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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 multiphoton 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 ultraintense 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], highorder 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-zeromomentum 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 μ m. 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

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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 μ s. 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 10⁴ 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/cm².



Figure 2: (color online) (a) Dependence of the ion yield of Xe⁺ (black square), Xe²⁺ (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 Xe²⁺. (b) Ratio of (Xe⁺)⁺/Xe⁺ and Xe²⁺/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/cm².

Different isotopes of Xe atoms, including ¹²⁹Xe, ¹³¹Xe, ¹³²Xe, ¹³⁴Xe and ¹³⁶Xe, are well distinguished in the TOF mass spectra of direct ionization. Both peaks of Xe⁺ and Xe²⁺ 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 μ s 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,