Laser pulse design for coherent control of Rydberg lithium atoms*

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Using the time-dependent multilevel approach (TDML), this paper studies the dynamics of coherent control of Rydberg lithium atoms and demonstrates that Rydberg lithium atoms can be transferred to states of higher principal quantum number by exposing them to specially designed frequency-chirped laser pulses. The population transfer from n=70 to n=75 states of lithium atoms with efficiency of more than 90% is achieved by means of the sequential adiabatic rapid passages. The results agree well with the experimental ones and show that the coherent control of the population transfer from the lower n to the higher n states can be accomplished by the optimization of the chirping parameters and the intensity of laser field.

Keywords: coherent control, population transfer, time-dependent multilevel approach (TDML), super-Gaussian laser pulse

PACC: 3100, 3120, 3280

1. Introduction

In recent years, the design of laser pulse sequences to achieve efficient and robust population transfer between quantum states has been the subject of many theoretical and experimental studies.[1−6] Coherent population transfer via adiabatic rapid passage (ARP) in atoms through two-photon[7] or multiphoton[8] transitions using chirped pulses has been recently demonstrated. ARP via chirped pulses is a robust and coherent method to transfer population from one state to another with essentially 100% efficiency,[9,10] in which the frequency of radiation coupling of the two levels is swept through the resonance at a rate smaller than the square of the Rabi frequency. The high efficiency of each step allows the population to be transferred through many levels by sequential ARPs, which can make population transfer by using a coherent chirped radiation field, unique and quite attractive in atomic physics and chemistry.

Frequency- or amplitude-chirped pulses have also been used to perform quantum control of physical and chemical processes by robustly driving transitions between quantum states of Rydberg atoms.[11,12] Meerson and Friedland[13] suggested that using a microwave pulse, initially at the Kepler frequency and chirped to lower frequency, would transfer atoms to a higher n state, leading to ionization at a lower microwave field. Recently, considerable attention has been paid to the manipulation and control of the atomic quantum states of Rydberg atoms using single laser pulses.[14] The result shows that the population transition probability is sensitive to laser pulse parameters and the chirped rate, so it is important to study how to choose suitable laser pulse parameters to enhance the population transition probability. In this paper, we numerically investigate the population transfer of Rydberg lithium atoms from $n = 70$ to 75 ($l = 0$, 1 for a given n) by exposing them to the specially designed laser pulses which are constant amplitude laser pulses, Gaussian laser pulses and super-Gaussian laser pulses. The motivation of this paper is twofold: the first is to study the effect of the specially designed frequency-chirped laser pulses on the coherent control of population transfer of Rydberg lithium atoms, and the second is to achieve as complete as possible population transfer between the selected states of lithium atoms by optimizing the chirping parameters and the intensity of laser field. In order to obtain more accurate numerical results, we use a number of cou-

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pled states instead of only the selected states involved. We expect to obtain the condition and probability to maximize the population transfer amplitude in the multilevel system by solving stationary Schrödinger equation using B-spline method.

2. Theory and method

We use the time-dependent multilevel approach (TDML) developed by Xian-Zhou Zhang *et al.*[15] The driving laser pulse is designed as (in atomic units):

$$E(t) = A(t)\cos[\omega(t)t], \tag{1}$$

where $\omega(t) = \omega_0 + \beta t$, ω_0 is the centre frequency of chirped pulse, β is the chirped rate, the expression of $A(t)$ reads:[16]

$$A(t) = A_0 \exp\left\{-c\left(\frac{t}{t_r}\right)^{m'}\right\}, \tag{2}$$

where A_0 is the peak amplitude of laser pulse, t_r is the pulse half width, c is the initial parameter, m' is the pulse shape parameter, $m' = 2$ corresponds to normal Gaussian pulse, and m' (even number)> 2 corresponds to super-Gaussian pulse. The Hamiltonian for the outer electron of the lithium atom takes the form:

$$H = H_0 + zE(t), \tag{3}$$

where H_0 is the field-free Hamiltonian. The stationary Schrödinger equation has the form (in atomic units):

$$\left[-\frac{1}{2}\nabla^2 + V(r)\right]\phi(r) = E\phi(r), \tag{4}$$

where $V(r)$ is the core potential of an alkali-metal atom, involving the effects of penetration and polarization, and is assumed to be the form:[17]

$$V(r) = -\frac{1}{r}\left\{(Z-1)\exp(c_1 r) + c_2 r\exp(c_3 r) + 1\right\}, \tag{5}$$

for lithium atom, $c_1 = -3.395$, $c_2 = 3.212$, $c_3 = -3.207$, and the wave function $\phi(r)$ has the form

$$\phi_{nlm}(r) = [U_{nl}(r)/r]\mathrm{Y}_{lm}(\theta,\varphi),$$

where n, l, m are principal quantum number, angular momentum and magnetic quantum number respectively, $\mathrm{Y}_{lm}(\theta,\varphi)$ is a spherical harmonic function, and the radial function $U_{nl}(r)$ is a solution of the reduced Schrödinger equation:

$$\left[-\frac{\mathrm{d}^2}{2\mathrm{d}r^2} + \frac{l(l+1)}{2r^2} + V(r)\right]U_{nl}(r) = E_{nl}U_{nl}(r), \tag{6}$$

using B-spline basis set, $U_{nl}(r)$ can be expanded as

$$U_{nl}(r) = \sum_{i=1}^{n} c_i B_{i,k}(r) \tag{7}$$

with the boundary condition $U_{nl}(r)(r=0) = 0$, where $B_{i,k}(r)$ is the i-th B-spline of order k. B-spline is a kind of piecewise polynomial, which possesses the characteristics of both analytical and numerical calculation. See Refs. [18] and [19] for detailed characteristics of B-spline. B-splines have been widely applied to the calculation of atomic and molecular physics, and are useful to solve different problems in this realm.[18−23] Xi *et al.*[20] were the first to use the B-splines to treat the ground and the lower excited states of atoms in external field. They calculated the energy levels of the ground and lower excited states of hydrogen in magnetic field of arbitrary strength. Rao *et al.*[23] calculated the ground and the lower excited Stark resonances of hydrogen atom in super-strong electric fields using the B-spline method.

Substituting Eq. (7) into Eq. (6), the following matrix equation is obtained

$$\boldsymbol{H}C = E\boldsymbol{S}C, \tag{8}$$

where $\boldsymbol{H}$ is the Hamiltonian matrix and $\boldsymbol{S}$ is the overlap matrix, E and C are eigenvalues and eigenvectors. By solving Eq. (8), we can get E and $U_{nl}(r)$. The wavefunction of the lithium atom in the presence of laser field $E(t)$can be written as:

$$\psi(r,t) = \sum_{i=1}^{n} a_{il}(t)\phi_i \mathrm{e}^{-\mathrm{i}E_{il}t}, \tag{9}$$

where $a_{il}(t)$ is the coefficient of the expansion which represents the amplitude of the transition probability. The $\mathrm{e}^{-\mathrm{i}E_{il}t}$ contains the phase information that determines the moment when the resonance occurs.[15] The wave function $\psi(r,t)$ obeys the time-dependent Schrödinger equation

$$\mathrm{i}\frac{\partial\psi(r,t)}{\partial t} = H\psi(r,t), \tag{10}$$

Substituting Eq. (9) into Eq. (10), and solving the time-dependent Schrödinger equation numerically, we can obtain $a_{il}(t)$. Then the probability of the outer electron inhabiting in the state i can be written as:

$$P_i = \sum_{l=0}^{l=1} |a_{il}|^2. \tag{11}$$

Using the above formula, we can calculate the state-to-state transition probability from which some interesting features can be observed directly and the population trapping can be obtained.

3. Results and discussion

We now discuss specially designed frequency-chirped laser pulses to move population from $n = 70$ to $n = 75$ using chirped 19.5 GHz to 15 GHz pulses. In quantum mechanical terms it is a sequence of overlapping ARPs through $\Delta n = 1$ transitions between the states of principal quantum numbers n and $n+1$, which occur at the Kepler frequency $1/n^3 = \omega$. We assume that only level 70s is initially populated, then choose $\omega_0 = 19.5$ GHz, and analyse three types of specially designed laser pulses: Constant amplitude laser pulses: $m' = c = 0$; Gaussian laser pulses: $m' = 2$ and $c = 0.5$; Super-Gaussian laser pulses: $m' = 4$ and $c = 0.5$.

3.1. Constant amplitude laser pulses: $m' = c = 0$

We choose a linear chirp with $A_0 = 0.05$ V/cm, $\beta = -0.00245$ GHz/ns, i.e., $\omega(t) = 19.5 - 0.00245t$. The obtained population transfer between atom states is illustrated in Fig. 1(a) which shows a clear evidence for population transfer with about 83% of $n = 70$ atoms transferred to $n = 75$. The transition probability between Rydberg lithium atom states is related to the slew rate of the chirped frequency. To achieve more population transfer from the initial state to the final state, the ratio of the instantaneous frequency to the time span must be small enough to allow atoms to completely pass over the avoided crossings at the corresponding resonance frequency. By changing $\beta = -0.0024$ GHz/ns, the obtained population is illustrated in Fig. 1(b) which shows that about 91% of the $n = 70$ atoms are transferred to $n = 75$. In the process of the transition, the initial frequency is located near the $n = 70$ to 71 resonance and the atoms undergo a sequence of ARPs through the Δn=1 resonances and the frequency chirp stops right after reaching the $n = 74$ to 75 resonance. The frequency sweeping range is from 19.5 GHz to 15 GHz. The population transfer stops at the state $n = 75$. The result agrees well with the experimental result of Gallagher T F[24] in which a 19-GHz to 13-GHz chirped microwave field phase locks the electron's motion of an atom in $n = 70$ and reduces its orbital frequency from 19 GHz to 13 GHz, increasing n from 70 to 79, the instantaneous frequency sequentially equals the Kepler frequency of each n. From Fig. 1, we know that the population transition probability from the initial state to the final state is related to the chirped rate of the frequency, and the ratio of the instantaneous frequency affects the population of the final state efficiently. Hence, to coherently control the population transfer, the chirped rate of the instantaneous frequency of a constant amplitude pulse must be optimized. During the pulses, the population of each state reveals the fine structure, slightly oscillation in time, which could be regarded as a small Rabi oscillation induced by the change of the chirped laser frequency.

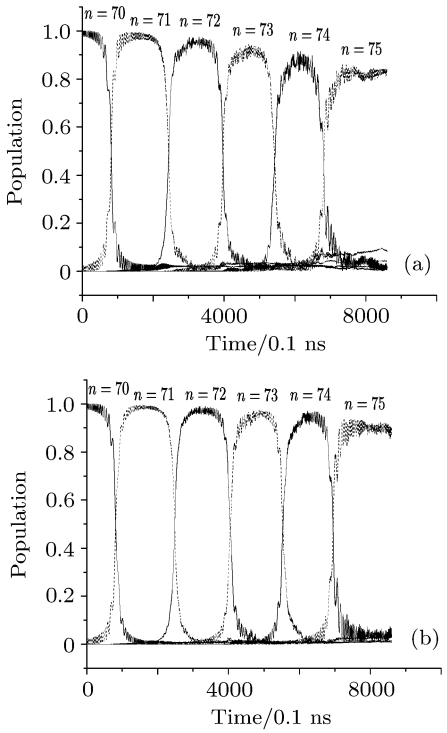


Fig. 1. Population transfer from $n = 70$ to 75 state in constant amplitude laser pulses. The parameters are laser pulse peak amplitude $A_0 = 0.05$ V/cm, centre frequency $\omega_0 = 19.5$ GHz, and the chirped rate for (a) $\beta = -0.00245$ GHz/ns and (b): $\beta = -0.0024$ GHz/ns.

3.2. Gaussian laser pulses: $m' = 2$ and $c = 0.5$

We choose $t_r = 558$ ns, $\beta = -0.0024$ GHz/ns, $A_0 = 0.069$ and 0.072 V/cm, the evolution curves of the field intensity are shown in Figs. 2(a) and 2(b), respectively. The obtained population transfer between atom states is illustrated in Figs. 3(a) and 3(b), respectively. In Fig. 3(a) $A_0 = 0.069$ V/cm, we can find that perfect population transfer can be achieved from $n = 70 \to 71 \to 72 \to 73$, and about 85% of the $n = 70$ atoms are transferred to $n = 74$, but only 77% of the $n = 70$ atoms are transferred to the final state. In Fig. 3(b) $A_0 = 0.072$ V/cm, we obtain almost

complete population transfer, about 94% of the n=70 atoms are transferred to the final state. When the n=70 atoms are exposed to Gaussian laser pulses with a peak amplitude of $A_0 < 0.024$ V/cm, no population is transferred to $n = 75$ state for any chirped rate; when they are exposed to Gaussian laser pulses with peak amplitude $0.024 \leq A_0 \leq 0.05$ V/cm, the populations transferred to $n = 75$ state increase with the increase of laser peak amplitude when keeping other parameters unchanged. In Fig. 4(a), we show the effects of the laser pulse peak amplitude from 0.05 V/cm to 0.095 V/cm on the population transfer from $n = 70$ to $n = 75$ state, which shows that the population transfer from the initial state to the final state can be realized under specially designed Gaussian laser pulses and be largely completely transferred to the final state. When $A_0 = 0.072$ V/cm, the population transition probability transferred to the final state is the biggest (94%), but at higher laser fields the population transfer decreases, as expected. From Figs. 3(a), 3(b) and 4(a), we can find that the population transition probability is sensitive to laser pulse peak amplitude. Hence, to coherently control the population transfer, the laser pulse peak amplitude as well as the chirped rate of frequency must be optimized. From Figs. 3(a) and 3(b) we can see that the population in each state slightly oscillates in time because of the decrease of Gaussian laser pulses in time and the change of the chirped laser frequency.

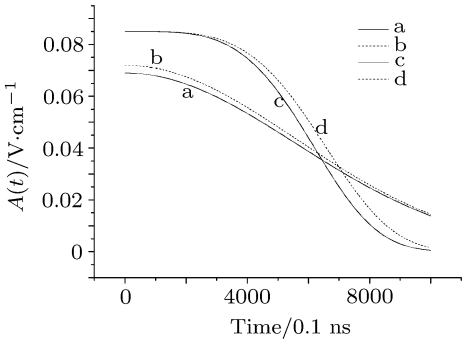


Fig. 2. The evolution curves of the field intensity. The parameters are taken as follows: $A_0 = 0.069$ V/cm, $m' = 2$, $c = 0.5$, $t_r = 558$ ns for curve a; $A_0 = 0.072$ V/cm for curve b; $A_0 = 0.085$ V/cm, $m' = 4$, $t_r = 560$ ns for curve c; $A_0 = 0.085$ V/cm, $m' = 4$, $t_r = 595$ ns for curve d. Other parameters in curves b, c, and d are the same as those in curve a.

3.3. Super-Gaussian laser pulses: $m' = 4$ and $c = 0.5$

We choose $A_0 = 0.085$ V/cm, $\beta = -0.0024$ GHz/ns, $t_r = 560$ ns and 595 ns, the evolution curves of the field intensity are shown in Fig. 2 (curve c and curve d). From Fig. 2 we can see that the super-Gaussian laser pulses decrease more rapidly than Gaussian laser pulses as time goes on. The obtained population transfer between atom states is illustrated in Figs. 3(c) and 3(d). In Fig. 3(c), $t_r = 560$ ns about 76.6% of the population is transferred to the final state; in Fig. 3(d), $t_r = 595$ ns about 88.6% of the population is transferred to the final state. In Fig. 4(b), we show the effects of pulse half width from 550 ns to 596 ns on the population transfer from $n = 70$ to $n = 75$ state, which shows that the population transfer from the initial state to the final state can be realized under specially designed super-Gaussian laser pulses and be largely completely transferred to the final state. From Figs. 3(c), 3(d) and 4(b), we can find that the population transition probability is sensitive to the laser pulse half width. Hence, to coherently control the population transfer, the pulse half width as well as the laser field intensity must be optimized. But in the process, the population of each state reveals the fine structure, a little bit high oscillation in time, which could be regarded as a little bit high Rabi oscillation induced by the rapid decrease of super-Gaussian laser pulses and the change of the chirped laser frequency.

From the above analysis, we can draw the conclusion that efficient population transfer can be realized and be trapped in the final state eventually by modulating the pulse shape parameter m', the chirped rate β, pulse half width t_r and laser pulse peak amplitude A_0 under specially designed frequency-chirped laser pulses. From Fig. 3 we can see that the population transfer can be realized more easily under Gaussian laser pulses and five sequential ARPs driven by chirped 19.5–15 GHz laser pulses, namely, firstly the population is transferred from the lower level to the intermediate level, and subsequently from the intermediate level to the upper level in the multilevel system, finally about 94% of the $n = 70$ atoms are transferred to the final state. From Figs. 3 and 4 we can see that the populations transferred to the final state are different at different pulse parameters, which shows that pulse parameter has an important effect on the population transfer from the initial state to the final state. In our calculation, complete population did not take place. We attribute its shortcomings to our ignoring the interference occurring in successive $\Delta n = 1$ transitions, while we assume that there are levels of $m = 0$, so extending our approach to take all the interferences into account is an interesting and practical work.

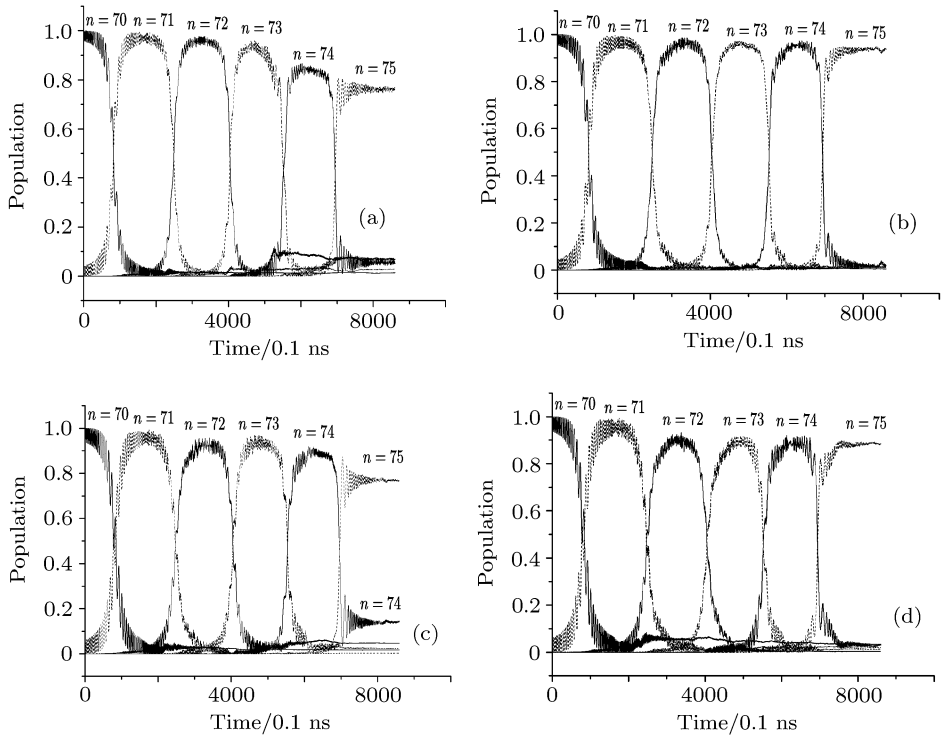


Fig. 3. Population transition probabilities of lithium atoms initially located in the state $n = 70$ and driven sequentially to $n = 75$ state in Gaussian and super-Gaussian laser pulses. The parameters are the same as those in Fig. 2. The chirped rate $\beta = -0.0024$ GHz/ns.

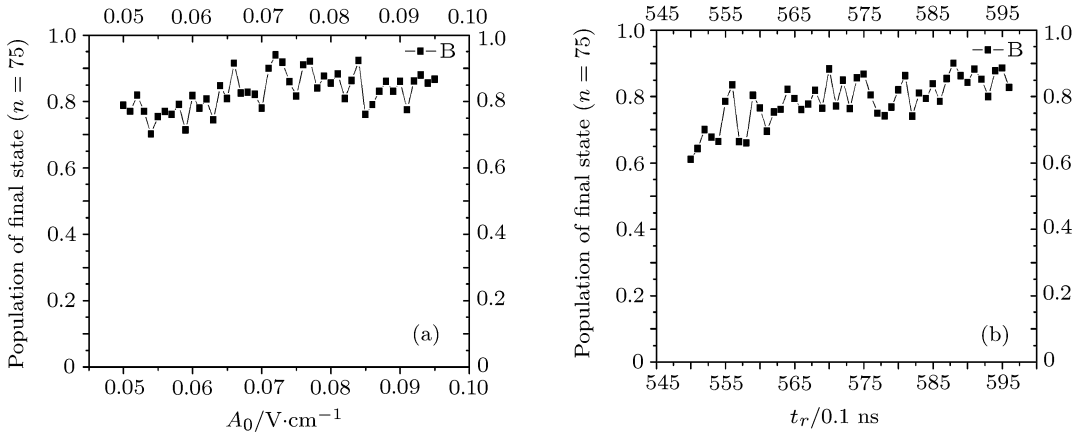


Fig. 4. Population of the final state ($n = 75$) of Li atom as a function of (a) the laser pulse peak amplitude A_0; (b) the pulse half width t_r.

4. Conclusion

In this paper, we have demonstrated that the population can be transferred to a higher state efficiently by exposing Rydberg lithium atoms to specially designed frequency-chirped laser pulses. We found that more than 90% of the population can be transferred from $n = 70$ to $n = 75$ state, and the population transfer can be optimized by modulating the parameters of laser pulses. The results suggest one to determine the suitable laser pulse parameters to achieve complete population transfer between the selected states in a multilevel system. The results also suggest that we can use this method to excite the atom to any state and trap it in this state for a long time by using single specially designed frequency-chirped laser pulses. In a sense, it is possible to achieve coherent control of the population transfer of Rydberg lithium atoms by designing appropriate laser pulses.

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