelength of 1064 nm is used for the first part of this study, while the second part of the study is carried out using the third harmonic output at 355 nm. Synchronization of the laser pulse and the gas pulse is critical for successful operation, and a systematic variation of the delay between the two pulses was carried out to establish optimum timing. For both wavelengths employed, the laser pulse width is 8 ns and the intensity is $\sim 10^{10}$ W/cm^2. The laser beam is focused using a convex lens of 20 cm focal length. The laser fluence is around 350 J/cm^2 with 1064 nm and 60 J/cm^2 with 355 nm.

The ionization mechanism of atoms exposed to intense laser fields can be separated into two different ionization regime, MPI and tunneling ionization, on the basis of Keldysh parameter (γ). The Keldysh parameter, is defined as $\gamma = (I_p/2U_p)^{1/2}$, where I_p is the ionization potential of atom and U_p is the pondermotive energy of the quivering electron in the laser field. The ponderomotive energy of the electron in a laser field is given by $U_p = 9.33 \times 10^{-14} \times I \times \lambda^2$ eV, where I is the laser pulse intensity in W/cm^2, and λ is the wavelength in μm [8,20]. MPI dominates the ionization process when $\gamma > 1$ [8,20], which is the case for both wavelengths (1064 nm and 355 nm) in the present study.

3. Results

3.1. IR irradiation: multiple ionization

When 1064 nm radiation is employed, there are singly ionized peaks of the monomer and the dimer, but, more interestingly, there are several multiply charged atomic ions (Xe^{q+}, $q = 1–5$) in the mass spectrum. A sample mass spectrum with IR ionization at 3 bar stagnation pressure is shown in Fig. 2. The presence of clusters is vital for the production of multiply charged atomic ions. This was confirmed by obtaining a mass spectrum with the pulsed nozzle beam replaced by a low number density effusive beam of Xe gas (which is devoid of clusters). The mass spectrum in the case of effusive beam