PHYSICAL REVIEW A **90**, 053428 (2014)

Photodissociation of Br_2 molecules in an intense femtosecond laser field

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(Received 25 August 2014; revised manuscript received 3 November 2014; published 24 November 2014)

We experimentally demonstrate the photodissociation process of Br_2 molecules in the intense femtosecond laser field by a dc-sliced ion velocity map imaging technique. We show that four fragment ions Br^{n+} ($n = 1$–4) are observed, and their kinetic energy increases while their angular distribution decreases with the increase of the charge number. We prove that the low (or high) charged fragment ions result from the photodissociation of the low (or high) charged parent ions. We explain the changes of the kinetic energy and angular distribution in these fragment ions by considering the potential energy curves of these parent ions that involve both the interaction of the Coulomb repulsive energy and chemical bonding energy. We also explain the experimental observation that the measured kinetic energy release in the experiment is much smaller than the theoretical calculation by enhanced ionization at a critical distance.

DOI: 10.1103/PhysRevA.90.053428 PACS number(s): 33.80.Gj, 34.20.Gj, 42.50.Hz

I. INTRODUCTION

Molecular photodissociation is a chemical reaction in which a chemical bond is broken down by photons; any photon with sufficient energy can affect the chemical bonds of the molecule. A single photon in the infrared spectral range is often not energetic enough for direct photodissociation, but the molecule may gain internal energy to overcome its barrier for dissociation by absorbing multiple infrared photons. With the advent of the femtosecond laser pulse, the ultrafast intense laser field can quickly strip away one or more electrons, and then the chemical bonds of the molecule are broken by the rest energy of the laser field or the Coulomb repulsive energy of the parent ions [1–4]. Recently, the molecular photodissociation in the intense femtosecond laser field has attracted considerable attention because of its potential applications in the study of the molecular dynamics, such as chemical bond breaking [5], internal conversion [6–8], concerted elimination [9–11], and so on.

The molecular photoionization and photodissociation in the laser field have been widely studied [12–26], and some theoretical models have been proposed to illustrate the dynamical process [1,27–29]. The multielectron dissociative ionization (MEDI) model showed that the femtosecond laser field can quickly strip several electrons off at the equilibrium nuclear distance, and then subsequent dissociation of the charged molecular ions is followed under the influence of the Coulomb repulsive energy [1]. However, the postdissociative ionization (PDI) model showed that the molecule can be ionized for the second time during the dissociation process [27]. Furthermore, Bandrauk *et al.* proposed a charge-resonance enhanced ionization (CREI) model to explain the ionization of the H_2^+ ion [28], and showed that there exists a pair of charge-resonant states that strongly coupled to the laser field at a critical distance R_c, where the ionization probability can be significantly enhanced. Chelkowski *et al.* proposed a two-step model that the molecule can lose electrons and Coulomb explosions take place twice at their equilibrium distance and critical distance, respectively [29].

Halogen and halogen compounds can damage the atmosphere ozone layer [30,31], and therefore their molecular photodissociation process under the laser field has been an active field for many years [12–26], especially for the halogen diatomic molecules Cl_2, Br_2, and I_2 [32–38]. In this paper, we experimentally demonstrate the photodissociation process of Br_2 molecules in the intense femtosecond laser field by a dc-sliced ion velocity map imaging technique. Our experimental results show that four fragment ions Br^{n+} ($n = 1$–4) can be observed, and their kinetic energy (KE) increases while their angular distribution decreases with the increase of the charge number. Our analysis indicates that the low (or high) charged fragment ions come from the photodissociation of the low (or high) charged parent ions. The potential energy curves of these parent ions are used to explain the changes of the kinetic energy and angular distribution in these fragment ions by considering both the interaction of the Coulomb repulsive energy and chemical bonding energy. Furthermore, the enhanced ionization at a critical distance of parent ions can explain such an experimental observation that the measured kinetic energy release (KER) in the experiment is much lower than the theoretical calculation.

II. EXPERIMENTAL SETUP

The experimental arrangement has been exactly described in our previous work [39], and here only a brief description is given. A mode-locked Ti:sapphire laser is used as excitation source with the central wavelength of 800 nm, the repetition rate of 1 kHz, and the pulse duration of 90 fs. The output femtosecond laser pulse is focused into the gaseous sample by a lens with 400-mm focus length, and the laser intensity at the focus position is estimated to be about 1.5×10^{14} W/cm^2. The gaseous molecules are seeded into the chamber by helium with 0.2 atm and then injected to the ionization region by a pulse valve with the repetition rate of 100 Hz and the duration

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time of 160 μs. By the multilens velocity mapping apparatus, these fragment ions are sent to a two-stage microchannel plate (MCP) coupled to a P47 phosphor screen, and finally their intensities are detected by photomultiplier tubes (PMTs) and their spatial distributions are imaged by a charge-coupled device (CCD). All of the timing sequence control is performed by a Stanford Instrument Digital Delay/Pulse Generator (DG535).

III. RESULTS AND DISCUSSION

Figure 1 shows the mass spectrum of Br_2 molecules irradiated by the femtosecond laser pulse with the laser intensity of 1.5×10^{14} W/cm^2. As can be seen, in addition to the parent ion $Br_2{}^+$, the fragment ions Br^+, Br^{2+}, Br^{3+}, and Br^{4+} can be observed. Considering that the spectral resolution of our experimental apparatus is not high enough to distinguish the two isotopes ^{79}Br and ^{81}Br, therefore the peak in $m/q = 80$ represents the combination of the two fragment ions $^{79}Br^+$ and $^{81}Br^+$. The same method is also employed in the other multicharged fragment ions Br^{2+}, Br^{3+}, and Br^{4+}. Furthermore, these fragment ions in the region of the low mass to charge ratio should result from the H_2O molecule in our experimental setup. To demonstrate the photodissociation process of these fragment ions Br^{n+} ($n = 1$–4), we present their dc-sliced ion images, as shown in Fig. 2. It is obvious that there are multiple photodissociation channels in each fragment ion, and the kinetic energy increases while the angular distribution decreases with the increase of the charge number. Especially, it is noteworthy that a spot with zero kinetic energy in the central position of Fig. 2(d) is observed, which is attributed to the parent ion $Br_2{}^{2+}$, where the fragment ion Br^+ has the same mass to charge ratio as the parent ion $Br_2{}^{2+}$.

The photodissociation channel assignment is critical to understanding the photodissociation process. One simple way to assign the photodissociation channel is by observing the kinetic energy. In the two-body Coulomb explosion process, the kinetic energy of a pair of fragment ions should satisfy the relation of

$$\frac{E_{\mathrm{kin}}(\mathrm{Br}^{P^+})}{E_{\mathrm{kin}}(\mathrm{Br}^{P^+})} = \frac{m(\mathrm{Br}^{Q^+})}{m(\mathrm{Br}^{Q^+})}, \tag{1}$$

where E_{Kin} is Kinetic energy, m is the mass of the fragment ion, and P and Q are the charge number of the two fragment ions, respectively. Figure 3 shows the kinetic energy of these fragment ions Br^{4+} (a), Br^{3+} (b), Br^{2+} (c), and Br^+ (d) calculated from Fig. 2. One can see that multiple peaks are observed in each fragment ion, and each peak is corresponding to one photodissociation channel. In order to facilitate the discussion below, these peaks from low to high kinetic energy are respectively labeled with P_k ($k = 1$–3). According to Eq. (1), it is easy to verify that P_1 peak in Br^{4+} ion and P_3 peak in Br^{3+} ion should come from the same photodissociation channel, which can be assigned to the channel of $Br_2{}^{7+} \rightarrow Br^{4+} + Br^{3+}$. Applying the same method, P_1 peak in Br^{3+} ion and P_3 peak in Br^{2+} ion can be assigned to the channel of $Br_2{}^{5+} \rightarrow Br^{3+} + Br^{2+}$, and P_1 peak in Br^{2+} ion and P_3 peak in Br^+ ion can be assigned to the channel of $Br_2{}^{3+} \rightarrow Br^{2+} + Br^+$. However, we cannot find a corresponding peak

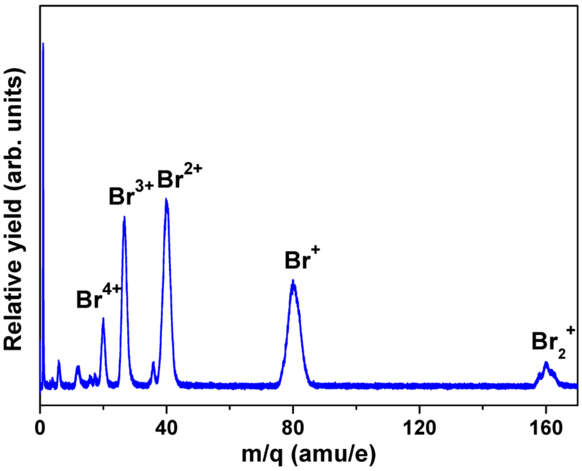

FIG. 1. (Color online) The mass spectrum of Br_2 molecules excited by the femtosecond laser pulse with the laser intensity of 1.5×10^{14} W/cm^2.

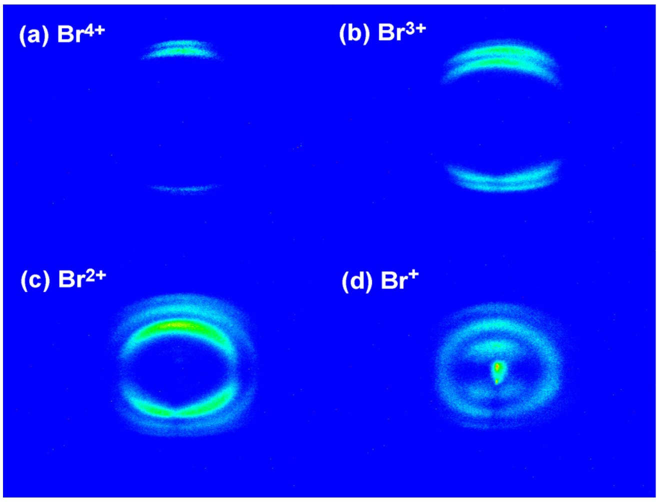

FIG. 2. (Color online) The sliced images of the fragmental ions Br^{4+} (a), Br^{3+} (b), Br^{2+} (c), and Br^+ (d).

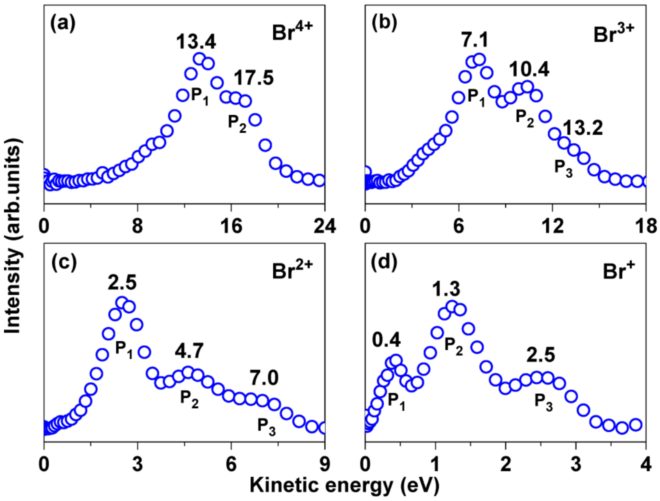

FIG. 3. (Color online) The kinetic energy diagrams of the fragment ions Br^{4+} (a), Br^{3+} (b), Br^{2+} (c), and Br^+ (d) calculated from Fig. 2.

TABLE I. Kinetic energy KE (eV) and photodissociation channel assignment in these fragmentation ions Br^{n+} (n = 1–4).

Fragment ions	KE(eV)	Channel assignment
Br^{4+}	17.5(±2.5)	$Br_2^{8+} \rightarrow Br^{4+} + Br^{4+}$
	13.4(±2.0)	$Br_2^{7+} \rightarrow Br^{4+} + Br^{3+}$
Br^{3+}	13.2(±1.1)	$Br_2^{7+} \rightarrow Br^{3+} + Br^{4+}$
	10.4(±0.7)	$Br_2^{6+} \rightarrow Br^{3+} + Br^{3+}$
	7.1(±0.5)	$Br_2^{5+} \rightarrow Br^{3+} + Br^{2+}$
Br^{2+}	7.0(±0.7)	$Br_2^{5+} \rightarrow Br^{2+} + Br^{3+}$
	4.7(±0.6)	$Br_2^{4+} \rightarrow Br^{2+} + Br^{2+}$
	2.5(±0.3)	$Br_2^{3+} \rightarrow Br^{2+} + Br$
Br^{+}	2.5(±0.2)	$Br_2^{3+} \rightarrow Br^{+} + Br^{2+}$
	1.2(±0.1)	$Br_2^{2+} \rightarrow Br^{+} + Br^{+}$
	0.4(±0.1)	$Br_2^{+} \rightarrow Br^{+} + Br$

in Fig. 3 to match P_2 peak in Br^{4+} ion based on Eq. (1), so we believe that P_2 peak in Br^{4+} ion should be assigned to the channel of $Br_2^{8+} \rightarrow Br^{4+} + Br^{4+}$. Similarly, these P_2 peaks in Br^{3+}, Br^{2+}, Br^{+} ions can be assigned to the channels of $Br_2^{6+} \rightarrow Br^{3+} + Br^{3+}$, $Br_2^{4+} \rightarrow Br^{2+} + Br^{2+}$, and $Br_2^{2+} \rightarrow Br^{+} + Br^{+}$, respectively. In addition, the P_1 peak in Br^{+} ion should be assigned to the channel of $Br_2^{+} \rightarrow Br^{+} + Br$. In this dissociation process, the parent ion Br_2^{+} in the electronic ground state is excited to the excited state by the laser-induced electron rescattering, and then dissociates into the fragment ion Br^{+}. All the photodissociation channels and corresponding kinetic energy in our experiment are listed in Table I.

These photodissociation channel assignments as mentioned above can be further confirmed by observing the angular distribution of a pair of fragment ions. For the two fragment ions from the same photodissociation channel, their angular distribution should be consistent. Figure 4 shows the angular distributions of these P_1 peaks in Br^{4+} (a), Br^{3+} (b), and Br^{2+} ions (c) (blue open circles), together with these P_3 peaks in Br^{3+} (a), Br^{2+} (b), and Br^{+} ions (c) (red open squares). As expected, the two fragment ions from the same photodissociation channel have the same angular distribution, which can well validate the above photodissociation channel assignments. Based on the above discussion and analysis, such a phenomenon can be found in our experiment that the photodissociation process is a charge symmetric or near-symmetrical dissociation, and thus the low (or high) charged fragment ions come from the photodissociation of low (or high) charged parent ions.

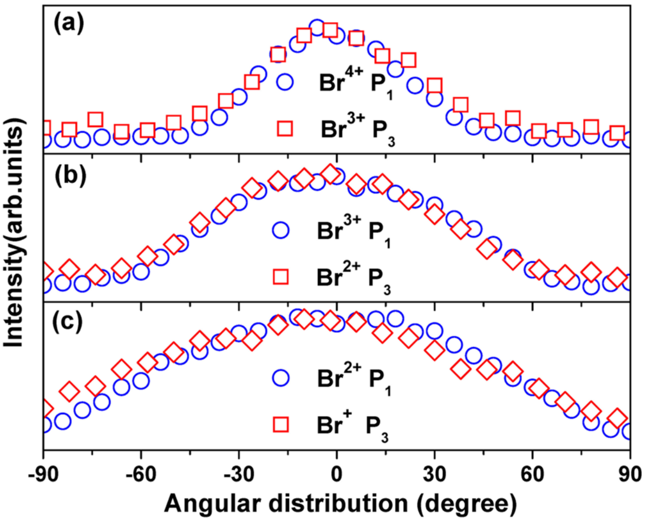

FIG. 4. (Color online) The angular distributions of these P_1 peaks in ions Br^{4+} (a), Br^{3+} (b), and Br^{2+} (c) (blue open circles), together with these P_3 peaks in ions Br^{3+} (a), Br^{2+} (b), and Br^{+} (c) (red open squares).

Since the low (or high) charged parent ions dissociate into the low (or high) charged fragment ions, the changes of the kinetic energy and angular distribution of these fragment ions Br^{n+} (n = 1–4) in Fig. 2 can be explained by considering the potential energy curves of these parent ions. Figure 5 shows the potential energy curves of the ground-state parent ions Br_2^{m+} (m = 1–8) calculated based on the level of MP2/6-311++G(d,p) in the GAUSSIAN 09 package. Since the main driving force for the chemical bond breaking of multicharged molecular ions is the Coulomb repulsive energy and the dissociation time of the molecule containing the heavy atoms (such as Br atom) is usually much longer than our pulse duration (~90 fs), these potential energy curves of parent ions Br_2^{m+} (m = 1–8) in Fig. 5 are considered under the field-free condition. As can be seen, the potential energy curves for the low charged parent ions Br_2^{m+} ($m = 1-4$) are relatively flat and even a potential well exists around the bond length of 2.3 Å; that is to say, these low charged parent ions Br_2^{m+} ($m = 1-4$) exist in the bound state, and the bound state of the parent ion Br_2^{2+} has been experimentally observed in the central spot of Fig. 2(d). However, the potential energy curves for the high charged parent ions Br_2^{m+} (m = 5–8) are monotonic decay; that is to say, the parent ions can spontaneously dissociate into fragment ions once they are formed by multiple ionization.

Since the potential energy of the parent ions is the result of the interaction of the chemical bonding energy and Coulomb repulsive energy, the experimental observation in Fig. 2 can be explained by analyzing the chemical bonding

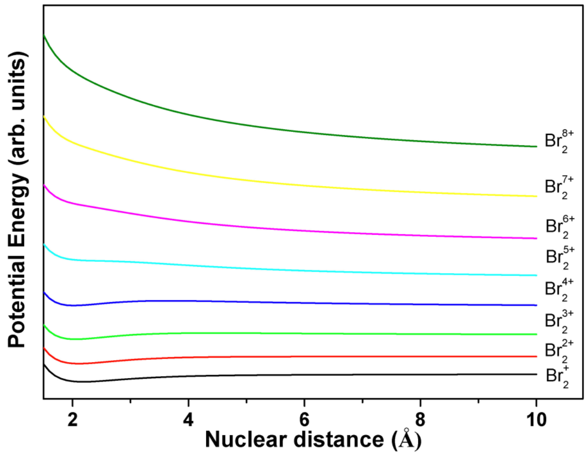

FIG. 5. (Color online) *Ab initio* potential energy curves of the parent ions Br_2^{m+} (m = 1–8) calculated by MP2/6-311G++(d,p) in GAUSSIAN 09 package.

TABLE II. Kinetic energy release KER (eV) and critical distance R_c (Å) for the eight photodissociation channels.

Channels	KER (eV)	R_c (Å)
(1,0)	0.4	
(1,1)	2.4	6.0
(1,2)	5.0	5.8
(2,2)	9.4	6.1
(2,3)	14.1	6.1
(3,3)	20.8	6.2
(3,4)	26.6	6.5
(4,4)	35.0	6.6

energy and Coulomb repulsive energy in these parent ions $Br_2{}^{m+}$ ($m = 1$–8). As can be seen in Fig. 5, the Coulomb repulsive energy is smaller than the chemical bonding energy for the low charged parent ions $Br_2{}^{m+}$ ($m = 2$–4), and therefore the photodissociation process is slow due to the existence of the potential well, which results in the low kinetic energy and wide angular distribution, and this phenomenon is similar to the predissociation process [40]. However, the Coulomb repulsive energy exceeds the chemical bonding energy for the high charged parent ions $Br_2{}^{m+}$ ($m = 5$–8); the chemical bond will be quickly broken, and thus the photodissociation process is fast, which results in the high kinetic energy and narrow angular distribution. In addition, the geometric alignment can occur in our experiment, which also has an effect on the angular distribution of these fragment ions. Since the Br_2 molecule in the strong femtosecond laser field is usually a sequential ionization, the higher charged parent ions are formed with a larger degree of anisotropy, and the anisotropy of the parent ions also contributes to the subsequent anisotropy of these fragment ions. This phenomenon has been experimentally observed in previous studies with the similarly experimental condition [32].

On the basis of the opinion that the dissociation process of multicharged molecular ions is uniquely dominated by the Coulomb repulsive energy and the Coulomb explosion occurs at the equilibrium nuclear distance, the kinetic energy releases for these photodissociation channels (4,4), (4,3), (3,3), (3,2), (2,2), (2,1), (1,1) can be respectively calculated as 100.2, 75.1, 56.3, 37.6, 25.0, 12.5, and 6.3 eV based on the Coulomb law of $\mathrm{KER} = 14.4PQ/R_e$, where R_e is the equilibrium nuclear distance of the Br_2 molecule at 2.3 Å. Here, the channel (P, Q) is corresponding to the dissociation process $Br_2{}^{(P+Q)+} \rightarrow Br^{P+} + Br^{Q+}$ in Table I. However, the kinetic energy releases for these channels in our experiment are respectively measured as 35.0, 26.6, 20.8, 14.1, 9.4, 5.0, and 2.4 eV, as shown in Table II. It is obvious that the measured kinetic energy release in the experiment is much smaller than the theoretical calculation. This experimental observation can be well explained by the enhanced ionization at a critical distance of the parent ions, where the ionization probability can be greatly enhanced when the bond length of the molecular ions elongates to a critical distance, which triggers the Coulomb repulsion, and then the Coulomb repulsive energy accelerates the fragmentation process. That is to say, the parent ions experience a bond elongation before the chemical bond breaking rather than explode at the equilibrium nuclear distance. According to the measured kinetic energy release in our experiment, the critical distances R_c for these dissociation channels can be calculated and are listed in Table II. It can be found that the Coulomb explosion in all these channels takes place around the critical distance of $R_c = 6.2$ Å. At this critical internuclear distance, the chemical bonding energy almost diminishes and only Coulomb repulsive energy dominates the dissociation process. Since the chemical bond breaking does not occur at the equilibrium nuclear distance but at a critical distance, the measured kinetic energy release in the experiment is always lower than the theoretical calculation. The enhanced ionization at a critical distance has been experimentally observed in molecular iodine (I_2), and it was shown that the elongation of the molecular ion to a critical distance $R_c = 3.8$ Å can drastically enhance the multiple ionization probability of the molecule [41].

IV. CONCLUSION

In summary, we have demonstrated the photodissociation process of Br_2 molecules irradiated by the intense femtosecond laser field using a dc-sliced ion velocity map imaging technique. Our experimental results showed that four fragment ions Br^{n+} ($n = 1$–4) can be observed, and the kinetic energy increases while the angular distribution decreases with the increase of the charge number. By observing the kinetic energy and angular distribution, it was shown that the low (or high) charged fragment ions come from the photodissociation of the low (or high) charged parent ions. Based on the potential energy curves of these parent ions that involve both the interaction of the Coulomb repulsive energy and the chemical bonding energy, the changes of the kinetic energy and the angular distribution in these fragment ions Br^{n+} ($n = 1$–4) could be well explained. Moreover, the experimental observation that the measured kinetic energy release is much smaller than the theoretical calculation was illustrated by the enhanced ionization at a critical distance. We believe that our results are useful for exploring the molecular photodissociation process under the ultrafast intense laser field and have potential applications in understanding and controlling the photochemical reaction.

ACKNOWLEDGMENTS

This work was partly supported by National Natural Science Fund (Grants No. 11474096 and No. 51132004) and Shanghai Municipal Science and Technology Commission (Grant No. 14JC1401500).

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