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Research paper

Dependence of multiply charged ions on the polarization state in nanosecond laser-benzene cluster interaction

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ABSTRACT

This paper investigated the dependence of multiply charged ions on the laser polarization state when benzene cluster was irradiated with 532 and 1064 nm nanosecond laser. A circle, square and flower distribution for C^{2+}, C^{3+} and C^{4+} were observed with 532 nm laser respectively, while flower petals for C^{2+}, C^{3+} and C^{4+} were observed at 1064 nm as the laser polarization varied. A theoretical calculation was performed to interpret the polarization state and wavelength dependence of the multiply charged ions. The simulated results agreed well with the experimental observation with considering the contribution from the cluster disintegration.

1. Introduction

The interactions of nanosecond laser with clusters have been an active research area over the past few decades, and many interesting phenomena have been observed such as the production of multiply charged ions and their dependences on the sizes of the clusters and the wavelength of the laser [1–6]. C_6H_6, a symmetric top molecule, has been used extensively to study the ionization and dissociation dynamic process [7–12]. Recently, much effort has been conducted in order to gain further information about the general mechanism behind the nanosecond laser–cluster interaction [13,14].

Kong et al. discovered the production of multiply charged ions C^{2+} and C^{3+} in the C_6H_6 cluster beam while only singly charged fragment ions were observed in the C_6H_6 diffused beam [13]. Zhang et al. investigated the influences of laser intensity on the electron energy and multiply charged ions using a nanosecond laser of 10^9–10^{11} W/cm^2, and proposed that electron impact ionization was the primary formation channel for the multiply charged ions in C_6H_6 clusters [14]. As the electric field strength of the nanosecond laser with intensity of 10^9–10^{11} W/cm^2 is small compared to that of the binding molecular fields of the valence electrons, the theoretical models based on field ionization were not suitable for explaining the production of multiply charged ions in nanosecond laser field although they did well in femtosecond laser field [16–20]. Previously, a theoretical model was proposed to explain the production of multiply charged ions and their dependences on wavelength of the laser, where three steps "multiphoton ionization (MPI) triggered-inverse bremsstrahlung (IB) heating-electron impact (EI) ionization" were involved [21]. Zhao et al. investigated the evolution of the multiple charged ion intensity as a function of the laser intensity, and proposed that the formation of more ionized sites in a cluster at MPI process would split the cluster due to Coulomb repulsion, which would suppress the formation of multiply charged ions [12]. To further study the influence of the MPI process in laser-cluster interaction, different laser wavelengths were chosen [15,21,22]. However, selective study of the effect of the MPI on multiply charged ions could not be achieved as the laser wavelength affects not only the MPI process but also the IB process.

David et al. compared the heating rates by inverse Bremsstrahlung for circularly and linearly polarized laser radiations, and found that the heating rate at intensity lower than 10^{14} W/cm^2 was independent of laser polarization [23]. Thus, laser polarization became an ideal parameter for selectively studying the influence of the MPI on the formation of the multiply charged ions. This paper investigated the effect of the MPI on the formation of multiply charged ions with 532 and 1064 nm nanosecond lasers by changing the laser polarization state. A theoretical calculation was performed to simulate the dependence of the multiple charged ion intensities on the laser polarization states.

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2. Experimental

The experiment setup used for this work was a linear dual polarity time-of-flight mass spectrometer (TOFMS), which has been described in detail previously [12,14]. Only the main features are outlined here. The liquid C_6H_6 vapor seeded in argon gas is passed through a stainless steel tube and sprayed out from a pulsed valve (Bosch, 026133 025A) into the source chamber of TOFMS. The clusters were formed by the supersonic expansion in the source chamber and then entered the ionization chamber through a skimmer (diameter = 1 mm). In the ionization chamber, two accelerating stages of mass spectrometer were constituted by three electrodes of repeller, extraction, and accelerator. The inner and outer diameters of the electrode are 10 and 90 mm, respectively. The distance between the repeller and extraction electrodes is 20 mm, and the ionization took place in middle between the repeller and extraction electrodes. The distance between the extraction and accelerator is 10 mm. The voltages on the repeller and extraction electrodes were set to 2800 and 1680 V, respectively. The accelerator electrode was grounded. After passing through the accelerator electrode, the ions entered the field-free region with length of 500 mm. Finally, ions were detected at the end of the field-free region by a two-stage microchannel plates.

The laser system was a commercially available Nd-YAG laser (New Wave Tempest 30). The output energy of the second harmonic at 532 nm was less than 30 mJ with pulse duration of 5 ns. The laser beam was focused by a quartz lens with $f = 25$ cm, and the diameter of the focus point was about 0.1 mm. The laser pulse energy was measured by an energy meter (Molectron, model PM500). The laser intensity I was calculated by the formula $I = E/(S \times t)$, where E is the power of the laser, S is the spot area, t is the pulse duration of the laser. A quarter-wave plate was used to change the laser polarization from linear to circular by rotating it.

3. Results and discussion

Fig. 1(a) displays the typical mass spectra as the benzene cluster beam was irradiated by linearly polarized 532 nm nanosecond laser with intensity of 4.4×10^{10} W/cm^2. In Fig. 1(a), the dominant multiple charged carbon ions were C^{2+} and C^{3+}, and the C^{4+} signal was weak, with intensities $C^{2+} > C^{3+} >> C^{4+}$. By rotating the quarter wave plate, the polarization state of the laser beam was changed from linearly polarized (0°, 90°, 270°, 360°) to circularly polarized state (45°, 135°, 225°, 315°). Fig. 1(b) showed the dependence of C^{2+}, C^{3+} and C^{4+} intensities on the laser polarization. When the polarization state changed from linear to circular, the normalized intensity of C^{3+} and C^{4+} increased by about 20% and 53%, while the intensity of C^{2+} almost did not change. It indicated that degrees of dependence on polarization state at 532 nm were $C^{4+} > C^{3+} > C^{2+}$.

At 532 nm, the back- and forward peaks of C^{2+} and C^{3+} were observed due to the different flight directions toward and backward to the detector. The interval of time depends on the laser intensity the valence state of the ions and also the size of the cluster, which was discussed in details [25].

Fig. 2(a) displays the typical mass spectra when the benzene cluster beam was irradiated by a 5 ns laser with wavelength of 1064 nm. The C^{4+} signal was dramatically higher than those of C^{2+} and C^{3+}, and the intensity order was $C^{4+} >> C^{3+} > C^{2+}$. Obviously, longer wavelength at 1064 nm facilitated the generation of higher charged ions compared with that at 532 nm, which was also observed in other cluster systems, such as NH_3 and CH_3I [2,21]. In Fig. 2(b), the evolution curves of C^{2+}, C^{3+} and C^{4+} look like a petal when the quarter-wave plate was rotated from 0° to 360°. However, the evolution trend of C^{3+} in Fig. 2(b) was quite different from that of C^{3+} in Fig. 1(b). When the polarization changed from linear to circular, the intensity of the C^{2+}, C^{3+} and C^{4+} decreased by about 50% at 1064 nm, while increased by about 20% and 53% for C^{3+} and C^{4+} at 532 nm as shown in Fig. 1(b).

How to explain the dependence of multiply charged ions on the laser polarization state? As known to all, the polarization state would change multiphoton ionization (MPI) rate. Shchatsinin et al. put forward a theory to describe the influence of polarization state on MPI [24], the overall ionization rate in an N-photon process depends on the time averaged intensity $\langle L^n(t,\alpha)\rangle$

$$\langle L^n(t,\alpha)\rangle = L^n \frac{w}{2\pi}\int_0^{\frac{2\pi}{w}} \left[1-\sin(2\alpha+\frac{\pi}{2})\cos(2wt)\right]^n dt$$
$$= L^n \cos^n(2\alpha+\frac{\pi}{2}) P_n\left(\frac{1}{\cos(2\alpha+\frac{\pi}{2})}\right), \quad (1)$$

where $P_n(x)$ are the Legendre polynomials, L is the laser intensity, α is the ellipticity angle, w is the laser angular frequency, n is the laser power index in the multi-photon process. The classical average displayed a drop in the effective multiphoton ionization cross section as the laser polarization changed from linear to circular, which was ascribed to the fact that the electric field vector for circularly polarized laser was only $1/\sqrt{2}$ of that for the linearly polarized laser [24].

Suppose that M is the number of cluster, which could produce multiply charged ions; P is the MPI probability of a molecule. We assume that the change of M was proportional to MdP as follow:

$$dM = -\beta M dP \quad (2)$$

where dP is a small change of MPI probability. A disintegration factor of cluster β is introduced to reflect the influence of MPI probability on the generation of multiply charged ions, which depends on the cluster size, charge state, laser wavelength, laser intensity and electronic properties of related molecules. By integrating Eq. (2), we could obtain:

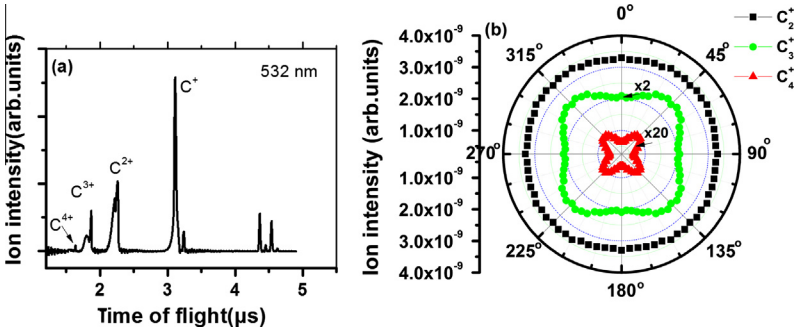

Fig. 1. (a) The typical mass spectrum of benzene cluster for linear polarized 532 nm laser with the intensity of 4.4×10^{10} W/cm^2; (b) The dependence of C^{2+}, C^{3+} and C^{4+} intensities on laser polarization state as the C_6H_6 cluster beam was irradiated by 532 nm laser.