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Excitation and Ionization of Xenon Atoms in Strong UV Laser Fields

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Abstract: Neutral Rydberg state excitation in strong UV laser fields of Xe atoms has been studied and compared with direct ionization. The yields of both strong-field excitation and ionization have been measured as a function of UV laser intensity and ellipticity. The underlying physical mechanism has been discussed based on the experimental results, indicating that the Rydberg states are populated via multi-photon resonance excitation in strong 400 nm laser fields. Finally, a comparative study of Xe and O_2 with similar ionization potentials have been performed, showing the suppressed excitation of molecules in strong UV laser fields.

Key words: Strong UV laser field, Rydberg state excitation, Multi-photon ionization.

1. Introduction

As a result of the rapid development of ultra-short and ultra-intense laser technology, the peak electric field strength of lasers can reach or even exceed the coulomb field strength seen by an electron in atoms or molecules. A lot of novel physical phenomena are found when strong laser fields interact with atoms and molecules, for instance, high harmonic generation (HHG) [1], high-order above threshold ionization (HATI) [2] and non-sequential double ionization (NSDI) [3]. Recently, it is surprised to find that, both theoretically [4] and experimentally [5], neutral atoms can survive strong laser fields in Rydberg states, known as Rydberg state excitation (RSE). Moreover, recent investigations have demonstrated that the RSE is closely connected to some interesting phenomena such as the absence of near-zero-momentum electrons [6] and the acceleration of neutral Rydberg atoms [7, 8], and is also very useful in the enhancement of HHG [9]. Elaborate experimental and theoretical studies have been performed on this new strong-field phenomenon in the past few years [10-13].

In the tunneling regime, strong field RSE has been measured and described by the frustrated tunneling ionization (FTI) model, which was claimed to be a complementary of the well-known three-step rescattering scenario [5]. This mechanism is partially due to the fact that RSE yield is strongly dependent on laser ellipticity [14]. The long-range Coulomb potential has been proved very important in strong field RSE by theoretical calculations [15]. On the other hand, a recent study indicates that the strong dependence of atomic RSE on laser ellipticity could not be explained in the framework of the tunneling-plus-rescattering scenario (as in HHG or NSDI). Instead, the yield of electrons with low kinetic energy decreases along with the laser ellipticity increasing, that leads to the reduction of the probability of the tunneled electron to be captured into the Rydberg states by the Coulomb potential [16], indicating the possible correlation between the RSE process and the low-energy structure in ATI in the strong laser fields [17].

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Actually, strong-field excitation of atoms has been initially observed in the multi-photon regime using visible or near-IR laser fields [18, 19]. Thereby, the excitation has been almost solely associated with the multi-photon picture [20]. This was supported by a few simulations with time dependent Schrödinger equation (TDSE) [21, 22]. As the laser field increasing, the effective ionization potential increases accordingly when the Stark shift of the ground state and continuum boundary are taken into account. Thus the N-photon ionization channel closed and N-photon resonance excitation can occur at certain laser intensities resulting into some local maximum of strong field RSE in the corresponding laser intensity [21]. Only until recently has this phenomenon been identified in RSE of Ar atoms in 400 nm strong laser fields [23]. The TDSE simulation also confirms the unprecedented enhancement of the RSE in the vicinity of N-photon ionization channel closing. As the laser intensity increasing into the tunneling regime, the resonance effects are strongly modified by the accumulation of the electronic phase in the continuum making the mechanism of strong field RSE translate from multi-photon to tunneling picture. Thus these two possible mechanisms are reconciled [23].

In this study, we investigate the RSE of Xe atoms in strong 400 nm laser fields, and compare that with the strong field ionization. We have observed a substantial fraction of Xe atoms to be survived in high Rydberg states, indicating the significant role of RSE in the interaction of atoms with strong UV laser fields. Possible mechanisms have been discussed based on the experimental results. A comparative study of RSE between Xe and O_2, which have similar ionization potentials, has also been investigated.

2. Experiment setup

The experiment setup used for strong-field ionization and excitation of atoms and molecules was similar to that described in our previous studies [16, 24, 25]. Briefly, a gaseous sample was introduced to the reaction zone through a leak valve with an aperture of 10 μm. The base pressure the interaction chamber is 1×10^{-7} Pa, and the operating pressure is about 3×10^{-6} Pa. A Ti: sapphire system running at a 1 kHz repetition rate, producing 4 mJ/pulse in 50 fs pulses with a center frequency of 800 nm was used in the study. The 400 nm laser pulses were produced by frequency doubling the 800 nm output laser pulses using a BBO

crystal with an efficiency of about 17%. A half-wave plant and a Glan prism were inserting into the laser beam to change the laser intensity continuously. The laser polarization was controlled by rotating a quarter-wave plate before it was focused into the reaction zone by a thin lens with a focal length of 25 cm.

A linear time-of-flight (TOF) mass spectrometer is used to detect the produced cations from strong-field ionization. In the case for detection of direct ionized ions Xe^+, standard direct-current electric fields were applied in the TOF mass spectrometer. In order to detect the neutral molecular Rydbergs, the direct ionized ions (Xe^+) are first pushed away from the detector by an electric field, the remaining high-lying neutral Rydbergs (Xe*) are then ionized by another electric field with a typical delay time of 0.5 μs. These cations are detected by dual micro-channel-plates at the end of flying about 50 cm. The voltages in both cases were kept the same to ensure identical detection efficiencies for Xe^+ and $(Xe^*)^+$. Mass resolved ion signals are recorded using the data acquisition card (National instruments, PXIe-5162) and sent to a PC for analysis. All experimental data are normally averaged over 10^4 laser shots.

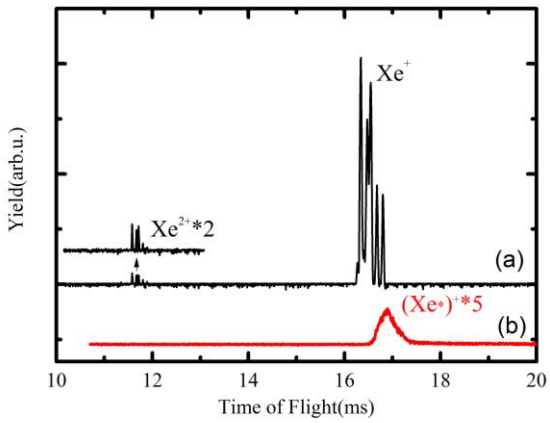

Figure 1: (color online) Typical TOF mass spectra of directly ionization (a) and pulsed field ionization of Rydbergs (b) recorded at linearly polarized 400 nm laser fields with intensity of 1.5×10^{14} W/cm^2.

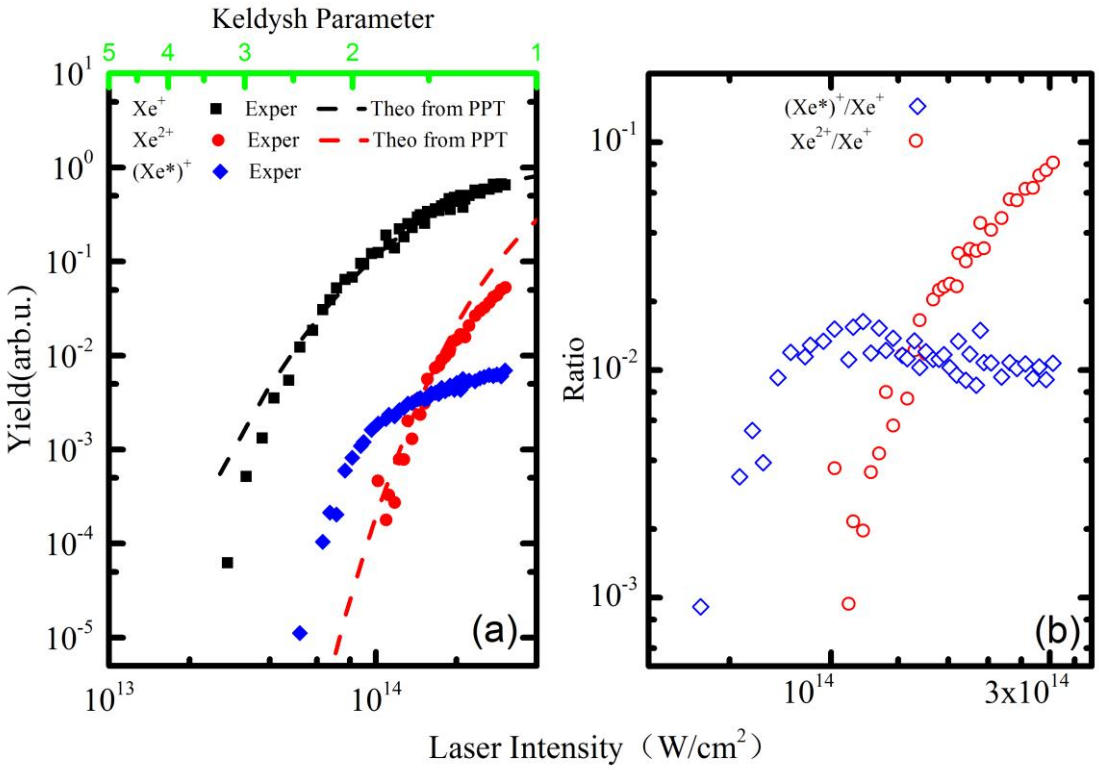

Figure 2: (color online) (a) Dependence of the ion yield of Xe^+ (black square), Xe^{2+} (red circle) and $(Xe^*)^+$ (blue diamond) on the peak intensity of the linearly polarized strong 400 nm laser pulse. The Keldysh γ parameter for Xe is shown on the upper x axis. PPT calculations are shown as black dash line for Xe^+ and red line for Xe^{2+}. (b) Ratio of $(Xe^*)^+/Xe^+$ and Xe^{2+}/Xe^+ as a function of laser intensity.

3. Results and discussion

3.1 TOF mass spectra

In our study, we investigated strong-field RSE of Xe in 400 nm laser fields using pulse-filed ionization combining with TOF mass spectroscopy, allowing us to compare it with strong-field single and double ionization processes under the same experimental conditions. In Fig 1(a), we show the TOF mass spectrum of direct ionization recorded at linearly polarized 400 nm laser fields with intensity of 1.5×10^{14} W/cm^2. Different isotopes of Xe atoms, including ^{129}Xe, ^{131}Xe, ^{132}Xe, ^{134}Xe and ^{136}Xe, are well distinguished in the TOF mass spectra of direct ionization. Both peaks of Xe^+ and Xe^{2+} are clearly identified, indicating strong field single and double ionization induced by the laser fields. Fig 1(b) presents the TOF mass spectrum of pulsed field ionization of neutral Rydbergs Xe* induced by strong UV laser fields. The flight time of the $(Xe^*)^+$ peak is 0.5 μs larger than that of Xe^+, which is exactly the same as the delay time between laser pulse and the pulsed electric field. As the delay time increasing, the corresponding flight time of $(Xe^*)^+$ increases linearly,

confirming it is produced from the field ionization of Rydberg atoms. While before the pulsed-field ionization, the Rydberg atoms move around randomly, which smears out the resolution of isotope as shown in Fig 1(a). As per the adiabatic ionization equation $F=1/9n^4$ [26], where F is the pulsed electric field and n is the principle quantum number of a Rydberg state, we estimated that the Rydberg atoms with n=20~30 are detected in our experiment. The peak intensity of (Xe*)+ is about 1% of that of Xe^+, however, considering that only a small part of Rydberg atoms are detected in this experiment [27], the RSE probability would be much higher. Indeed, in 800 nm strong laser fields, it has been demonstrated that the RSE probability of He atoms is about 10% of direct ionization [5], indicating a substantial fraction of atoms can be survived in neutral Rydbergs after irradiated by strong laser fields.

3.2 Dependence of the direct ionization and RSE on laser intensity

Fig 2(a) shows the dependence of RSE $(Xe^*)^+$, single and double ionization (Xe^+ and Xe^{2+}) on the laser intensity in the range of 3×10^{13} W/cm^2～3×10^{14} W/cm^2. All the three yield curves are normalized as per the maximum of Xe^+ yield. In the strong laser field ionization, the multi-photon ionization will be dominant if the adiabatic Keldysh parameter γ > 1, while for γ< 1, the ionization is in the tunneling ionization regime ($\gamma= (I_p/2U_p)^{1/2}$, where I_p and U_p are ionization potential and ponderomotive energy respectively, $Up=F_0^2/4\omega^2$ where F_0 is laser peak electric field and ω is the laser frequency) [28]. For the intensity range of the 400 nm laser field investigated, the corresponding Keldysh parameter γ is in the range of 4.5～1.2. It is therefore expected that the strong field ionization/excitation occurs in the multi-photon regime. We have performed calculations on direct ionization in linear polarized 400 nm laser fields with different laser intensities using Perelomov-Popov-Terent'ev (PPT) theory [29] and Ammosov-Delone-Krainov (ADK) model [30]. While the ADK results show a large deviation from the experimental results (data not shown), the theoretical results from PPT model well reproduce the experimental measurements as shown in Fig 2(a). It could be understood that the ADK model is based on the tunneling ionization while the PPT model could describe the atomic strong field ionization well in the MPI regime [31], which further infers that the multi-photon ionization is dominant for Xe atoms at 400 nm in the investigated laser intensity range.

The TOF peak of Xe^{2+} appears at laser intensity higher than 10^{14} W/cm^2. The Xe^{2+} yield increases as the laser intensity is increasing, and does not get saturated in the whole intensity range. There is no clear 'Knee' structure upon varying the laser intensity, which is a signature of NSDI induced by rescattering of the tunnelled electrons with the nucleus. This is more obvious in the ratio of Xe^{2+}/Xe^+, which sharply increases as the laser intensity is increase (Fig 2(b)). Moreover, the yield of Xe^{2+} calculated by the PPT model, in which the double ionization is assumed to be a sequential removal of electrons, is in good agreement with the experimental results (see Fig 2(a)). Thus the sequential double ionization (SDI) process, rather than NSDI, is dominant for double ionization of Xe atoms in UV laser fields, owing to the reduced probability of rescattering with the ionic core by increasing the frequency of the laser field.

We now turn to the neutral RSE of Xe produced in the strong UV laser field. It can be seen from Fig 2(a) that as the increasing of laser intensity, the yield of Rydberg atoms $(Xe^*)^+$ increases and becomes saturated above 1×10^{14} W/cm^2. The appearance of $(Xe^*)^+$ is at lower intensity than that of Xe^{2+}, and the yield of $(Xe^*)^+$ is larger than that of Xe^{2+} below 1.5×10^{14} W/cm^2. In addition, unlike that of the Xe^{2+}/Xe^+ ratio, the ratio of $(Xe^*)^+/Xe^+$ shows weak dependence on the laser intensity, as shown in Fig 2(b). Our results strongly indicate that the RSE process plays a significant role in the interaction of Xe with a strong UV laser field.

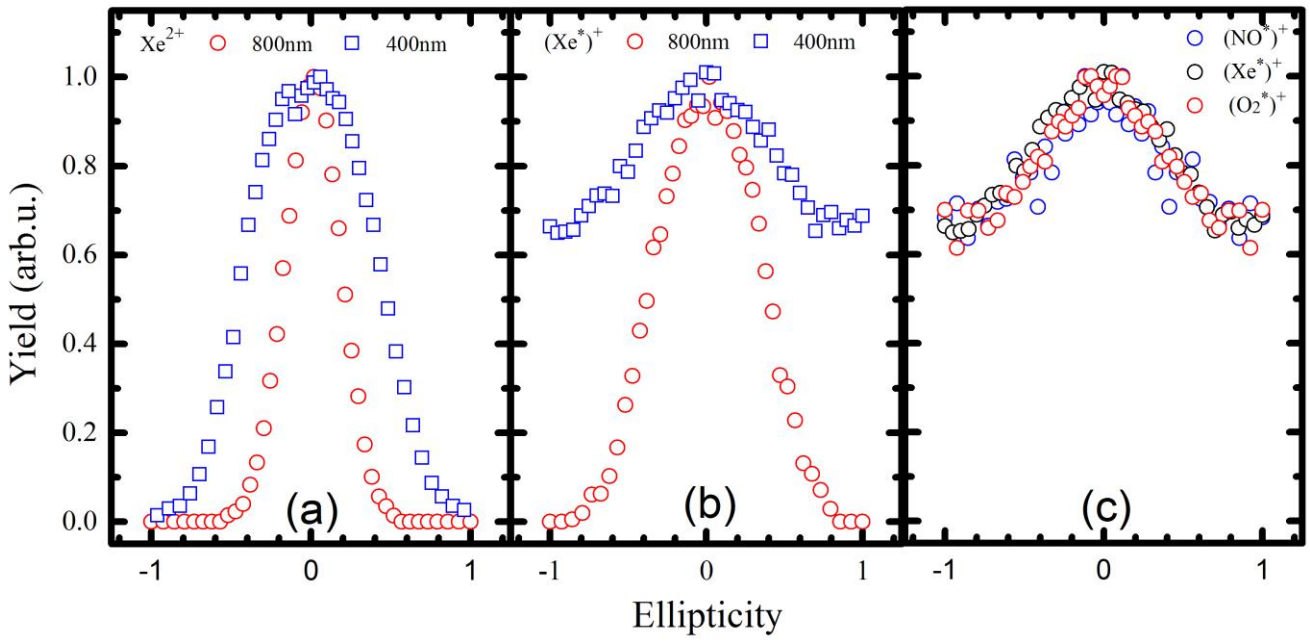

Figure 3: (color online) Ellipticity dependence of the Xe^{2+} (a) and $(Xe^*)^+$ (b) at 400 nm (Blue square) and 800 nm (red circle). (c) Ellipticity dependence of $(Xe^*)^+$ (black circle), $(NO^*)^+$ (blue circle) and $(O_2^*)^+$ (red circle) in the 400 nm laser fields.

For the atomic RSE in 800 nm laser fields, the underlying physical mechanism is still under debate. Both Freeman resonance [32] (via multiphoton absorption [21, 33]) and frustrated tunneling ionization (FTI) [5] (or recapture [4]) mechanisms have been attributed to explain the strong field RSE process in the tunneling ionization regime. A recent study indicates that the RSE is formed by capturing ionized electrons with low kinetic energy (E<0.4eV) into Rydberg states by the ionic Coulomb potential at the end of the laser field [16]. In the MPI regime, however, most ionized electrons could have higher kinetic energy, making it hard to be captured into the Rydberg states by the Coulomb potential of ions. In the case of Xe irradiated by 400 nm laser fields, it only requires absorption of four photons to get ionized. As we have discussed above, strong field direct ionization happens via MPI. The RSE process in 400 nm laser fields thus is induced by multi-photon resonance excitation, also supported by the results of RSE-yield vs laser ellipticity, as we will show in the following section.

3.3 Dependence of the direct ionization and RSE yield on laser ellipticity

In the tunneling ionization regime, it has been proved that the trajectory of the freed electrons can be altered by changing laser ellipticity, and both the probabilities of rescattering with nucleus (for HHG and NSDI) and capturing into the Rydbergs (RSE) diminish in circular polarized laser fields. In order to further investigate the different mechanism of the strong field process in the UV laser field, we have measured the yields of Xe^{2+} and $(Xe^*)^+$ as a function of laser ellipticity of the 400 nm laser fields, and have compared with those obtained in the 800 nm laser fields. The results are presented in Fig 3 for Xe^{2+} (Fig 3(a)) and $(Xe^*)^+$ (Fig 3(b)), respectively. All the data are normalized to the each maximum respectively.

In Fig 3(a), it can be seen the yield of Xe^{2+} produced in the 800 nm laser field drastically decreases with an increase of the laser ellipticity. This behaviour is typical for a rescattering process as it has also been measured for HHG consistent with that the Xe^{2+} is mainly produced by NSDI at 800 nm laser field. While in the 400 nm laser field, a much weaker dependence of Xe^{2+} yield on laser ellipticity is observed, as shown in Fig 3(a). By Gaussian fitting our experimental data, the full width at half maximum of the Xe^{2+} yield versus ellipticity is evaluated to be σ_{400}=0.82 at 400 nm, larger than that σ_{800}=0.43 at 800 nm. This further supports that the SDI are dominant for Xe at 400 nm in this experiment. The decline of Xe^{2+} at 400 nm may result from the reduction of transition probability in elliptical and circular polarized laser fields.

The yield of RSE is measured as a function of laser ellipticity at 400 nm and 800 nm respectively, and the results are shown in Fig 3(b). In the 800 nm laser field, the yield of RSE strongly depends on the laser ellipticity. The decline of the RSE yield with increasing ellipticity can be attributed to a decrease of electrons with low kinetic energy that could be captured in the Rydberg states by the Coulomb potential [16]. Completely different ellipticity dependence is observed for RSE in the 400 nm laser fields. The yield of $(Xe^*)^+$ peaks at linear polarized laser field, and only slightly decreases as laser ellipticity is increasing. The $(Xe^*)^+$ yield under circular polarization is as high as 70% of that under linear polarization. We also measured the RSE yields as a function of laser ellipticity at 400 nm for different atomic and molecular targets, Xe (I_p=12.13eV), O_2 (I_p=12.07eV) and NO (I_p=9.26eV)), and the results are shown in Fig 3(c). It is interesting to see that all the ellipticity-dependence curves are almost the same with each other. Unlike the results in the 800 nm laser field [16], the elliptical dependence of RSE in the 400 nm laser field is irrelevant to the ionization potential and atom molecule species.

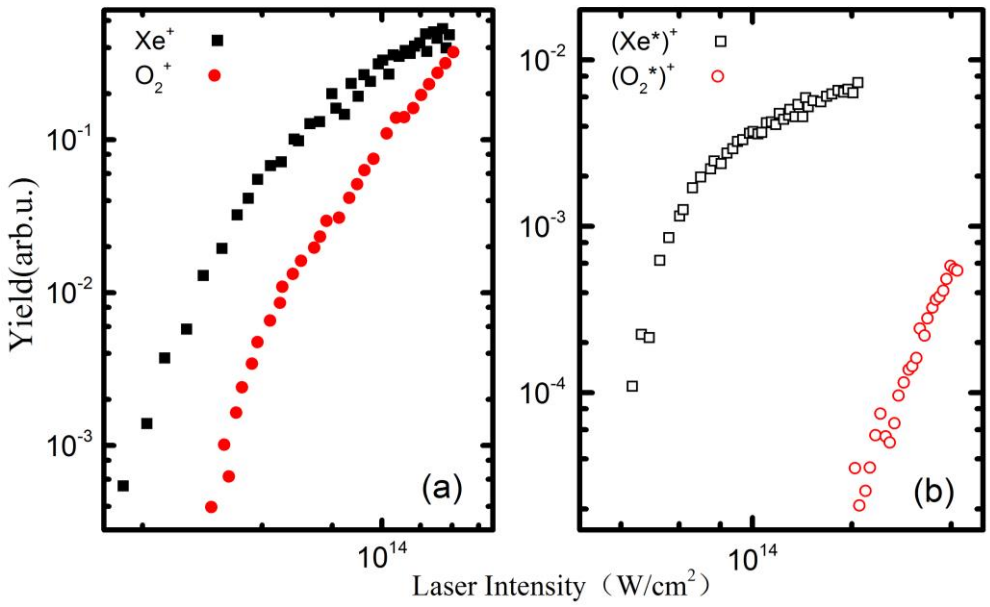

Figure 4: (color online) (a) Dependences of the ionization yield of Xe (black square) and O_2 (red circle) on the laser intensity at the linearly polarized 400 nm laser field. (b) The same with (a) but for RSE of Xe and O_2 respectively.

As mentioned above, the multi-photon resonance between the ground state and a group of excited (Rydberg) states contributes to Rydberg state excitation at 400 nm in this laser intensity range. Because the Rydberg states observed in this study have high- principle quantum numbers, the small energy interval of the adjacent states is less than the laser bandwidth, and the probability of multi-photon resonance strongly depends on the laser intensity instead on laser ellipticity. This is consistent with the observation shown in Fig 3(b) and 3(c), indicating that all these three atoms and molecules are excited by multi-photon resonance. Considering that Xe, NO and O_2 possess similar Rydberg levels owing to the large orbits of the almost decoupled electron, it's no surprise to find that the ellipticity dependence curves are similar to each other for these three species, no matter how many photons are needed in the RSE (4 photons are needed for RSE of O_2 and Xe, 3 photons are needed for NO). Since the laser intensity was kept constant in Fig 3, the electric field intensities decreases as the laser ellipticity increasing, this maybe the reason that the yield of $(Xe^*)^+$ decreases slightly in circular polarized laser fields. Another feature of multi-photon resonance excitation is the oscillating yield along with laser intensity [21, 33], which is absent in our experiment. This could be due to the focal average effect and the fluctuation of laser intensity that smooth the yield curve of $(Xe^*)^+$. However, a closer inspection on the ratio $(Xe^*)^+/Xe^+$ *vs* laser intensity (Fig 2(b)) may see a hump around 10^{14} W/cm^2, which may be the residue of the oscillating structure. This could stimulate more insight experimental and theoretical studies in the future.

3.4 Suppressed ionization and RSE in 400 nm strong laser fields: O_2 VS Xe

With a comparative study between a molecule and its companion atom (an atom with I_p similar to that of the molecule), various studies have revealed the molecular structure effect in strong field single ionization [34], ATI [35] and NSDI [36]. Here, we have investigated and compared strong field ionization and RSE probability in the 400 nm laser field, for O_2 and Xe that have a similar ionization potential. The results are shown in Fig 4(a) for ionization and Fig 4(b) for RSE. It is clear to see that strong-field direct ionization is suppressed for O_2 comparing to its companion atom Xe, while the suppression in RSE is much more significant than that in direct ionization. The $(O_2^*)^+$ yield is too low to be detected until the laser intensity reaches 2×10^{14} W/cm^2, and the excitation yield of $(O_2^*)^+$ is at least two order of magnitudes lower comparing to that of $(Xe^*)^+$.

In strong 800 nm laser fields, suppressed ionization of O_2 comparing to that of Xe has been observed for years, and recently, by comparing with the experimental data at near-infrared wavelength and the S-matrix theory calculations, it has been unambiguously identified the role of two-center interference in suppressed ionization of O_2 [34]. In UV laser fields, the only study was performed by Wu *et al* in 2006 [37], which also showed the suppressed ionization in MPI regime, at laser intensity lower than that used in the present study. The suppressed multi-photon ionization was attributed to the multi-electron screen effect [37]. Very recently, we have carried out the first study on comparing the RSE process on atoms and molecules in 800 nm laser fields [25]. The results indicate that besides the ionization suppression, the molecular orbital structure of O_2 also leads to a dispersion of electron wave packets which finally reduce the capture probability in strong field RSE. Since the mechanism of RSE in 400 nm laser fields could be multi-photon resonance excitation as aforementioned, the excitation suppression of O_2 observed in MPI regime can't be explain by the photoelectron wave packets dispersion as in the TI regime. Moreover, the RSE suppression is much stronger than the ionization process, indicating that besides the multi-electron screen effect, there should be some other effects remaining to be explored. This demands further theoretical investigation in the future to reveal the important role of molecular structure in the strong-field multi-photon resonance excitation.

4. Conclusions

In this study, we have investigated neutral RSE as well as ionization processes of Xe in strong 400 nm laser fields. The yields of both strong-field excitation and ionization have been measured as a function of laser intensity in the range of 3×10^{13} W/cm^2～3×10^{14} W/cm^2 and laser ellipticity. By comparing with PPT calculations, we show that single and double ionization are mainly produced through MPI and SDI respectively. For RSE, a weak ellipticity dependence was observed in 400 nm laser fields comparing to that in 800 nm laser fields in the TI regime, and the RSE of different atoms and molecules exhibit quite similar ellipticity dependence in strong 400 nm laser fields. It is indicated that the Rydbergs are produced through multi-photon resonance excitation. A suppression of O_2 compared to Xe is observed in either ionization or excitation, which will stimulate further insight studies to reveal the essential role of molecular structure in strong-field physical processes induced by UV laser pulses.

Acknowledgements

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