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Population trapping in Xe atoms

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Abstract. We study the interference stabilization (population trapping) of Xe atoms using a fs Ti – Sapphire laser both experimentally and theoretically. The investigation is performed for two pulses of different duration. The signature of population trapping arising from the dynamic multiphoton resonance of the initial state and a group of Rydberg states of the atom is found to exist. The results obtained can be considered as the manifestation that population trapping is indeed a universal phenomena.

1. Introduction

The idea of interference stabilization (IS) of Rydberg atomic states in a strong laser field was proposed by Fedorov and co-workers [1] in 1988. In accordance with [1,2], the physical reason for the stabilization is the interference of the amplitudes of transitions to the continuum from the excited Rydberg states that are coherently repopulated by the Λ -type Raman transitions in the strong laser field. The suppression of the ionization in this case is a result of the overlapping of the ionization widths of all these Rydberg states and the destructive interference of the amplitudes of the transitions to the continuum. The threshold of the interference stabilization phenomenon can be estimated as

$$\varepsilon_0 / \omega^{5/3} > 1 \quad (1)$$

where ω is the laser frequency and ε_0 is the amplitude of the laser's electric field strength [2]. For the Ti:Sapphire laser ($\hbar\omega = 1.55$ eV) condition (1) is fulfilled for laser intensities larger than $I > I^* \approx 10^{13}$ W/cm². The existence of the interference stabilization was first experimentally demonstrated in the most unambiguous way in [3-5].

The possibility of IS for the atom in the ground atomic state which is coupled with the continuum by the multiphoton transition was proposed in [6] and studied theoretically in detail in [7]. The key element of such interpretation is the multiphoton resonance of the ground state and a set of high-lying Rydberg states AC Stark shift taking into account, and then the IS of population trapped in excited Rydberg states. Recent data on population trapping in Rydberg states obtained by the numerical solution of the time – dependent Schroedinger equation (TDSE) for Hydrogen and model Silver atoms are discussed in [8-10].

Independently, the same idea of population trapping in Rydberg states was suggested by S.L. Chin et al [11,12] for interpretation of experimental results on the strong – field ionization yields in atoms of noble gases and molecules, respectively. It was found that in a certain intensity range below the saturation intensity, the ion yield would decrease subtly before increasing again. This was explained by the dynamic resonance of the ponderomotively shifted Rydberg states resulting in population trapping. In [13] the population trapping in high – lying Rydberg states was demonstrated to exist also in another molecular system. Paper [14] gives a review of experimental work of S.L. Chin et al. on this subject in which real application to facilitate the remote detection of THz pulses through filamentation was discussed.

The trapping of population in highly excited Rydberg states was also observed in recent experiments with He atoms irradiated by Ti-Sa femtosecond pulse [15]. The authors of this paper proposed the model of the frustrated tunnel ionization. In accordance with this theoretical model the capture to highly excited states take place as a result of the recombination process on parent ion during the turn-off time of the laser pulse caused by the Coulomb interaction with the atomic core. Such a temporal dynamics is in contradiction with the model based on IS phenomenon where the population of excited Rydberg states is found to be pronounced already at the first ramp of the laser pulse as soon as the field amplitude becomes close to its peak value [10].

In this work we study both experimentally and theoretically the ionization dynamics features of Xe atom in the strong pulse of the Ti-Sapphire laser. The step structure of ionization yield versus laser intensity is interpreted as a manifestation of the population trapping in excited states and confirmation of interference stabilization phenomenon. The effect of the pulse duration and spectral bandwidth on population trapping are analyzed in the frames of IS model.

2. Experimental Setup

A schematic diagram of the experimental setup is shown in figure 1. Briefly, a commercial ~0.2 TW femtosecond Ti-sapphire laser system was used. It consists of an oscillator (Spectra Physics Tsunami) followed by a chirped pulse amplification (CPA) module (Spectra Physics Spitfire). In the CPA section, a stretcher is used to stretch the pulses from 30 fs transform limited to around 200 ps positively chirped pulses. These were then amplified by a regenerative amplifier at 1 kHz repetition

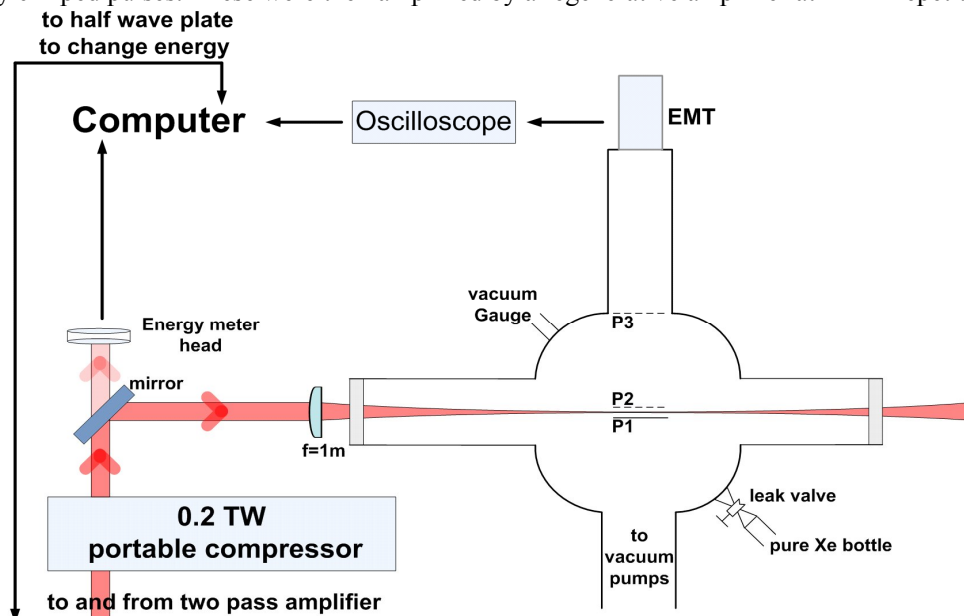


Figure 1. Sketch of the experimental setup.

rate. A pulse picker was used to select a ten hertz beam from the 1 kHz beam. The 10 Hz beam was sent into a two pass Ti-sapphire amplifier. A portable parallel grating compressor is used to give a transform – limited pulse with pulse duration of about 45 fs at FWHM (full width at half-maximum). The pulse duration is measured using a second – order single – shot autocorrelator (SSA, Positive Light). The output beam has a maximum energy of 20 mJ/pulse with a slight elliptic shape beam pattern with diameter of about 4.2 (vertically) and 4.7 (horizontally) mm (Full width at half maximum of the fluence). A half – wave plate and a polarizer are used inside the two pass amplifier to change the output energy of the linearly polarized pulses. They are installed before the gain medium.

To change the pulse duration and to compare two cases with the same pulse duration and different bandwidth, we used a narrow bandwidth band pass filter. Since after the amplification stage the power of the pulse can damage any optical component, we put the band pass filter before the two pass amplifier stage. Based on uncertainty principle and considering Gaussian pulse shape the transform limited pulse duration of 175 and 45 fs has the bandwidth of 5.3 and 20.9 nm respectively with central wavelength of 800 nm. In both cases, the beam was focused into an ultrahigh – vacuum (UHV) chamber by a 1 m plano–convex lens. The background pressure of the UHV chamber was 3×10^{-9} Torr. The UHV chamber was evacuated by a turbo molecular pump connected to a TriScroll pump (oil free rotary pump). The Xe pressure in the interaction chamber was controlled by a precision leak valve (see figure 1) and ranged from 8×10^{-8} to 7×10^{-4} Torr. Ion curves were produced by combining a series of intensity scans, each having a different fill pressure in the interaction chamber. Ion species were collected using a time – of – flight mass spectrometer with a 60 cm long drift tube. The laser was focused through the centre between two plates P1 and P2 (with an opening covered by a metallic mesh) spaced by 5 mm (see figure 1). The voltages applied to P1 and P2 were 4 and 3.5 kV, respectively. The opening of the TOF spectrometer, P3, was 25 mm covered by a metallic mesh.

3. Results and Discussions

3.1. Experimental results

First, let us discuss the experimental data for the ionization yield of Xe atoms (which is proportional to

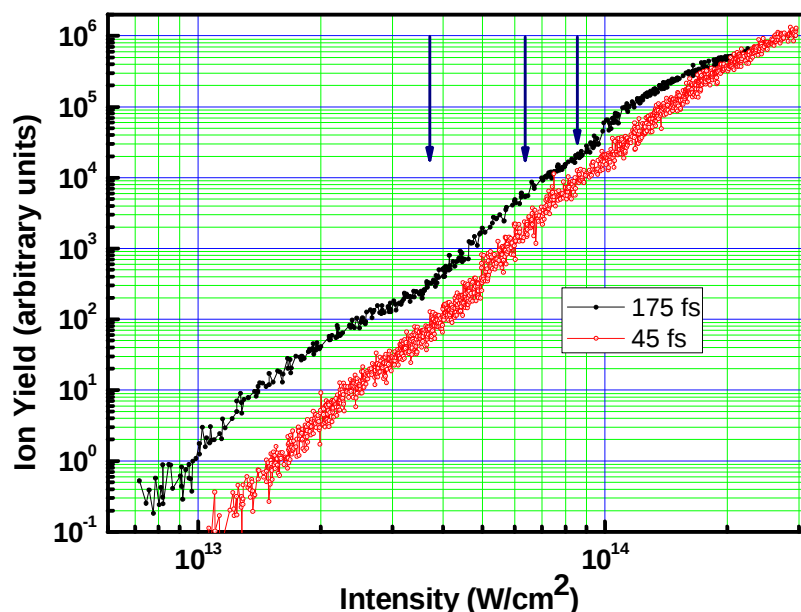


Figure 2. The dependence of the ion yield of Xe atom on the intensity for “long” (black) 175 fs pulse, and “short” (red) 45 fs pulse. Arrows indicate the intensities mentioned in the text.

the average probability W_i as a function of laser intensity I . The results obtained in our experiment for "short" (45 fs) and "long" (175 fs) laser pulses with two different bandwidths are presented in figure 2. Both pulses are transform limited pulses. The main specific feature of the dependence obtained for the 175 fs pulse is its step – like structure. The steps on the dependence $W_i(I)$ are well pronounced for the intensity values around 3.7×10^{13} , 6.2×10^{13} and $8.3 \times 10^{13} \text{ W/cm}^2$. As it was mentioned above, in [11, 12] it was supposed that the reason for such step – like dependence is the resonance character of the population trapping in a number of high-lying Rydberg states that are resistant to the ionization if the laser field strength exceeds the critical value (1). In comparison with the case of the "long" laser pulse such step – like structure is much less pronounced for the curve corresponding to the "shorter" 45 fs laser pulse. It means that the resonant character of the excitation process might have been smeared out due to the broader bandwidth in this case. Also there is a shift of the steps to higher intensities. As a result, the ratio of the ion yields for "long" and "short" pulses $w_i^{(long)} / w_i^{(short)}$ demonstrates a non – monotonous behavior of the dependence on laser intensity (see figure 3). In the intensity range from $3 \times 10^{13} \text{ W/cm}^2$ up to 10^{14} W/cm^2 below the saturation of ionization process, the ratio of ionization probabilities $w_i^{(long)} / w_i^{(short)}$ oscillates near the value of 3.8 corresponding to the ratio of pulse durations for "long" and "short" pulses. For the intensities higher than 10^{14} W/cm^2 the saturation of ionization process for "long" pulse appears to exist and the ratio of the yields $w_i^{(long)} / w_i^{(short)}$ has the tendency to decrease. On the other hand for intensities lower than $3 \times 10^{13} \text{ W/cm}^2$ the ionization probability is dramatically enhanced for the case of the "long" pulse; the ratio $w_i^{(long)} / w_i^{(short)}$ at the intensity $\approx 2 \times 10^{13} \text{ W/cm}^2$ reaches almost one order of magnitude higher than the level at the highest intensity. The physical reason for this dramatic enhancement of ionization will be discussed later.

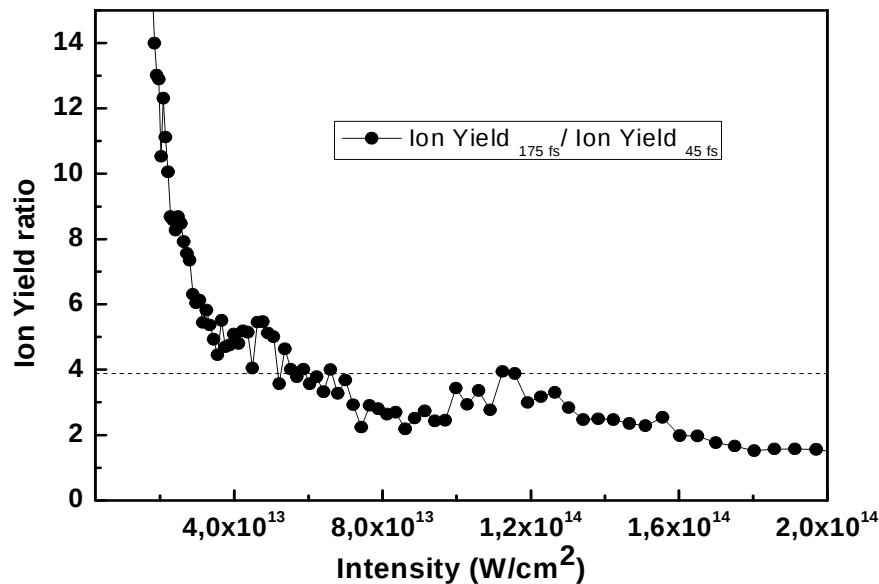


Figure 3. The ratio of ion yields $w_i^{(long)} / w_i^{(short)}$ for "long" (175 fs) and "short" (45 fs) laser pulses. Dash line indicates the ratio of pulse durations 3.8.

3.2. Numerical simulation

To confirm the effect of population trapping in high – lying Rydberg states we performed the numerical simulation of the dynamics of ionization of Xe atom for laser parameters similar to the experimental ones. Our simulation was based on the solution of the TDSE (time dependent Schroedinger equation) for the model single electron Xe – like atom. In our calculations the potential for the model Xe atom was chosen in the form

$$V(r) = -\frac{e^2}{\sqrt{r^2 + \alpha^2}} \quad (2)$$

with smoothing parameter $\alpha = 0.0943$ A. For such a value of α the ionization potential of the ground state is equal to $I_i = 12.13$ eV and coincides with the ionization potential of the real Xe atom. Two lowest s and p excited states have the energies of -3.21 eV and -3.37 eV respectively, which are close enough to the position of the real levels. This model is rather simple, but it provides both the exact value of ionization potential and existence of Rydberg series which is quite important for the qualitative comparison of the experimental and numerical data.

The laser pulse with $\lambda = 800$ nm ($\hbar\omega = 1.55$ eV) had a smoothed envelope with sine – squared ramps of duration t_f equal to integer number of optical cycles:

$$\varepsilon_0(t) = \varepsilon_0 f(t) \cos(\omega t) \quad (3)$$

where $f(t) = \sin^2(\pi t/2t_f)$ is the pulse envelope, so that the total pulse duration is $2t_f$ and pulse duration at FWHM is $\tau = t_f$. For example, for $t_f = 20T$ ($T = 2\pi/\omega$ is the optical cycle) we have $\tau = 53$ fs. All the details of the numerical procedure can be found in [16].

The main result of our numerical simulations is presented in figure 4 and demonstrates the non –

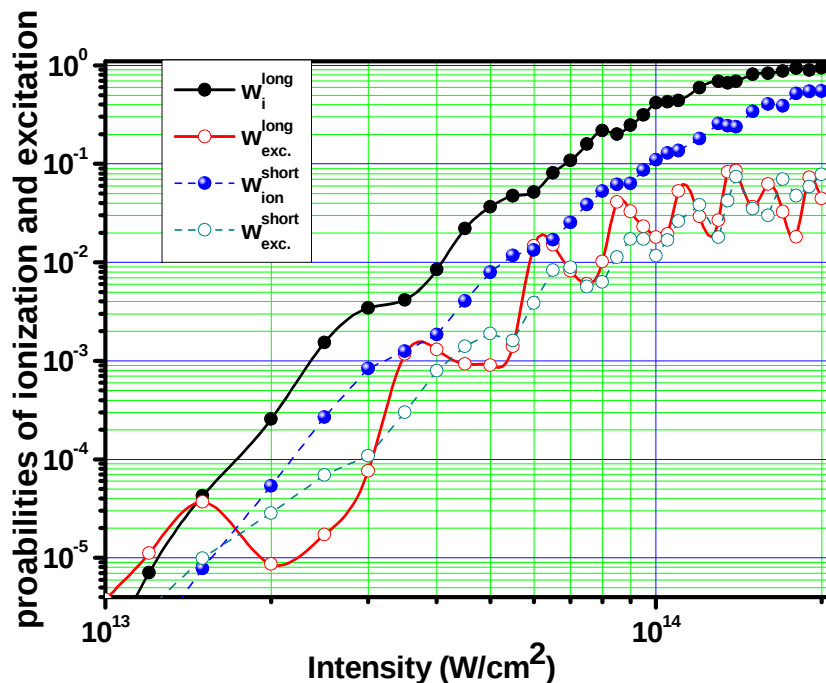


Figure 4. Probabilities of ionization (full circles) and excitation (open circles) per pulse in dependence of intensity of Ti:Sapphire laser pulse with $t_f = 20T$ (solid curves) and $t_f = 5T$ fs (dash curves) for a Xe atom obtained in numerical simulation.

monotonous dependence of the ionization and excitation probabilities of the Xenon atom versus laser intensity for two different pulse durations ($t_f = 20T$ and $t_f = 5T$). These two pulses are shorter in comparison with our experimental pulses of 45 and 175 fs. Nevertheless the comparison is possible and the ratio of durations $R=4$ is similar to the experimental ratio $R=175/45\sim 3.8$. So, qualitative comparison is possible. The obtained dependence for both pulses demonstrates non – monotonous structure with each excitation maximum being accompanied by the minimum of ionization probability. The maxima of excitation probability correspond to the trapping of the population in high – lying Rydberg states with binding energies in a range of 0.1 – 0.4 eV and are similar to that were found in simulations [8,9]. These maxima are separated by approximately the same value of laser intensity equal to $\Delta I \approx 2.5 \times 10^{13} \text{ W/cm}^2$, which exactly corresponds to the ponderomotive shift of the continuum boundary $U_p = e^2 \varepsilon_0^2 / 4m\omega^2$ equal to photon energy $\hbar\omega$. The maxima in excitation probability are better pronounced for longer pulse with $t_f = 20T$ and observed at laser intensities of $3,7 \times 10^{13}$, $6,2 \times 10^{13}$ and $8,6 \times 10^{13} \text{ W/cm}^2$. These values correspond to the “steps” on the ionization curve and we would like to stress that these values of intensity are in rather good coincidence with the positions of the “steps” on the ionization curve obtained experimentally (see figure 2). For shorter laser pulse with $t_f = 5T$ the dependence of excitation probability versus intensity is smoother, but it is also characterized by a set of minima and maxima. Their positions are shifted a little in comparison with the case of long ($t_f = 20T$) pulse. As a result the steps on the ionization curve for longer pulse duration are better pronounced and also shifted a little in comparison with their positions for shorter pulse. This shift in position of maxima in the dependence of population trapped in Rydberg states versus intensity manifests itself in the non – monotonous character of the ratio of ionization yields $w_i^{(long)} / w_i^{(short)}$ calculated for pulses with $t_f = 20T$ and $t_f = 5T$. This dependence is presented at figure 5 and demonstrates the features similar to that obtained in experiments discussed above.

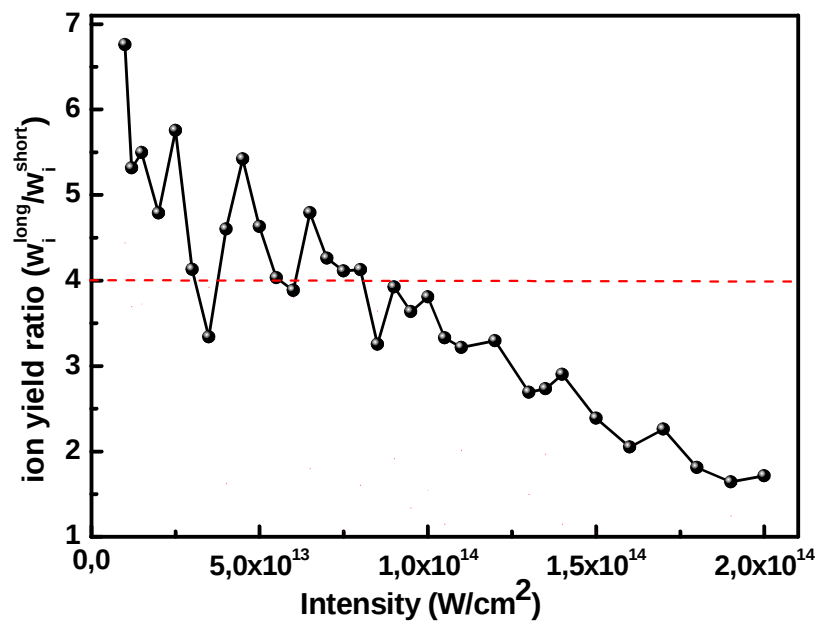


Figure 5. The ratio of ion yields $w_i^{(long)} / w_i^{(short)}$ calculated for “long” (20T) and “short” (5T) laser pulses. Dash line indicates the ratio of the pulse durations.

Indeed, the curve oscillates near the value of 4 corresponding to the ratio of pulse durations for “long” and “short” pulses, has the tendency to decrease in the range of high intensities ($I \geq 9 \times 10^{13} \text{ W/cm}^2$). Also the significant increment of the ratio is seen for low intensity limit ($I \geq 1 - 3 \times 10^{13} \text{ W/cm}^2$). Moreover, the position of minima on the dependence $w_i^{(long)} / w_i^{(short)}$ on laser intensity are in excellent coincidence with the maxima of excitation probability for long pulse that observed on the curve (see figure 4). Hence, the observed oscillation structure of the ratio of ionization yields can be treated as the manifestation of population trapping in high – lying Rydberg states in our experiments.

To provide more insight into the process of ionization and population trapping in high – lying Rydberg states, let us analyze the energy spectra of photoelectrons obtained for a 20T pulse for intensities corresponding to the maximum and minimum of the excitation on the curve in Fig. 4. These spectra represent a number of ATI peaks separated by $\hbar\omega$ from each other. Due to the ponderomotive shift of the continuum boundary the energy of each ATI peak decreases with increment of laser intensity. The “channel closing” effect [17] and the change of the multiphoton order of the ionization process should take place as soon as the peak laser intensity is increased by $2.5 \times 10^{13} \text{ W/cm}^2$. Such a behavior of ATI peaks is clearly seen at figure 6, where the dependence of positions of several first ATI peaks on the peak laser intensity is presented.

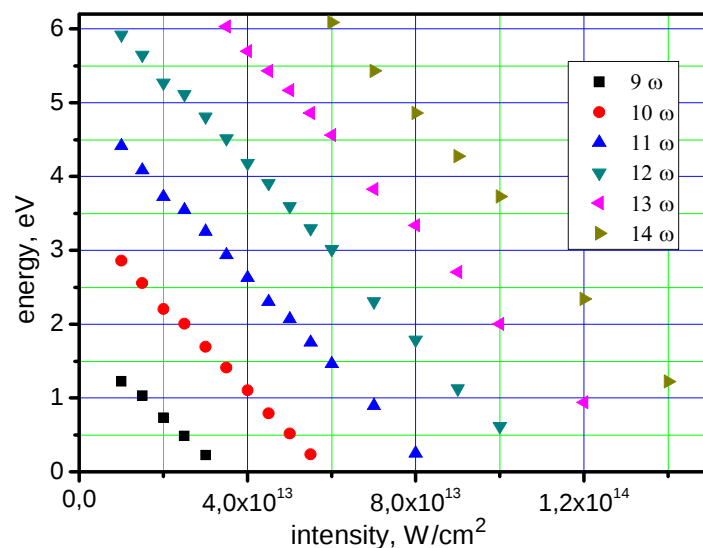


Figure 6. Energies of several first ATI peaks of the photoelectron spectra versus the laser intensity, n is the number of absorbed photons.

As can be seen from figure 6 the intensities 3.7×10^{13} , 6.2×10^{13} and $8.6 \times 10^{13} \text{ W/cm}^2$ providing approximately the maximum of excitation probability correspond to the case just after closing of the 9-th, 10-th and 11-th channels of multiphoton ionization when one more photon is needed to be absorbed for the ionization. In this situation the energy of the first ATI peak is about 1.5 eV. The absorption of 9, 10 or 11 photons for the above mentioned intensities lead to the transition to the high – lying Rydberg states near the continuum boundary that will be efficiently populated. The reason of the observed population trapping consists in the repopulation of the resonantly coupled Rydberg band by Raman – type transitions via the continuum that are very efficient in a strong field and results in the significant suppression of the ionization process (see [8-10] for details).

In contrast, for intermediate values of laser intensity the multiphoton transition to the continuum provides the first ATI peak at the energy $\approx 0.75 \text{ eV}$ and multiphoton excitation gives the possibility to

populate only relatively low – lying states with binding energy around 1 eV and lower principal quantum numbers. Since for this situation the density of Rydberg states is significantly lower and their resistance to ionization is smaller in comparison with the high – lying states near the boundary of the continuum, the excitation probability is found to be suppressed in this case.

In order to compare results of numerical simulations with real experimental observations the integration over the focal volume should be taken into account. In assumption of cylindrical form of the focal volume and the Gaussian radial profile with halfwidth r_0

$$I(r) = I_0 \exp(-r^2/r_0^2) \quad (4)$$

we obtain

$$W_{i(excit)} = \frac{2}{r_0^2} \int_0^\infty w_{i(excit)}(I(r)) r dr \quad (6)$$

The averaged over the focal spot numerical data for ionization and excitation probabilities from figure 4 are presented at figure 7. As can be seen, the existing maxima and minima of ionization/excitation probabilities are partly smoothed and the obtained curves for ionization yield are quite similar to the experimental data (see figure 2).

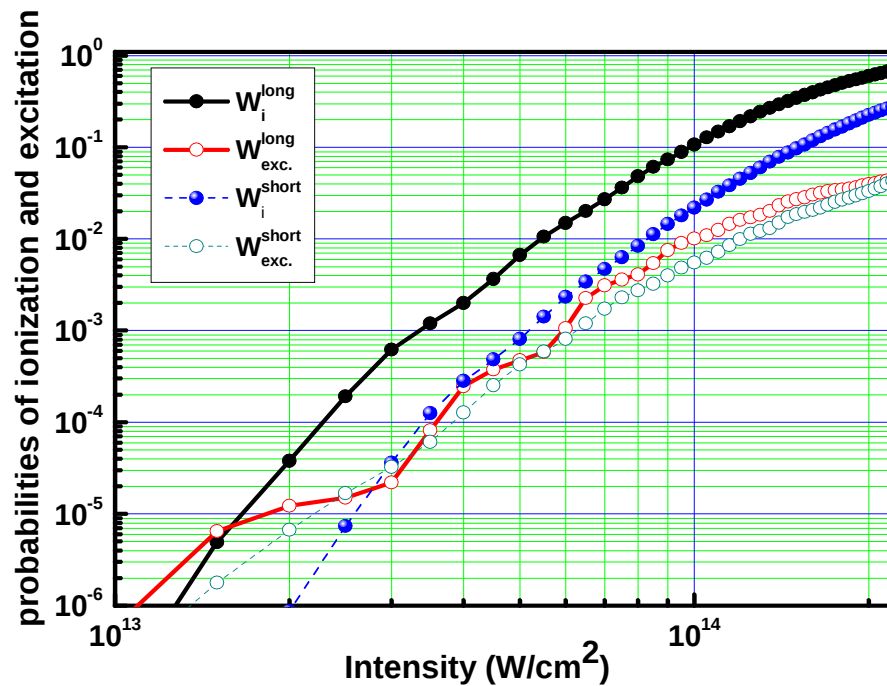


Figure 7. Probabilities of ionization (full circles) and excitation (open circles) averaged over focal spot distribution of intensity in dependence on intensity of Ti:Sa laser pulse with $t_f = 20T$ (solid curves) and $t_f = 5T$ fs (dash curves) for a Xe atom obtained in numerical simulation.

To conclude the discussion of both experimental and numerical results we would like to stress that the population trapped is really an important phenomenon in physics of strong laser field interaction with atoms. To support this statement we present the ratio of trapped population to the ionization yield

versus laser intensity for both “short” and “long” pulses averaged over the Gaussian distribution of the intensity in the focal spot (see figure 8). Both curves have the nonmonotonous structure which can be

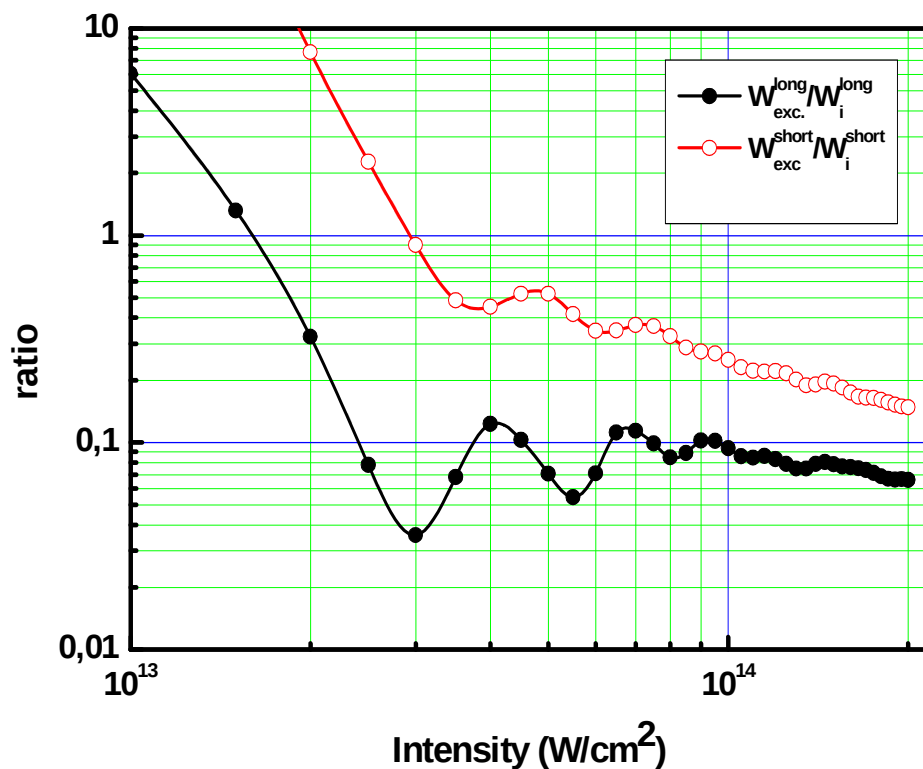


Figure 8. The ratio of trapped populations to ionization yields for “long” (full circles) and “short” (open circles) laser pulses averaged over focal spot versus the intensity of Ti:Sapphire laser pulse for a Xe atom obtained in numerical simulation.

considered as the manifestation of the multiphoton resonances between the ground and highly excited Rydberg states near the boundary of the continuum. The ratio w_{exc} / w_i is larger for “short” pulse. That is due to the wider bandwidth of the short pulse and more effective interaction of the ground state with highly excited states via the multiphoton transition. On the other hand, the resonance features of the curve are less pronounced in comparison with the case of the “long” pulse. It should be emphasized that it is in weak fields ($I \approx 1 - 2 \times 10^{13}$ W/cm²) the probability of excitation is even larger than the probability of ionization, and the ratio w_{exc} / w_i exceeds the value of unity. Unfortunately for such intensities the level of both signals is too low and can be difficultly studied experimentally or numerically. Nevertheless even for “long” pulse with rather high intensity the ratio of the population trapped in the set of Rydberg states to the ion yield oscillates at a level of 10%.

4. Conclusion

So far, population trapping in atoms in a strong laser field was observed through measuring the ion yield as a function of intensity. The qualitative agreement was found to exist between the experimental data and results of numerical simulation based on TDSE solution. The obtained dependences are very similar to the experimental data. The specific features of the ionization yields (step structure) as well

as non – monotonous dependence of the ratio of ionization yields for long and short pulses $w_i^{(long)} / w_i^{(short)}$ vs laser intensity can be considered as the manifestation of the population trapping in high-lying Rydberg states. The observed phenomena were interpreted as the confirmation of interference stabilization phenomenon, which is indeed a universal process during strong laser field interaction with atoms and molecules.

Acknowledgments

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