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Controlling Rydberg excitation process with shaped intense ultrashort laser pulses*

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We perform a theoretical investigation on the control over the atomic excitation of Rydberg states with shaped intense ultrashort laser pulses. By numerically solving the time-dependent Schrödinger equation (TDSE), we systematically study the dependence of the population of the Rydberg states on the π phase step position in the frequency spectra of the laser pulse for different intensities, central wavelengths and pulse durations. Our results show that the Rydberg excitation process can be effectively modulated using shaped intense laser pulses with the laser intensity as high as 1×10^{14} W/cm^2. Our work also have benefit to the future investigation to find out the dominant mechanism behind the excitation of Rydberg states in strong laser fields.

Keywords: shaped laser pulse, Rydberg states excitation, intense field

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1. Introduction

Survival of neutral atoms in (long-lived) Rydberg states in strong laser field has not been paid too much attention in the early time. Only in recent years, both theoretical and experimental works surprisingly found that neutral atoms in Rydberg states can survive very strong laser fields.[1–3] This becomes the subject of elaborate experimental and theoretical studies accompanying new important applications of neutral atoms with high excited states exposed to an oscillating electric field.[4–7] Eichmann *et al.* identified the ponderomotive force on electrons as the driving mechanism, leading to ultrastrong acceleration of neutral atoms with a very high magnitude.[8] Two theoretical mechanisms have been proposed in order to explain how these excited states are populated and how they remain stable in the intense laser field. In the tunneling regime, the frustrated tunneling ionization (FTI) model successfully describes strong field excitation, which says that the electron does not gain enough drift energy from the laser pulse after tunneling and will eventually be captured by the coulomb field of the ion,[9–13] In the multi-photon regime, the high population of Rydberg states is thought to be resulted from the multiphoton resonance of the ground state and a set of high-lying Rydberg states with AC stack shift, and the interference stabilization (IS) of population trapped in Rydberg states.[14–16] However, there is still no precise and unambiguous theory to explain all the existing phenomena. And whether there exists a effective way to coherently control the strong field Rydberg excitation process is also an interesting question.

On the other hand, the ability to modulate the temporal shape of an ultrashort pulse enables the manipulation of the evolution of a quantum system and thus the control of various atomic and molecular processes. In particular, control of the spectral phase of the pulse and thus its temporal profile has been demonstrated effectively for control over numerous physical systems and processes.[17–20] Corresponding examples of past control studies include the control over multiphoton absorption processes in atoms, over nonresonant two-photon and three-photon absorptions in large molecules, and over molecular Raman transitions.[21–27] And in recent years, there has been increasing interest in the control over the ionization dynamics of atoms and molecules using shaped intense ultrashort laser pulses with the peak intensity higher than 1×10^{13} W/cm^2.[28–31] However, the excitation dynamics of atoms and molecules in shaped intense ultrashort laser pulse is still lack of investigation. Generally, both the ionization and excitation dynamics in strong laser field will be different from

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the cases in weak field. This is because the atomic potential will be significantly modified by the strong laser electric field, which can not be treated as perturbation any more. In contrast to the multi-photon absorption picture in weak field, electron's ionization dynamics is governed predominantly by tunneling process in strong laser field. While for excitation process in strong laser field, the mechanism as well as the result of the control will be also not the same as that in weak field.

In this paper, we perform an investigation on the control over the process of Rydberg states excitation in the interaction between the Hydrogen atom with shaped intense ultrashort laser pulse. We test the effect of the coherent control in different laser intensities, center wavelengths, and pulse durations of the shaped intense laser pulses, by numerically solving the time-dependent Schrödinger equation (TDSE). We mainly focus on the potentialities of the scheme for control over the Rydberg excitation using shaped intense laser pulse, and attempt to provide results that would benefit the future work to find out which one of the mechanisms is more suitable to explain the Rydberg excitation process in intense laser fields.

2. Theory

The time-dependent Schröinger equation for atomic hydrogen in intense laser fields can be written as (atomic units $\hbar = e = m = 1$ are used unless indicated)

$$\mathrm{i}\frac{\partial}{\partial t}\Psi(\boldsymbol{r},t) = \left(-\frac{1}{2}\nabla^2 - \frac{1}{r} - \boldsymbol{r}\cdot\boldsymbol{E}(t)\right)\Psi(\boldsymbol{r},t). \quad (1)$$

For a linearly polarized laser pulse, the temporal electric field $\boldsymbol{E}(t)$ along the polarized direction (z axis) is

$$\boldsymbol{E}(t) = \frac{1}{2}E_0 f(t)\,\mathrm{e}^{\mathrm{i}\omega_0 t}\hat{z} + \text{c.c.}, \quad (2)$$

where E_0 is the amplitude of the electric field, ω_0 is the center frequency, and $f(t)$ is the envelope and is assumed to be in the Gaussian shape

$$f(t) = \mathrm{e}^{-t^2/s^2}. \quad (3)$$

Here the shaped pulse is realized by a simple and intuitive pulse shape parametrization, *i.e.*, a π phase jump in the spectral phase. Experimentally, the pulse shaper is composed of a pair of diffraction gratings, a pair of achromatic lens, and a spatial filter. Briefly, after passing through the first lens and grating, the complex spectrum of the input pulse is spatially mapped at the Fourier plane. A one-dimensional programmable liquid-crystal spatial light-modulator array, composed of many computer-controlled discrete phase elements was placed at the Fourier plane of the shaper, and was used as a dynamic filter which can modify the spectral phase of the pulses. Finally, the second lens and grating reassemble the spectral components to form a modified time-shaped pulse.

Theoretically, we can obtain the shaped pulse in a similar way. We first obtain the unshaped pulse envelope function $f(\omega)$ in the frequency domain through Fourier transformation,

$$f(\omega) = \int_{-\infty}^{+\infty} f(t)\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t. \quad (4)$$

Next, by adding a phase π to the spectrum before a given frequency ω and applying inverse Fourier transform, we gain a new temporal envelope function:

$$\begin{aligned} f_{\text{new}}(t) &= \frac{1}{2\pi}\left[\int_{-\infty}^{\omega_1} f(\omega)\,\mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{e}^{\mathrm{i}\pi}\,\mathrm{d}\omega + \int_{\omega_1}^{+\infty} f(\omega)\,\mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{d}\omega\right] \\ &= |f_{\text{new}}(t)|\,\mathrm{e}^{\mathrm{i}\theta(t)}, \end{aligned} \quad (5)$$

where $\theta(t)$ is the temporal phase introduced by the π phase step. Applying Eq. (5) into Eq. (2), we can obtain a shaped laser electric field as shown in Fig. 1. As the π step position increases from 735 nm to 800 nm, the shape of the pulse gradually transforms from a Gaussian shape (Fig. 1(a)) to a flattop shape (Fig. 1(b)), and then becomes a double-peak pulse with the central valley getting deeper (Fig. 1(c) and Fig. 1(d)).

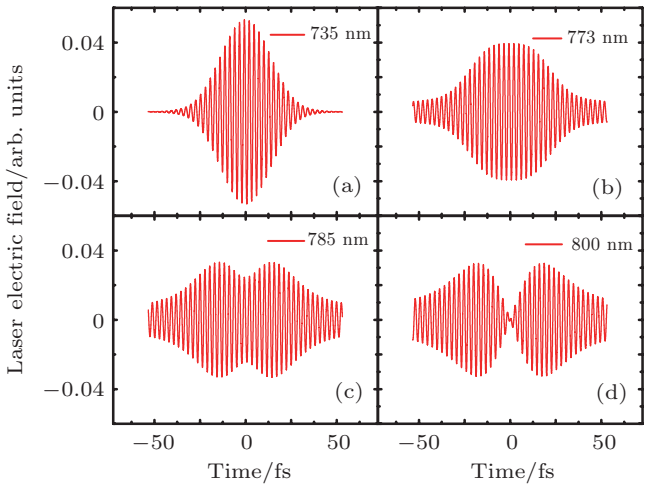


Fig. 1. Temporal evolution of laser electric field for different π step positions, all panels are for an intensity of 1×10^{14} W/cm^2 and central wavelength of 800 nm. The position of the π step are 735 nm, 773 nm , 785 nm, and 800 nm for panels (a), (b), (c), and (d), respectively.

In the numerical calculations, the atom is initially in the 1s state, and $\Psi(\boldsymbol{r},t)$ is expanded with spherical harmonic function and each angular momentum channel expanded on a radial grid

$$\Psi(\boldsymbol{r},t) = \sum_{l=0}^{L}\Phi_l(r,t)\,\mathrm{Y}_l^0(\theta,\phi). \quad (6)$$

The resulting coupled equations are solved using a Peaceman–Rachford propagator.[32] An absorbing boundary is employed to prevent the unphysical reflection from the boundary. This code has been used successfully to study the HHG

process,[33] the low energy structure in ATI,[34] and tunneling dynamics.[35] The numerical convergence is checked by varying the simulation parameters.

3. Results and discussion

In the following we will show the simulated results for different laser parameters. Figure 2 presents the dependence of the population at different levels on the π step position at the end of the laser pulse. Here we only show the results of the π step position lower than the central wavelength since the result is symmetric about the central wavelength in our calculation. The energy levels are denoted by the principal quantum number n. In Fig. 2, we apply 800-nm laser pulses with pulse duration of 24 fs but for three intensities: $0.5I_0$ (Figs. 2(a)–2(d), I_0 (Figs. 2(e)–2(h)), and $2I_0$ (Figs. 2(i)–2(l)), with $I_0 = 1 \times 10^{14}$ W/cm^2. In general, the population of different levels vary with the π step position in different ways. In the case of the lowest intensity, the low lying excited state of $n = 3$ shows a prominent peak around 795 nm near the central wavelength. While for excited states of $n \geq 4$, the most prominent peak appears around 760 nm far from the central wavelength. Especially for the high lying excited states of $n \geq 5$, the excitations are dramatically suppressed at long wavelength, and such a behavior is ideal for control over the excitation process.

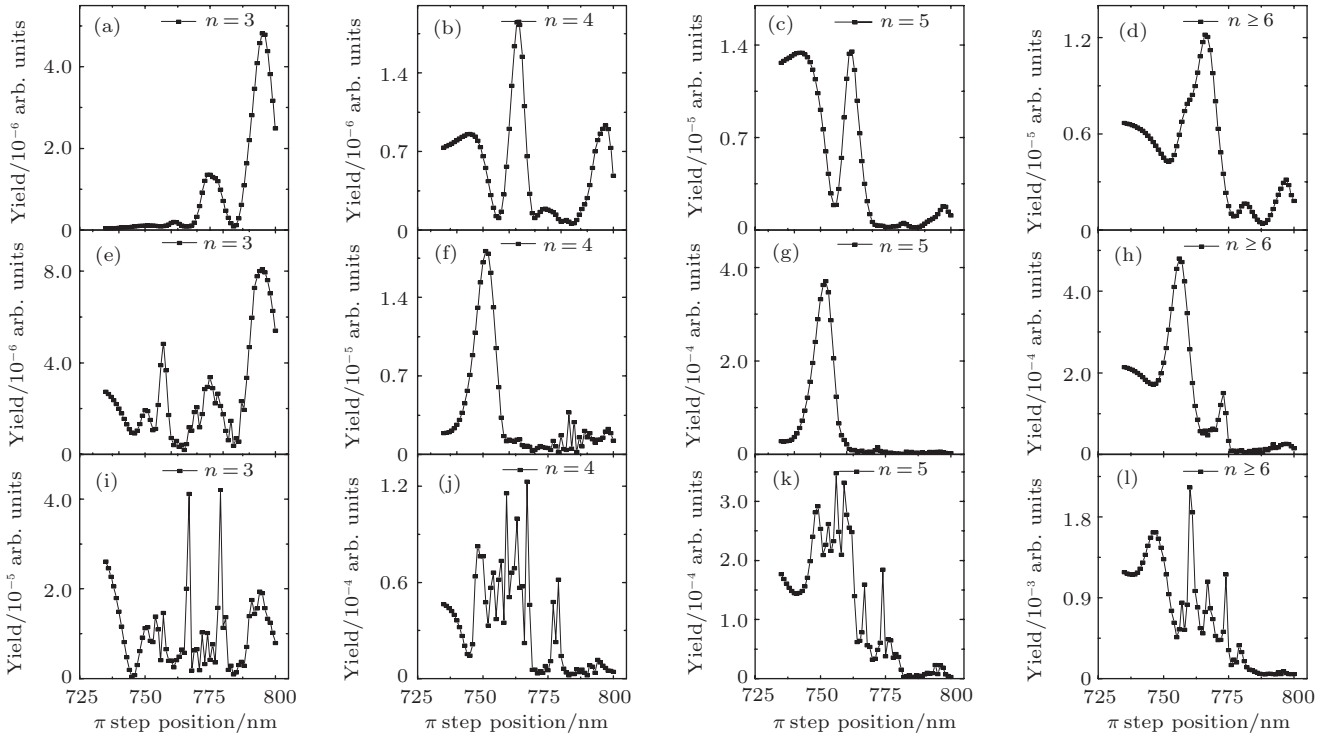


Fig. 2. Simulated population of the excited stats of $n = 3, 4, 5$ and $n \geq 6$ as a function of the phase π step position. The central wavelength is 800 nm and the pulse duration for the unshaped pulse is 24 fs. Panels in the first, second, and third lines correspond to intensity of $0.5I_0$, I_0, and $2I_0$, respectively, with $I_0 = 1 \times 10^{14}$ W/cm^2.

When increasing the laser intensity, the population becomes more and more sensitive on the π step position. At the intensity of I_0 (Figs. 2(e)–2(h)), more narrow peaks arise for the population of the low excited states of $n = 3$. For the high excited states of $n \geq 4$, the principle peak shifts to short wavelength position, and excitation at large π step position is dramatically suppressed. It seems that the control over the Rydberg excitation at the intensity of $I_0 = 1 \times 10^{14}$ W/cm^2 is still effective. However, when the intensity increases to $2I_0$, the situation becomes more complex. There are many narrow peaks on the population for all excited states as seen in Figs. 2(i)–2(l). This means that more channels contribute to the excitation, which makes precise control over excitation more difficult. Therefore, the method of control over the Rydberg excitation using shaped intense laser pulse works well if the intensity is not very high.

In Fig. 3 we present the results of 400 nm to show the effect of the coherent control in shorter wavelength. The pulse duration is 24 fs and the intensity is $I = 1 \times 10^{14}$ W/cm^2. Compared with the case of 800 nm, the curves of 400 nm are more clean and smooth. The low-lying excited states of $n \leq 4$ show a series of peaks which is not suitable for control. For excited states of $n = 5$, the population first decreases quickly and then exhibits minor oscillation in the range of large π step position. While for high-lying excited states of $n \geq 6$, the population shows a monotonous decrease with the π step position and tends to vanish in the range of 375 nm–400 nm, which indicate that the control over the Rydberg excitation at 400 nm is

also effective.

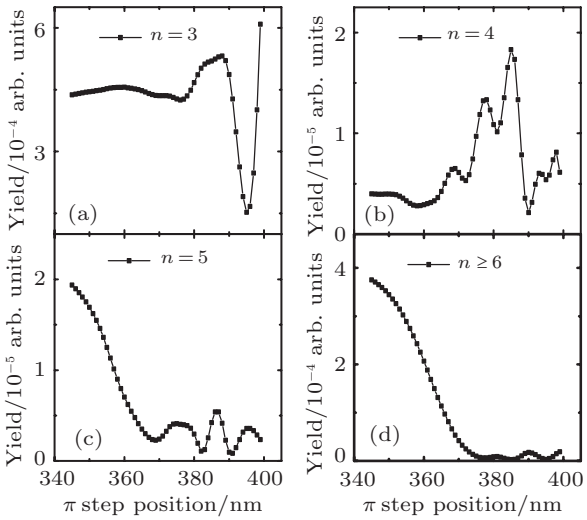


Fig. 3. Simulation of the $n = 3,4,5$ and $n \geq 6$ populations in dependence of the π step position at a central wavelength of 400 nm, intensity of 1×10^{14} W/cm^2, pulse duration of 24 fs.

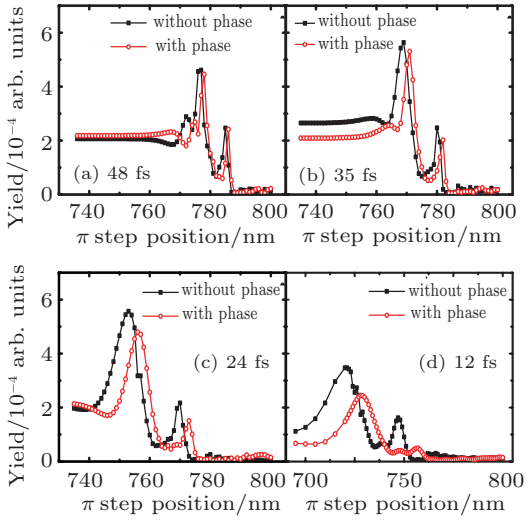


Fig. 4. Simulated population of the Rydberg states of $n \geq 6$ as a function of the π step position for pulse durations of 48 fs, 35 fs, 24 fs, and 12 fs, respectively. In each panel there are two curves corresponding to two different situations, taking the temporal phase $\theta(t)$ in Eq. (5) into account or not. The laser intensity is 1×10^{14} W/cm^2 and the central wavelength is 800 nm.

Finally, we calculate the population of different excited states for different pulse durations. In Fig. 4 we only show the high-lying Rydberg states of $n \geq 6$ in 800-nm laser pulses with intensity of $I = 1 \times 10^{14}$ W/cm^2. As seen in Eq. (5), the π flip operations not only change the envelope of the pulse but will also introduce a time-dependent phase $\theta(t)$ in the shaped pulse in time domain, compared with the unshaped pulse. Therefore, in order to clarify the effects of the envelope and the time-dependent phase, we perform the calculations with (open circle) and without (full square) the above phase, respectively, for comparison. It can be seen from Fig. 4 that as the laser pulse duration decreases, the shape of the curve does not change, *i.e.*, one high peak followed by a low peak, but the position of the peaks moves to the smaller π step position. We can also see that the main effect of the temporal phase is to suppress the excitation and at the same time shift the peaks to larger position but will not change the shape of the curves. At long pulse the effect of the temporal phase is minor, but it becomes more and more prominent when pulse duration decreases. At the shortest pulse of 12 fs, the low peak nearly disappears after taking the temporal phase into account. This is mainly due to the carrier envelope phase (CEP) effect becomes significant in short laser pulse.

4. Conclusion

In conclusion, we theoretically verify the availability of coherent control over atomic Rydberg excitation with shaped intense ultrashort laser pulses and investigate its dependence on the laser intensity, central wavelength and pulse duration of the laser pulses. Our results demonstrate that the Rydberg excitation process can be effectively regulated with shaped intense laser pulses only if the laser intensity is not very high. Our work serves as a basis for the future control over the excitation process of Rydberg states and we expect that it can also pave the way towards understanding of the underlying mechanisms of the Rydberg state excitation in intense laser fields.

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