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Enhanced ionization of vibrational hot carbon disulfide molecules in strong femtosecond laser fields*

Wan-Long Zuo(左万龙)[1,2], Hang Lv(吕航)[1], Hong-Jing Liang(梁红静)[1], Shi-Min Shan(单石敏)[1], Ri Ma(马日)[1,†], Bing Yan(闫冰)[1], and Hai-Feng Xu(徐海峰)[1,‡]

[1] *Institute of Atomic and Molecular Physics, Jilin University, Changchun* 130012, *China*
[2] *Anhui Provincial Key Laboratory of Optoelectric Materials Science and Technology, Anhui Normal University, Wuhu* 241000, *China*

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By using a heated molecular beam in combination with a time-of-flight mass spectrometer, we experimentally study the ionization of vibrational-hot carbon disulfide (CS_2) molecules irradiated by a linearly polarized 800-nm 50-fs strong laser field. The ion yields are measured in a laser intensity range of 7.0×10^{12} W/cm^2–1.5×10^{14} W/cm^2 at different molecular temperatures of up to 1400 K. Enhanced ionization yield is observed for vibrationally excited CS_2 molecules. The results show that the enhancement decreases as the laser intensity increases, and exhibits non-monotonical dependence on the molecular temperature. According to the calculated potential energy curves of the neutral and ionic electronic states of CS_2, as well as the theoretical models of molecular strong-field ionization available in the literature, we discuss the mechanism of the enhanced ionization of vibrational-hot molecules. It is indicated that the enhanced ionization could be attributed to both the reduced ionization potential with vibrational excitation and the Frank–Condon factors between the neutral and ionic electronic states. Our study paves the way to understanding the effect of nuclear motion on the strong-field ionization of molecules, which would give a further insight into theoretical and experimental investigations on the interaction of polyatomic molecules with strong laser fields.

Keywords: strong field ionization, vibrational excited, time-of-flight-mass spectrum

PACS: 33.80.RV **DOI:** 10.1088/1674-1056/27/6/063301

1. Introduction

Compared with atoms, the interaction of molecules with a strong laser field is much more complicated and exhibits many peculiar behaviors, which can be attributed to the complexity in molecular structure and additional nuclear degree-of-freedom. The effect of molecular properties on the strong-field physical process has attracted increasing research interest during the past two decades. It has been demonstrated that molecular geometry,[1,2] alignment and orientation of molecular axis with respect to the laser polarization,[3,4] as well as the structure of molecular orbitals,[5,6] play crucial roles in various molecular strong-field physical processes, such as strong field ionization and fragmentation, high-order harmonic generation (HHG), above-threshold ionization (ATI), and non-sequential double ionization (NSDI).

One particular issue is the effect of the nuclear vibrational motion on the strong-field ionization of molecules. For diatomic molecules, since there is only the stretching vibrational mode, the effect could be reasonably considered as a consequence of the dependence of ionization on the internuclear distance R. Various experimental and theoretical studies have shown that the tunneling ionization probability of diatomic molecules increases as R increases, and reaches a maxima at a critical internuclear distance R_c, which is well known as the charge resonance enhanced ionization (CREI).[7–9] It is indicated that CREI highly depends on molecular symmetry as well as molecular orbitals from which the electron is ionized.[10] The pronounced R-dependent ionization could be used as an interesting method of forming the coherent vibrational wavepackets in ground electronic states with strong laser fields.[11–13] Theoretical studies have predicted that for some diatomic molecules such as N_2[14] and H_2,[15] the HHG efficiency could be improved significantly by vibrational motion, which is also attributed to the enhanced ionization. Furthermore, by numerically solving the time-dependent Schrödinger equation for H_2^+, Kästneret *et al.* have shown that the ionization and dissociation channels of the molecule could be steered if the molecule is prepared in a specific vibrational state.[16]

For polyatomic molecules, to the best of our knowledge, there are no experimental studies concerning vibrational-dependent strong-field ionization. Using a wavefunction-modified Ammosov–Delone–Krainov (ADK) method, Brichta *et al.* calculated the ionization rate of triatomic molecules CO_2 (and diatomic molecules H_2 and N_2) as a function of the initial vibrational level.[17] Their results indicated that the enhanced

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†Corresponding author. E-mail: rma@jlu.edu.cn
‡Corresponding author. E-mail: xuhf@jlu.edu.cn

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ionization is sensitive to the field-free potential energy surfaces of neutral and cationic ground electronic states. In their study, only the symmetric stretching vibrational mode is considered, thus the case of CO_2 is treated in the same way as a diatomic molecule system. It remains a challenging task to understand the interaction of strong laser fields with vibrational excited polyatomic molecules, due to the increasing of the vibrational degree-of-freedom, which is $3n$-5 (or $3n$-6) for linear (or nonlinear) molecules (n is the number of atoms contained in a molecule).

Here we present the first experimental study on the enhanced ionization of vibrational hot polyatomic molecules in a strong 800-nm femtosecond laser field. Triatomic molecules CS_2 are investigated in the study, which exhibits four vibrational normal modes, i.e., symmetric stretching (ν_1), two-fold degenerate bending (ν_2), and asymmetric stretching (ν_3). The vibrational frequencies in the ground electronic state $X^1\Sigma_g^+$ are 659.23, 396.55, and 1537.63 cm^{-1} respectively.[18] Using a heated molecular beam, we can prepare the molecules with a vibrational population, mostly in the ν_1 and ν_2 excited states because of their low vibrational frequencies. We measure the ionization yields at different molecular temperatures (corresponding to different initial vibrational populations) in a laser intensity range of 7.0×10^{12} W/cm^2–1.5×10^{14} W/cm^2. The results show the enhanced ionization for vibrational hot CS_2 molecules in a strong laser field. Our study should shed light on the strong-field ionization of polyatomic molecules with nuclear vibrational motion.

2. Experiment

The experimental setup was similar to that described in our previous study,[19] except that a heated molecular beam based on the design of Chen and coworkers[20] was used in the present study to increase the initial vibrational population of CS_2 molecules. Briefly, CS_2 molecules seeded in Ar were introduced into the vacuum chamber through an effusive nozzle. The CS_2/Ar gas mixture from the nozzle was then expanded into an SiC tube (31-mm length, 1-mm i.d. and 2-mm o.d.), which was attached to an alumina shield to isolate the tube thermally and electrically from the face-plate of the nozzle. The SiC tube was heated via two graphite electrodes that were press-fitted at the ends of the tube. The temperature was measured using a remote portable infrared thermometer (IR-HS, CHINO). The tube could be maintained at up to 1500 K for more than five hours with a derivation of $\pm$10 K. Upon heating, CS_2 molecules could be populated at the vibrational excited states of the ground $X^1\Sigma_g^+$ state according to the Boltzmann distribution. The stagnation pressure of CS_2/Ar mixture was kept at approximately 1.5 atm (1 atm = 1.01325×10^5 Pa), and the operating pressure in the chamber was about 3×10^{-4} Pa.

The vibrational hot molecules were interacted with strong laser pulses at a 5-cm distance from the exit of the SiC tube. The laser system was a Ti:sapphire chirped-pulse amplified laser (Libra-USP-HE, Coherent Inc.) with a central wavelength of 800-nm, pulse duration of 50 fs, repetition rate of 1 kHz, and maximum pulse energy of 4 mJ. A half-wave plate and a Glan prism were inserted into the laser beam to vary the laser intensity continuously. The laser pulses were focused onto the molecular beam by a lens with the focal length of 25 cm to ionize the CS_2 molecules. The peak intensity of the focused laser pulse was calibrated by comparing the measured saturation intensity of Xe with that calculated by the Ammosov–Delome–Krainov (ADK) model.[21] The produced cations from strong-field ionization were detected by a linear time-of-flight (TOF) mass spectrometer operated under the Wiley–McLaren condition. All the cations were extracted, accelerated, and finally detected by a dual microchannel plate (MCP) detector at the end of the flight about 55 cm. Mass-resolved ion signals from MCP were recorded using a digital oscilloscope (Tektronix TDS 3054B) and sent to a PC for analysis. All experimental data were normally averaged over 10^3 laser shots.

3. Results and discussion

3.1. Enhanced strong-field ionization of vibrational-hot CS_2 molecules

Because of their relatively low frequencies of the symmetric stretching and bending vibrational modes (ν_1 = 659.23 cm^{-1} and ν_2 = 396.55 cm^{-1}), CS_2 molecules can be efficiently populated in the vibrational excited states upon heating the molecule beam. Assuming a Boltzman distribution in the molecular beam, more than 80% of CS_2 molecules will be populated at the excited vibrational states for T = 140 K, while the molecules are mainly populated at the ground vibrational states without heating. To retrieve strong field ionization of vibrationally excited molecules, the effect of thermal motion on heating, which would cause the molecule/atom-beam density in the laser interaction region to change, should be eliminated. In our study, we address this issue by calibrating ion yields to that of atom Ar^+, the intensity of which only depends on the thermal motion. Typical TOF mass spectra of CS_2 molecules irradiated by linearly polarized 800-nm laser fields at an intensity of 7.0×10^{13} W/cm^2 are presented in Fig. 1, without and with heating the molecules at T = 1400 K respectively. As shown in the figure, the dominant peak appearing in the TOF mass spectra is due to the singly charged parent ions CS_2^+, along with apparent daughter ions S^+ and CS^+, as well as doubly charged parent ions CS_2^{2+}, which appear at a laser intensity larger than 4.0×10^{13} W/cm^2. In the present study, we focus on the strong-field single ionization of vibrational-hot CS_2 molecules in a laser intensity range of

7.0×10^{12} W/cm^2–1.5×10^{14} W/cm^2. It should be mentioned that the fragment ions S^+ and CS^+ are mainly produced by the dissociative ionization of CS_2. Thus, for the more accurate representation of the strong field single ionization, we use the total ion yields of CS_2^+, S^+, and CS^+ ions (instead of the yield of only CS_2^+) as the single ionization yield in the following discussion.

It can be seen from Fig. 1 that although there is no significant change in the overall feature of each TOF spectrum, the peak intensity apparently increases upon heating the molecular beam. In Fig. 2, we further present the measured ion yields as a function of laser intensity with heating the molecular beam at 1400 K. For comparison, the results without heating the molecular beam are also presented in the figure. Because the vibration of a molecule cannot be cooled sufficiently in a diffusive molecular beam, the temperature is tentatively set to be at 300 K when no heating is applied. At each temperature, the ionization yield increases as the laser intensity increases and turns to be saturated at about 1.0×10^{14} W/cm^2, which is in agreement with previous studies.[20] Clearly, within the laser intensity range used in the study, the probability of strong field ionization is enhanced for vibrational-hot CS_2 molecules.

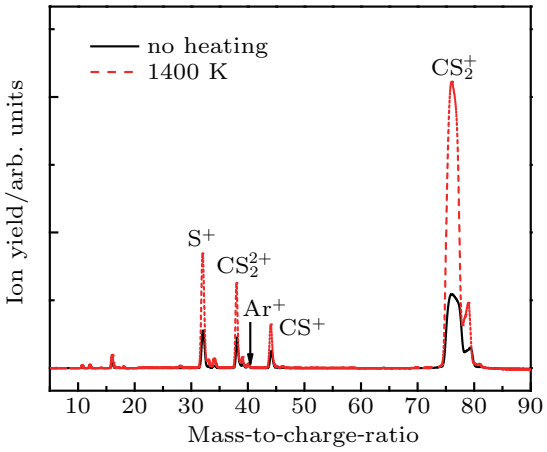


Fig. 1. (color online) Typical TOF mass spectra of CS_2 molecules irradiated by a linearly polarized 800-nm laser field at an intensity of 7.0×10^{13} W/cm^2 without heating the molecules (solid black) at $T = 14$ K (dashed red).

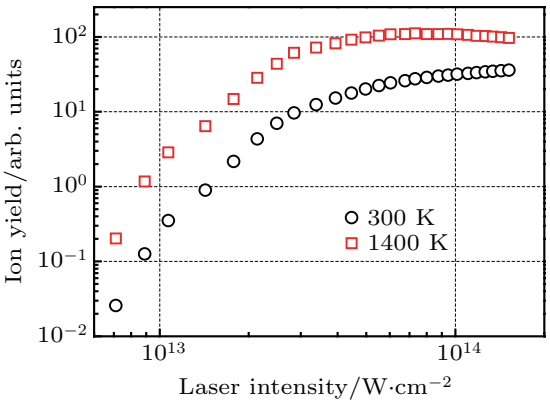


Fig. 2. (color online) Measured ion yields as a function of laser intensity at molecular beam temperatures of 300 K (black circle) and 1400 K (red square).

In order to further study the strong field ionization of vibrational hot CS_2 molecules, we show in Fig. 3 the dependence of the enhancement ratio on the laser intensity for different T values. Here, the enhancement ratio refers to the ratio of the ion yield of hot molecules to that without heating the molecular beam. Again, one can see that the enhancement ratio is larger for higher temperature of the molecular beam. In addition, the enhancement ratio gradually decreases as the laser intensity increases for either of the heating temperatures of the molecular beam. Figure 4 shows the measured ion yields at different T values of 300 K, 600 K, 900 K, 1200 K, and 1400 K. The laser intensity is fixed at 7.0×10^{13} W/cm^2, and the ion yield at each temperature is normalized to that of 1400 K. It is interesting to note that the ionization does not increase monotonically with the increase of temperature, which exhibits a plateau at 900 K–1200 K as shown in Fig. 4.

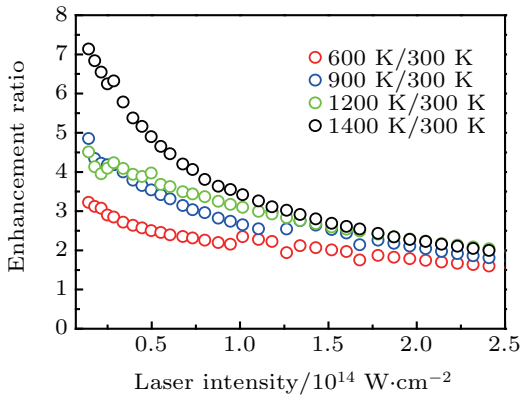


Fig. 3. (color online) Variations of enhancement ratio with laser intensity for different T values (see text for details).

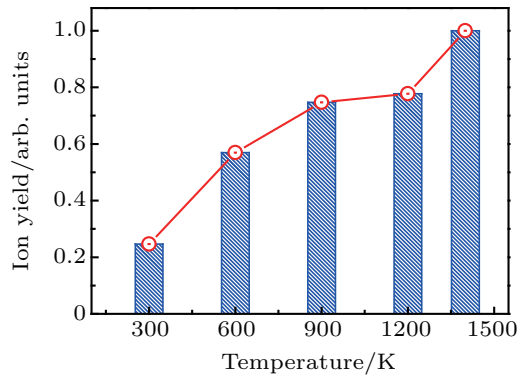


Fig. 4. (color online) Ion yields at different T values of 300 K, 600 K, 900 K, 1200 K, and 1400 K, with laser intensity fixed at 7.0×10^{13} W/cm^2 and the ion yield at each temperature normalized to that of 1400 K.

3.2. Discussions

For different laser intensities, the strong-field ionization could be divided into two regimes, i.e., multiphoton ionization (MPI) and tunneling ionization (TI), according to the adiabatic Keldysh parameter γ[22]

$$\gamma = \sqrt{\frac{I_p}{1.8 \times 10^{-13} \times I \times \lambda^2}}, \quad (1)$$

where I_p, I, and λ represent the field-free ionization potential (in unit eV), the peak intensity of the laser pulse (in units W/cm^2), and the laser wave-length (in unit μm), respectively. If $\gamma \gg 1$, MPI will be dominant, while for $\gamma \ll 1$, the ionization is mainly in the TI regime. In our study, laser intensities are in the range of 7.0×10^{12} W/cm^2–1.5×10^{14} W/cm^2, corresponding to the Keldysh parameter γ of $3.47-0.75(I_p(CS_2) = 10.078$ eV),[18] indicating that both MPI and TI could be attributed to the strong field ionization of CS_2.

For MPI, the ionization of CS_2 occurs as vertical transition happens from the neutral ground electronic state to the ionic ground electronic state, $CS_2^+(X^2\Pi_g;v_1^+,v_2^+,v_3^+) \leftarrow CS_2(X^1\Sigma_g^+;v_1,v_2,v_3)+nh$. The vibrational-state-dependent ionization is usually understood by using the adiabatic approximation and the Frank–Condon principle. The relative ionization probability from different initial vibrational states in the same electronic state depends on the overlap between the vibrational wavefunctions of the initial neutral electronic state and the final ionic electronic state, $|\langle\psi_{v^+}|\psi_v\rangle|$, which is determined by the shape of potential energy surfaces and the equilibrium geometries of the electronic states. We perform a high-level *ab initio* calculation on the $CS_2(X^1\Sigma_g^+)$ and $CS_2^+(X^2\Pi_g)$ states via the MOLPRO 2012 program package.[23,24] The ground state $X^1\Sigma_g^+$ of CS_2 is linear with respect to the dominant electronic configuration of $(1\sigma_g)^2(1\sigma_u)^2(1\pi_u)^4(1\pi_g)^4\ (2\sigma_g)^2(2\sigma_u)^2(3\sigma_g)^2(3\sigma_u)^2(4\sigma_g)^2(5\sigma_g)^2(4\sigma_u)^2(6\sigma_g)^2(5\sigma_u)^2(2\pi_u)^4(2\pi_g)^4$. The $X^2\Pi_g$ state of CS_2^+ is also a linear state that corresponds to the removal of an electron from the highest occupied molecular orbital, $2\pi_g$. Figure 5 shows the potential energy curves (PECs) of $CS_2(X^1\Sigma_g^+)$ and $CS_2^+(X^2\Pi_g)$ versus C–S bond (Fig. 5(a)) and ∠S–C–S angle (Fig. 5(b)), which are calculated using the method of coupled-cluster with singles, doubles, and perturbative triples (CCSD(T)) with the aug-cc-pwcvQZ basis set. The results show that the shape of the PEC and the equilibrium geometries of $CS_2^+(X^2\Pi_g)$ are almost identical to those of $CS_2(X^1\Sigma_g^+)$. This indicates that the diagonal elements (for $\Delta v = v^+ - v = 0$) are dominant in the overlap integral matrix $|\langle\psi_{v^+}|\psi_v\rangle|$, and that from the ground vibrational state $(0,0,0)$ is the maximum. Indeed, the high-resolution VUV-PFI-PE (vacuum ultraviolet-pulsed field ionization-photoelectron) study has demonstrated that the $CS_2^+(X^2\Pi_g;0,0,0) \leftarrow CS_2(X^1\Sigma_g^+;0,0,0)$ band is the strongest in the PE spectrum.[18] Thus, if the MPI mechanism is dominant, the ionization probability would decrease upon initial vibrational excitation of CS_2 molecules, which is opposite to the experimental observations.

We now turn to discuss the vibrational-state-dependent ionization of CS_2 in the TI regime. According to the quasi static theory of Ammosov, Delome, and Krainov (ADK model), which can well describe the strong-field ionization of atoms in the TI regime, the ionization rate will increase if the ionization potential (I_p) decreases. Using a molecular wavefunction modified ADK model (MO-ADK),[25] several studies have indicated that the enhanced ionization could also be attributed to the reduced I_p values of vibrational excited molecules.[17,25] The energy difference between the neutral $CS_2(X^1\Sigma_g^+)$ and ionic $CS_2^+(X^2\Pi_g)$ states is depicted as the dashed lines in Fig. 5, which indicates the reduced I_p values for the excitation of either the symmetric stretching (along the C–S bond (Fig. 5(a)) or the bending (along the ∠S–C–S angle (Fig. 5(b)) vibrational states of $CS_2(X^1\Sigma_g^+)$. Thus it is not a surprise that the ionization probability of vibrational-hot CS_2 molecules are observed experimentally to be enhanced according to the ADK model. In addition, a recent theoretical study on diatomic molecules predicts that the enhancement ratio is controlled by a parameter ζ, which is in inverse proportion to the laser intensity.[26] That is, the enhancement of the ionization rate for a vibrational excited state of a molecule is more pronounced at low laser intensity, which is consistent with our experimental results as shown in Fig. 3.

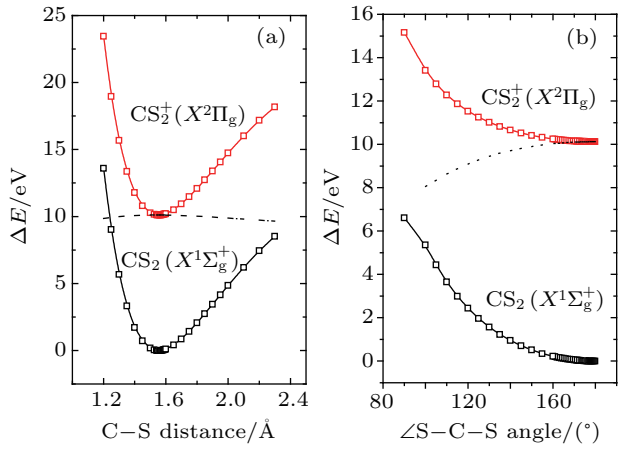


Fig. 5. (color online) PECs of $CS_2(X^1\Sigma_g^+)$ and $CS_2^+(X^2\Pi_g)$ versus (a) C–S bond and (b) ∠S–C–S angle, respectively.

According to the above discussion, it is indicated that the enhanced ionization of vibrational-hot CS_2 molecules can be attributed to the increased tunneling ionization probability due to the reduced I_p values for vibrational excited states. On the other hand, our results imply that the enhancement is not monotonically increased as vibrational excitation. As shown in Fig. 4, as the temperature increases from 900 K to 1200 K, the ion yield remains almost unchanged. This observation is hard to understand by the ADK model, since I_p decreases monotonically upon vibrational excitation (see the dashed lines in Fig. 5). Alternatively, based on the MO-ADK model, Zon *et al.* proposed an anti-stokes-enhanced tunneling ionization (ASETI) method to investigate the effect of vibrational motion on the strong field ionization of molecules.[27–29] In their method, the Dyson orbital wavefunctions including nuclear motion are used as the molecular wavefunctions, and

the ionization rate of molecules in the TI regime is described by the combination of the Frank–Condon factors between the neutral and ionic electronic states and the ADK ionization rate, which is dependent on the ionization potential.[27–29] According to this ASETI model, we can qualitatively understand the experimental observation in Fig. 4. As we have discussed above, the effect of the Frank-Condon factors will result in a decreasing ionization yield of vibrational hot CS_2 molecules, in opposite to the prediction of the ADK model, which could slow down the enhancement for higher vibrational excitation. It is noted that as the temperature is further increased from 1200 K to 1400 K, the ionization yield starts to increase again (see Fig. 4), which may be due to the fact that the population on the asymmetric stretching (ν_3) vibrational excited states occurs at higher temperature of CS_2 molecules.

We should mention that it would be far from completely understanding the mechanism of the strong-field ionization of vibrationally excited polyatomic molecules. Several essential issues should be addressed which are not possible to accomplish in the present study, for example, accurate molecular wavefunctions including nuclear motion, anharmonic effect for vibrational excited states, coupling between different vibrational modes, etc. The present experimental study indicates the apparent effect of vibrational excitation on strong field ionization of polyatomic molecules, and would stimulate further studies to develop a new theoretical model to reveal the underlying physical mechanism.

4. Conclusions

In this work, we have shown experimentally that by heating the molecular beam to populate CS_2 molecules in the vibrational excited states, the ionization yield is enhanced significantly after interacting with a linearly polarized 50-fs 800-nm strong laser field. The enhancement ratio decreases as the laser intensity increases in a range of 7.0×10^{12} W/cm^2–1.5×10^{14} W/cm^2, and increases non-monotonically as the molecular temperature increases from 300 K to 1400 K. The enhanced ionization of vibrational-hot molecules can be attributed to the reduced ionization potential of the initial vibrational state, according to the MO-ADK model. It is also discussed that the Frank–Condon factors between the neutral and ionic electronic state can also affect the ionization probability in the tunneling ionization regime. Our study sheds light on the effect of vibrational motion on the interaction between polyatomic molecules and the strong laser field.

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