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Effect of laser polarization on strong-field ionization and fragmentation of nitrous oxide molecules*

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Ionization of molecules in femtosecond laser fields is the most fundamental and important step of various strong-field physical processes. In this study, we experimentally investigate strong field ionization of linear N_2O molecules using a time-of-flight mass spectrometer in 800-nm laser fields. Yields of the parent ion and different fragment ions are measured as a function of laser intensity in the range of 2.0×10^{13} W/cm^2 to 3.6×10^{14} W/cm^2. We also investigate the dependence of strong field ionization and dissociation of N_2O on laser ellipticity and polarization direction. The significant role of laser induced electron re-collision in the formation of highly charged fragment ions is proved. The physical mechanism of strong field ionization and fragmentation is discussed, based on our experimental results.

Keywords: strong laser field, re-collision, angular distribution, nitrous oxide

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1. Introduction

Following Mourou and Strickland's ground-breaking invention of chirped pulse amplification (CPA) to generate high-intensity, ultra-short optical pulses,[1] studies on the interaction of an atom/molecule with strong laser fields have developed rapidly, and a variety of new non-perturbative and highly non-linear phenomena have been revealed.[2–4] Tunneling ionization is viewed as a fundamental process in atomic/molecular strong-field physics, in which the electron is freed by tunneling the barrier formed by combination of laser electric fields and Coulomb potential of atoms and molecules. Many strong field physical processes are induced by tunneling ionization, such as non-sequential double/multiple ionization (NSD(M)I),[5,6] high-order harmonic generation (HHG),[7] high-order above threshold ionization (HATI),[8,9] and neutral high-Rydberg excitation.[10,11] Therefore, understanding the strong-field ionization process is of vital importance in many research fields ,including attosecond pulse generation,[12,13] molecular orbital imaging,[14] and vacuum extreme ultraviolet light sources.[15]

Many atomic strong field processes can be qualitatively understood in the frame of so-called three-step re-collision scenario:[16,17] an electron in an atom first tunnels out, is then accelerated by the laser electric field, and finally as the laser electric field is reversed, the free electron can be driven back and collide with the nucleus to produce various strong field physical phenomena. According to this scenario, it is expected that re-collision will be suppressed as we increase the laser's ellipticity due to the additional drift momentum spread of the returning electron wavepacket.[18] Thus, a dramatically decreasing probability of the re-collision induced processes (NSDI, HATI, HHG) is observed in an elliptically polarized laser field.[19] Meanwhile, the trajectory of the returning electron can be modulated with laser ellipticity, which could play a significant role in revealing and controlling the dynamics of strong laser-driven electron re-collision processes.[20,21]

Comparing to atoms, molecules have much more complex ionization processes due to their diverse geometric structure, additional nuclear degrees of freedom, and complicated molecular orbitals. Indeed, previous studies have proven that the multi-center interference,[22] multiple orbitals,[23] and molecular nuclear motions[24] can affect strong field molecular ionization, which stimulate more elaborate experimental and theoretical investigations. In addition, molecules can undergo various processes that accompany strong field ionization, including excitation, dissociative ionization, Coulomb explosion, and so on. The role of laser-driven electron re-collision in the strong field ionization/dissociation of molecules is far from completely understood, especially for polyatomic molecules.

Here, we present an experimental study on the ionization/dissociation of nitrous oxide (N_2O) in both linearly and elliptically polarized strong laser fields. Early studies on N_2O have mainly focused on dissociation and Coulomb explosion processes of parent ions at highly charged states in an intense laser field. In 1991, Frasinski *et al.* distinguished

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the complex dynamics of multiphoton multiple ionization of N_2O molecules using the three-dimensional (3D) covariance method.[25] The coincidence momentum imaging method was used to discriminate the concerted and sequential Coulomb explosion processes of N_2O molecules.[26] Identification of two-body and three-body fragmentation pathways of Coulomb explosion of N_2O in an intense laser field was also investigated by high-resolution time-of-flight spectroscopy.[27] By combining the femtosecond multi-pulse length spectroscopy technique with 3D-ion-momentum coincidence method, Karimi *et al.* studied the dynamics of N_2O in the ionization process and found two stepwise dissociation pathways of N_2O^{3+}.[28] Recently, single and double ionizations of N_2O in short elliptically polarized 800-nm laser pulses were imaged by COLTRIMS technique, which showed that the shape of ionization orbits has a major effect on the ionization process of N_2O molecules.[29] In our study, we investigate the ionization and dissociation of N_2O in a 50-fs 800-nm laser field using a time-of-flight (TOF) mass spectrometer. We focus on the dependence of yields of parent and fragment ion on laser polarization, as well as laser intensity in the range of 2×10^{13} W/cm^2 to 3.6×10^{14} W/cm^2. Based on our experimental results, we discuss the role of tunneling electron induced re-collision in strong field ionization and dissociation of N_2O molecules.

2. Experimental setup

The experimental setup used for ionization/dissociation in femtosecond laser fields is similar to those described in our previous studies.[30,31] The laser system used in the study is a chirped-pulse-amplified Ti: sapphire femtosecond laser, with a repetition frequency of 1 kHz, a central wavelength of 800-nm, a pulse duration of 50-fs, and a maximum pulse energy of 4 mJ. The laser's intensity was continuously changed by rotating a half waveplate in front of a Glan prism. A quarter waveplate was used to control the state of the laser polarization which was inserted in the laser beam next to the Glan prism. Before the laser beam entered the vacuum chamber to interact with molecules, the laser beam was focused through a lens with a focal length of 25 cm.

N_2O molecules were introduced directly into the chamber through a leak valve with a stagnation pressure of ~1 atm. The operating pressure in the chamber was below 1×10^{-5} Pa. A standard linear TOF mass spectrometer operating under the Wiley–McLaren condition was used to detect the ions produced by strong field ionization/dissociation in strong 800-nm laser fields. All the ions were extracted, accelerated, and finally detected by a dual-microchannel-plate detector after a 45-cm long field-free flight. For the measurement of ion angular distributions, a slit with a 0.5 mm width was mounted at the end of the flight tube to ensure that only those ions produced with initial velocity along the flight tube were detected. An acquisition card (National instruments, PXIe-5162) was used to record and analyze mass-resolved ion signals. All experimental data were normally averaged over 5000 laser shots.

3. Results and discussion

Using a TOF mass spectrometer, we measured yields of different ions produced by irradiating N_2O molecules with a 50-fs 800-nm strong laser field, and investigated the dependence on laser polarization and intensity in the range of 2.0×10^{13} W/cm^2 to 3.6×10^{14} W/cm^2. In our experiments, we observed singly-charged parent ion (N_2O^+) and various fragment ions including diatomic molecular fragments (NO^+ and N_2^+) and atomic fragments (O^{n+} and N^{m+}, with $n=1$, 2 and $m=1$–3). Doubly-charged and multiple-charged parent ions were not observed in the whole laser intensity range, which could be attributed to their short lifetime that cannot survive the flight-time to the detector (for instance, the lifetime of the metastable state of N_2O^{2+} is only 660 ± 70 ns[32]).

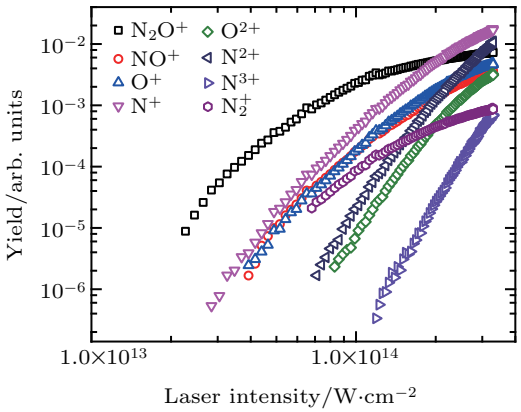


Fig. 1. Yields of parent and fragment ions of N_2O irradiated by linearly polarized laser fields in the laser intensity range of 2.0×10^{13} W/cm^2 to 3.6×10^{14} W/cm^2.

In Fig. 1, we show the yields of N_2O^+ and different fragment ions as a function of laser intensity. It can be seen from the figure that compared to singly-charged fragment ions that appear at a relatively low laser intensity, double charged ions (O^{2+} and N^{2+}) begin to appear at a stronger laser intensity of 7.0×10^{13} W/cm^2, and N^{3+} ions are not detectable until the laser intensity reaches 1.2×10^{14} W/cm^2. This is easy to be understood because singly-charged fragments can be produced by dissociative ionization of N_2O^+ with absorption multi-photons in the strong laser fields, while highly-charged fragments have to be formed by dissociation or Coulomb explosion of multiple-charged parent ions. We note that at a laser intensity higher than 1.0×10^{14} W/cm^2, the corresponding Keldysh parameter $\gamma<1$ ($\gamma=\sqrt{I_p/(2U_p)}$, where I_p represents the ionization potential of the target, $U_p=e^2E^2/(4m\omega^2)$ rep-

resents the ponderomotive potential[33]), indicating that tunneling ionization would be dominant in the strong field ionization of N_2O. It has already been demonstrated that for atom Xe, which has a similar ionization potential ($I_p = 12.13$ eV) to that of N_2O molecule ($I_p = 12.889$ eV), double (multiple) ionization in 800-nm strong laser fields with the intensity range used in the present study occurs mainly via tunneling electron re-collision induced NSD(M)I mechanism.[34] Thus, it can be expected that re-collision would play a significant role in the double (multiple) ionization of N_2O, thus the formation of highly-charged fragments.

To further investigate the mechanism of the formation of fragment ions by strong field ionization of N_2O, we measured the yield dependence of different ions on laser ellipticity. We first show the TOF mass spectra irradiated by 50-fs 800-nm laser fields at a peak intensity of 2×10^{14} W/cm^2 with linear polarization (LP) or circular polarization (CP) in Fig. 2. It is obvious that comparing to the parent ions and singly-charged fragment ions, the O^{2+} and N^{2+} ions exhibit a stronger laser-polarization dependence, while N^{3+} even completely disappears in the CP laser field, which indicates strong suppression of highly-charged fragment ions in the circular polarized laser field.

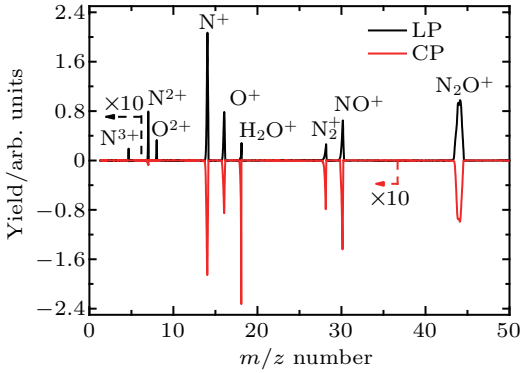


Fig. 2. TOF mass spectra of N_2O irradiated by 800-nm laser fields at a peak intensity of 2×10^{14} W/cm^2, with linear or circular polarization. Parts of the mass spectra which are pointed out by the black and red dotted arrows in the figure are magnified by ten times.

We then study the ellipticity dependence of the strong field ionization/fragmentation of N_2O. A slightly deduce of the yield of parent ion N_2O^+ in elliptically polarized laser fields could be attributed to the decrease of the amplitude of the electric field. Thus, to take into account the small decrease of ionization probability caused by the reduced electric field in elliptically polarized laser, we employed the ratio between the yields of each fragment ion to those of the singly charged parent ion N_2O^+. The results are shown in Fig. 3. For each ion, the ratios at different ellipticities were normalized to the maximal value at linear polarization ($\varepsilon = 0$). It can be seen that the ratio decreases as the laser ellipticity is increased. The ellipticity dependence is stronger for the highly charged fragment ions. While the ratio of singly-charged fragment ions in the CP laser fields is about 15%–25% of that in the LP laser fields, for N^{3+} it dramatically drops to zero at a small ellipticity ($\varepsilon = 0.3$).

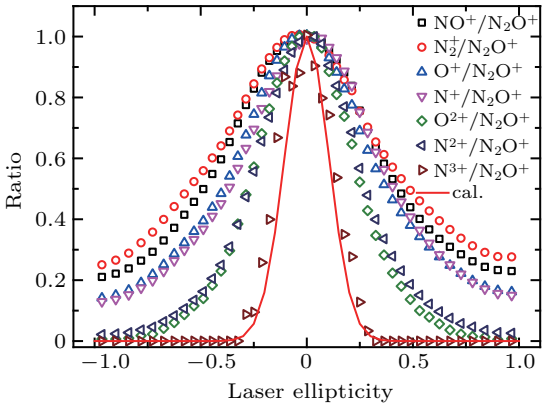


Fig. 3. Ellipticity dependence of the yield ratio between the fragment ion and N_2O^+ with laser field intensities of 1.4×10^{14} W/cm^2. The red solid line is the result obtained using the equation deduced from the three-step re-collision model.

As previously mentioned, the highly-charged fragments are mainly formed by dissociation or Coulomb explosion of multiple-charged parent ions from NSD(M)I of N_2O. Thus, the strong ellipticity dependence can be explained in the frame of the three-step re-collision model. For NSD(M)I, the tunneling electron should return to the tunnel exit and re-collide with the ionic core to release other electrons. However, in an elliptically polarized laser field, only those tunneling electrons whose additional drift motion induced by the laser field is compensated by the initial vertical velocity gained in the tunneling process would collide with the ionic core and induce the NSD(M)I. According to this scenario, the ellipticity dependence can be described using (in atomic units)[19]

$$\frac{I(\varepsilon)}{I(\varepsilon=0)} \approx \exp\left(-\frac{1.25\sqrt{2I_p}F}{\omega_0^2}\varepsilon^2\right),$$

where F represents the electric field, ε represents the laser ellipticity, and ω_0 represents the frequency of the driving laser.

The ellipticity dependence of the re-collision probability obtained using the equation deduced from the three-step re-collision model is also presented in Fig. 3 (solid-red line). Good agreement with the experimental results of N^{3+} fragment is observed, indicating the significant role of tunneling-plus-recollision in formation of highly-charged ions in strong field ionization of N_2O molecules. Meanwhile, some deviation from the prediction of the three-step re-collision model is also apparent for the doubly-charged fragment ions. This can be attributed to the fact that, at the intensity of 1.4×10^{14} W/cm^2, sequential ionization occurs (for reference, see Xe results in Ref. [34]), which can also lead to formation of N^{2+} or O^{2+}, thus a weaker dependence on laser ellipticity as observed in Fig. 3 is expected.

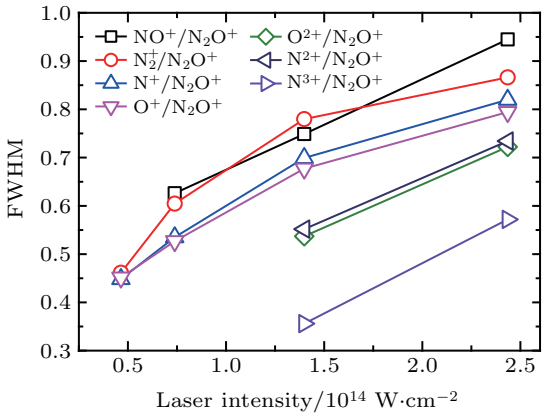


Fig. 4. Full width at half maximum of the ellipticity dependence of each ion measured at different intensities of the 800-nm laser field.

We further show the full width at half maximum (FWHM) of the ellipticity dependence of each ion measured at different laser intensities in Fig. 4. Here, the FWHM is obtained by Gaussian fitting of the experimental results in Fig. 3. For each laser intensity, when the fragment ion is in a more highly charged state, the FWHM is smaller, which indicates a stronger dependence on laser ellipticity. It is interesting to see that for each ion, the FWHM increases as the laser intensity is increased, indicating less dependence on the laser ellipticity at higher laser intensity. This observation can be attributed to the fact that as the laser intensity is increased, sequential ionization would contribute to the formation of these fragment ions, which differs from the non-sequential process that is induced by the laser-driven electron re-collision with the parent ions and has strong ellipticity dependence. Therefore, our study could shed some light on the physical mechanism of strong field dissociation/ionization of molecules by investigating the dependence of the ion yields on the laser's parameters.

In addition, we measured the angular distributions of different ions of N_2O molecules under linearly polarized light. The results are shown in Fig. 5. The angle here refers to the angle between the laser polarization direction and the TOF flight axis. In our experiments, an anisotropic distribution of an ion, i.e., angle-dependent ionic yields, is due to the non-ignorable kinetic energy release of the ion from the strong-field ionization/dissociation that reduces the detection efficiency at particular angles because of the small slit at the end of the flight tube. As shown in the figure, the angular distribution of parent ions N_2O^+ is almost isotropic. It is a little anisotropic for diatomic molecular fragments N_2^+ and NO^+, which is due to the small kinetic energy obtained from the dissociative ionization. The O^{n+} ion angular distribution is also anisotropic, and as the charge state of O^{n+} increases, the angular distribution becomes narrower. Both NO^+ and O^{n+} ions are peaked at angles of 0° and ±180°, that is, the laser polarization is parallel to the TOF axis. Such anisotropy is due to the geometric alignment of the strong field ionization in the femtosecond laser field, i.e., variation of the ionization rate with the polarization angle in a strong laser field.[35] The N_2O molecules are more easily ionized when the molecular bond is along the laser polarization direction, resulting in a maximum distribution at the parallel polarization. Highly charged fragment ions are mainly produced via Coulomb explosion of unstable multiple charged parent ions, which will carry large kinetic energy, thus a narrower or a more anisotropic distribution is expected; as shown in the figure. It is interesting to see that the angle distributions for N^{m+} fragments have two peaks, one major distribution at parallel polarization and the other minor one at perpendicular polarization. This is due to the fact that N^{m+} fragments can be from two different nitrogen atoms (central and peripheral) of the asymmetry N_2O molecule. For the peripheral nitrogen atom, the angular distribution of N^{m+} should be similar to that of O^{n+} with a maximum at parallel polarization. However, the central nitrogen atom prefers a perpendicular distribution considering the additional bending force on the nuclear structure and momentum conservation during Coulomb explosion. Our observation reproduces the results of Graham *et al.*[36] using a labeled molecule ($^{15}N^{14}N^{16}O$). This perpendicular distribution is also observed in the central atom of other linear triatomic molecules CS_2[31] and CO_2,[37,38] which indicates the change of molecular structure during Coulomb explosion in strong laser fields.

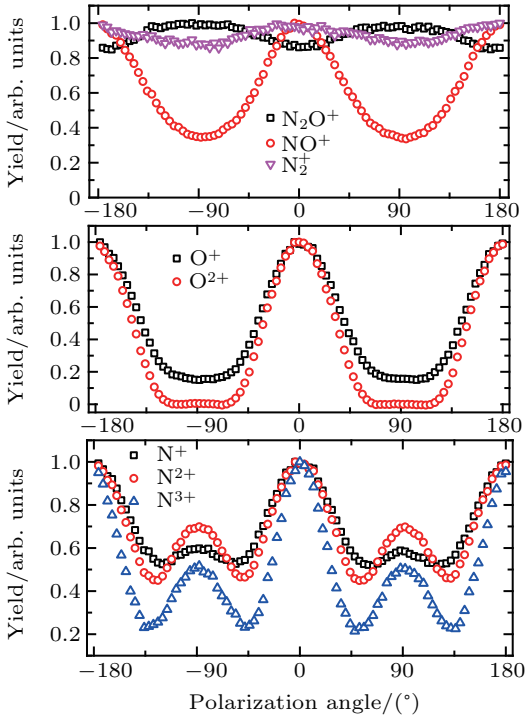


Fig. 5. Distributions of different ions yields as a function of the angle between the laser polarization direction and the flight axis.

As previously mentioned, the electron re-collison plays an important role in the formation of high charged parent ions. Recently, Lin *et al.* demonstrated that a two-

dimensional re-collision driven by the counter-rotating two-color circular laser fields would enhance the double ionization probability.[39] In this case, one could expect a more complex geometric alignment of the molecules, leading to a completely different angular distribution of these fragmental ions compared to that in the linearly polarized 800-nm laser fields that are used in the present study. Further studies would be stimulated to investigate strong field ionization and Coulomb explosion of polyatomic molecules in a two-color femtosecond laser field.

4. Conclusion

In summary, we investigate the strong field ionization and dissociation of N_2O molecules under different laser polarization states. It has been shown that highly charged fragments are produced by dissociation or Coulomb explosion of highly charged parent ions which are formed by NSD(M)I of N_2O. Our results show that the laser induced re-collision process is very important to the formation of highly charged parent ions. In addition, the angular distribution of various fragment ions of the N_2O molecule is obtained, and the N fragment ion angular distribution has two maxima in the horizontal and vertical directions, respectively, which is attributed to the different positions of the two nitrogen atoms in the linear molecule.

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