# Population coherent control of Rydberg potassium atom via adiabatic passage*

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The time-dependent multilevel approach (TDMA) and B-spline expansion technique are used to study the coherent population transfer between the quantum states of a potassium atom by a single frequency-chirped microwave pulse. The Rydberg potassium atom energy levels of $n = 6$–15, $l = 0$–5 states in zero field are calculated and the results are in good agreement with other theoretical values. The time evolutions of the population transfer of the six states from $n = 70$ to $n = 75$ in different microwave fields are obtained. The results show that the coherent control of the population transfer from the lower states to the higher ones can be accomplished by optimizing the microwave pulse parameters.

**Keywords:** adiabatic rapid passage, population transfer, multilevel system

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## 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.[1–5] This subject is related to many applications, including spectroscopy, collision dynamics, and optical control of chemical reactions. The tunable dye laser allows excited high Rydberg $n$ or $l$ states in atoms to reach an efficient population. A number of studies concerning highly excited states of different atoms, such as Na,[6,7] Li,[8] K,[9,10] Sr,[11] Ba,[12,13] He,[14] and Xe[15] have been the hot subjects in the last few decades. Meerson and Friedlang[16] suggested that using a microwave pulse, initially at the Kepler frequency and chirped to lower frequency, would transfer atoms to a higher $n$ state which leads to ionization at a lower microwave field. Bensky *et al.*[17] and Wesdorp *et al.*[18] suggested that it might be more interesting to chirp the frequency in the other direction; they demonstrated that it was possible to induce electron–ion recombination into a high-lying Rydberg state with a half cycle pulse, and the technique could be a way to produce antihydrogen.[19]

Adiabatic rapid passage (ARP) is an approximately 100% efficient way to transfer population from one state to another.[20,21] The high efficiency of each step allows a population to be transferred through many levels by sequential ARPs, which can make population transfer by using a coherent chirped radiation field. Recently the ARP using a single pulse to trap population in a multi-level ladder atom has been extensively discussed. Lambert *et al.*[2] demonstrated that Rydberg atoms could be transferred to states of lower principal quantum number by exposing them to a frequency chirped microwave pulse. In their experiment, Li Rydberg atoms were moved from $n = 75$ to $n = 66$ using chirped 7.8-GHz to 11.8-GHz pulses at the two-photon resonance between states differing in $n$ by 1. Djotyan *et al.*[22] proposed a scheme of population transfer between two ground states of the $\Lambda$ atom without considerable excitation of the atom using a single frequency-chirped laser pulse. This scheme could be realized when the width of the transform-limited laser pulse envelope frequency spectrum (without chirp) was smaller and the peak Rabi frequency of the pulse was larger than that of the frequency interval between the two ground states of the atom. In their analytical studies, two transition dipole elements were assumed to be equal to each other, i.e. $|d_{12}| = |d_{23}|$.[22] But for a real system, this is not always satisfied.

In this paper, we numerically investigate the population transfer in the Rydberg potassium atom with a single frequency-chirped microwave pulse. In order to obtain more accurate numerical results, we use a number of coupled states instead of only the selected states involved. We obtain the energy values and wave functions by solving the stationary Schrödinger equation using B-spline method. We expect to obtain the condition of complete population transfer in the multi-level ladder system.

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## 2. Theory

We use the time-dependent multilevel approach (TDMA) developed by Zhang *et al*.[23] The driving microwave pulse is designed as (in atomic units)

$$E(t) = E_0 f(t)\cos[\omega(t)t], \tag{1}$$

where $f(t) = \sin^2 \pi t/mT$, $\omega(t) = \omega_0 + \beta t$, $E_0$ is the amplitude of the microwave field, $\omega_0$ the initial frequency, $\beta$ the chirped rate, $T$ the microwave pulse period, and $m$ the number of the microwave pulse period. The Hamiltonian for the outer electron of the potassium atom takes the following form

$$H = H_0 + zE(t), \tag{2}$$

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

$$\left[-\frac{1}{2}\nabla^2 + V(r)\right]\varphi(r) = E\varphi(r), \tag{3}$$

where $V(r)$ is the core potential of an alkali-metal atom, involving the effects of penetration and polarization, and is assumed to be in the form of[24]

$$V(r) = -\frac{1}{r}\left\{(Z-1)\exp(c_1 r) + c_2 r\exp(c_3 r) + 1\right\}, \tag{4}$$

for potassium atom, with $Z = 19$, $c_1 = 3.491$, $c_2 = 10.591$, and $c_3 = 1.730$; wave function $\phi(r)$ has the form

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

with $n$, $l$, and $m$ being the principal, angular momentum, and magnetic quantum numbers, $\mathrm{Y}_{lm}(\theta,\varphi)$ the spherical harmonic function, radial function $U_{nl}(r)$ a solution for the reduced Schrödinger equation:

$$\left[-\frac{\mathrm{d}^2}{2\mathrm{d}r^2} + \frac{l(l+1)}{2r^2} + V(r)\right]U_{nl}(r) = E_{nl}U_{nl}(r). \tag{5}$$

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

$$U_{nl}(r) = \sum_{i=1}^{n} c_i B_{i,k}(r). \tag{6}$$

**Table 1.** Calculated Rydberg potassium atom energy levels of $n$ = 6–15, $l$ = 0–5 states in zero field (in Hartree), 1 Hartree=27.21 eV.

| $n/l$ | | 0 | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|---|---|
| 6 | $E^{\mathrm{MP}}$ | –0.034435058 | –0.027369895 | –0.015090411 | –0.013898657 | –0.013889008 | –0.013888889 |
| | $E^{\mathrm{MP1}}$[24] | –0.034439066 | –0.027369987 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.034442841 | –0.027364784 | | | | |
| 7 | $E^{\mathrm{MP}}$ | –0.021579092 | –0.017949549 | –0.010984297 | –0.010210947 | –0.010204183 | –0.010204082 |
| | $E^{\mathrm{MP1}}$[24] | –0.021581072 | –0.017949598 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.021577148 | –0.017941188 | | | | |
| 8 | $E^{\mathrm{MP}}$ | –0.014785766 | –0.012678313 | –0.008344289 | –0.007817418 | –0.007812580 | –0.007812501 |
| | $E^{\mathrm{MP1}}$[24] | –0.014786886 | –0.012678342 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.014783022 | –0.012671445 | | | | |
| 9 | $E^{\mathrm{MP}}$ | –0.010762069 | –0.009431190 | –0.006550232 | –0.006176450 | –0.006172902 | –0.006172840 |
| | $E^{\mathrm{MP1}}$[24] | –0.010762764 | –0.009431208 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.010759782 | –0.009426028 | | | | |
| 10 | $E^{\mathrm{MP}}$ | –0.008183161 | –0.007289521 | –0.005276925 | –0.005002715 | –0.005000049 | –0.005000001 |
| | $E^{\mathrm{MP1}}$[24] | –0.008183622 | –0.007289534 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.008181403 | –0.007285674 | | | | |
| 11 | $E^{\mathrm{MP}}$ | –0.006431556 | –0.005802738 | –0.004341177 | –0.004134317 | –0.004132271 | –0.004132232 |
| | $E^{\mathrm{MP1}}$[24] | –0.006431877 | –0.005802747 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.006430218 | –0.005799839 | | | | |
| 12 | $E^{\mathrm{MP}}$ | –0.005187713 | –0.004728586 | –0.003633620 | –0.003473856 | –0.003472254 | –0.003472223 |
| | $E^{\mathrm{MP1}}$[24] | –0.005187945 | –0.004728593 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.005186686 | –0.004726365 | | | | |
| 13 | $E^{\mathrm{MP}}$ | –0.004272738 | –0.003927337 | –0.003085766 | –0.002959882 | –0.002958605 | –0.002958580 |
| | $E^{\mathrm{MP1}}$[24] | –0.004272912 | –0.003927342 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.004271939 | –0.003925605 | | | | |
| 14 | $E^{\mathrm{MP}}$ | –0.003580123 | –0.003313780 | –0.002652985 | –0.002552073 | –0.002551041 | –0.002551020 |
| | $E^{\mathrm{MP1}}$[24] | –0.003580256 | –0.003313784 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.003579492 | –0.003312408 | | | | |
| 15 | $E^{\mathrm{MP}}$ | –0.003043237 | –0.002833551 | –0.002305195 | –0.002223085 | –0.002222240 | –0.002222222 |
| | $E^{\mathrm{MP1}}$[24] | –0.003043341 | –0.002833554 | | | | |
| | $E^{\mathrm{QD}}$[24] | –0.003042731 | –0.002832448 | | | | |

Using the boundary condition $U_{nl}(r)(r=0)=0$, where $B_{i,k}(r)$[25–30] is the $i$-th B-spline of order, and substituting Eq. (6) into Eq. (5), the following matrix equation is obtained

$$HC = ESC, \tag{7}$$

where $H$ is the Hamiltonian matrix and $S$ is the overlap matrix; $E$ and $C$ are eigenvalues and eigenvectors. By solving Eq. (7), we can obtain $E$ and $U_{nl}(r)$. In Table 1, we list the calculated $n=6$–15, $l=0$–5 state energy values ($E^{\mathrm{MP}}$) of $H_0$ by B-spline technique. It can be observed that the calculated results accord well with the theoretical values obtained from other model potentials ($E^{\mathrm{MP1}}$) and the quantum defect method ($E^{\mathrm{QD}}$) given by Schweizer *et al*.[24] In Table 2 we list the principal quantum numbers selected and the calculated Rydberg potassium atom levels of $n=70$–75, $l=0$–5 states in zero field. We select three hundred base sets to guarantee the convergence of the calculated results, so the calculated results in Table 2 are very reliable and accurate. The wavefunction of the potassium atom in the presence of microwave field $E(t)$ can be written as

$$\psi(r,t) = \sum_{i=1}^{n} a_{il}(t)\phi_i \mathrm{e}^{-\mathrm{i}E_{il}t}. \tag{8}$$

In the above equation, $a_{il}(t)$ is the coefficient of the expansion which represents the amplitude of the transition probability, $\mathrm{e}^{-\mathrm{i}E_{il}t}$ contains the phase information that determines the moment when the resonance occurs,[23] wave function $\psi(r,t)$ obeys the time-dependent Schrödinger equation,

$$\mathrm{i}\frac{\partial\psi(r,t)}{\partial t} = H\psi(r,t). \tag{9}$$

Substituting Eq. (8) into Eq. (9), and solving the time-dependent Schrödinger equation numerically, we can obtain $a_{il}(t)$. Then the probability of the outer electron inhabiting state $i$ can be written as

$$P_i = \sum_{l=0}^{l=5} |a_{il}|^2. \tag{10}$$

Using the above formula, we can obtain the state-to-state transition probability, from which we can observe the famous Rabi oscillation and some other interesting features directly.

**Table 2.** Calculated Rydberg potassium atom energy levels of $n=70$–75, $l=0$–5 states in zero field (in Hartree).

| $n/l$ | 0 | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|---|
| 70 | –0.0001087108 | –0.0001072315 | –0.0001028538 | –0.0001020498 | –0.0001020410 | –0.0001020408 |
| 71 | –0.0001055744 | –0.0001041585 | –0.0000999657 | –0.0000991952 | –0.0000991869 | –0.0000991867 |
| 72 | –0.0001025718 | –0.0001012157 | –0.0000971976 | –0.0000964588 | –0.0000964508 | –0.0000964506 |
| 73 | –0.0000996955 | –0.0000983958 | –0.0000945429 | –0.0000938341 | –0.0000938264 | –0.0000938262 |
| 74 | –0.0000969385 | –0.0000956922 | –0.0000919955 | –0.0000913151 | –0.0000913077 | –0.0000913075 |
| 75 | –0.0000942943 | –0.0000930985 | –0.0000895496 | –0.0000888962 | –0.0000888890 | –0.0000888889 |

## 3. Results

In this section, the population transfer of a potassium atom in a chirped microwave field from a lower ($n=70$) to a higher ($n=75$) state is discussed. First we investigate the population transfer of a potassium atom in a microwave field with $m=0.4$, $T=100$ ns, $E_0=30$ V/cm, $\omega_0=21.23$ GHz, and $\beta=0.0011$ GHz/ns. The obtained population transfer between atom states is illustrated in Fig. 1: about 99.4% of the $n=70$ state atoms are transferred to $n=75$. In this process, six sequential adiabatic passages are formed by a single frequency-swept pulse. The population traverses from the $n=70$ state to $n=71$, 72, 73, 74 states and finally comes to $n=75$ state. Next $\omega_0=21.19$ GHz/ns and keeping the other parameters unchanged, about 99.1% of the $n=70$ atoms are transferred to $n=75$ as shown in Fig. 2. We can see that the population in $n=75$ state slightly oscillates with time and finally can be trapped there, which could be regarded as a small Rabi oscillation induced in the $n=75$ state. From Figs. 1 and 2, we know that the population transition probability from the initial state to the final state is related to the initial frequency $\omega_0$. Hence, to control the population transfer coherently, the initial frequency $\omega_0$ must be optimized. Then we change the chirped rate $\beta=0.00106$ GHz/ns, and the other parameters are the same as those in Fig. 1. The obtained population is illustrated in Fig. 3, and about 94.3% of the $n=70$ atoms are transferred to $n=75$. In this process, about 99.3% of $n=70$ atoms are transferred to $n=71$ state, 96.5% of the population is transferred to $n=72$ state, 94.4% of the population is transferred to $n=73$ state, and 94.3% of the population is transferred to the final state. This shows that the population transition probability is related to the chirped rate $\beta$. Finally, in Fig. 4, $E_0=10$ V/cm, and the other parameters are the same as those in Fig. 1, we can find that population transfer can be achieved from $n\to70\to71\to72\to73\to74$ state to $n=75$ state; about 38.8% of the population is transferred to the final state. From all the figures, we can know that the population transition probability from the initial state to the final state is related to the initial frequency $\omega_0$, the chirped rate $\beta$ and the microwave field amplitude $E_0$.

From the above analyses, we can draw the conclusion that the efficient population transfer can be realized and trapped in

the final state eventually by modulating the initial frequency $\omega_0$, the chirped rate $\beta$, and the microwave field amplitude $E_0$, which shows that the coherent control of the population transfer from the lower states to the higher ones can be accomplished by optimizing the microwave pulse parameters.

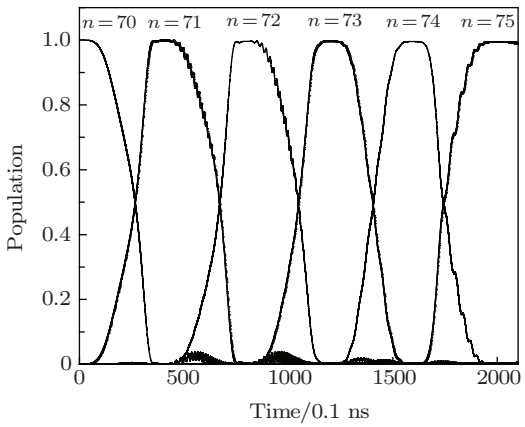



**Fig. 1.** Population transfer in the microwave field of frequency sweep from $n = 70$ to $n = 75$, with parameters $m = 0.4$, $T = 100$ ns, $E_0 = 30$ V/cm, $\omega_0 = 21.23$ GHz, and $\beta = 0.0011$ GHz/ns.

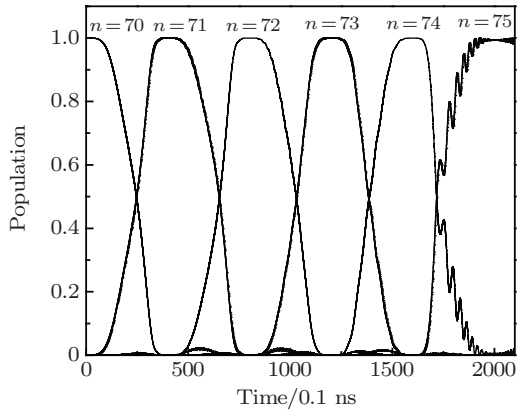



**Fig. 2.** Population transfer in the microwave field of frequency sweep from $n = 70$ to $n = 75$, with parameters $m = 0.4$, $T = 100$ ns, $E_0 = 30$ V/cm, $\omega_0 = 21.19$ GHz, and $\beta = 0.0011$ GHz/ns.

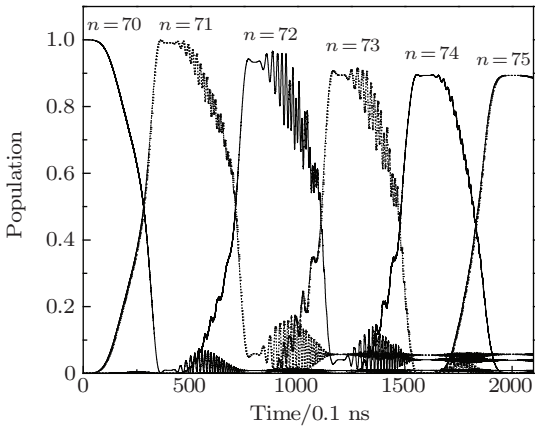



**Fig. 3.** Population transfer in the microwave field of frequency sweep from $n = 70$ to $n = 75$, with parameters $m = 0.4$, $T = 100$ ns, $E_0 = 30$ V/cm, $\omega_0 = 21.23$ GHz, and $\beta = 0.00106$ GHz/ns.

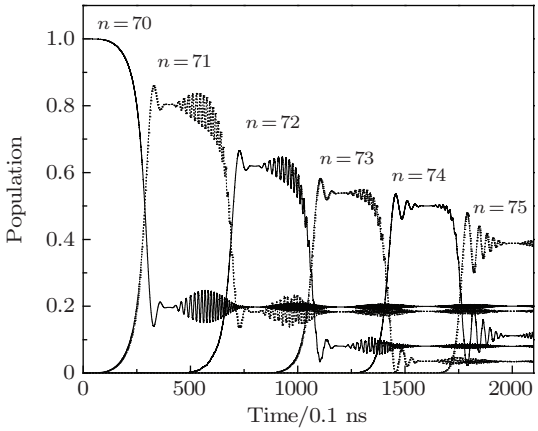



**Fig. 4.** Population transfer in the microwave field of frequency sweep from $n = 70$ to $n = 75$, with parameters taken as follows: $m = 0.4$, $T = 100$ ns, $E_0 = 10$ V/cm, $\omega_0 = 21.23$ GHz, and $\beta = 0.0011$ GHz/ns.

## 4. Conclusion

In this paper, we calculated Rydberg potassium atom energy levels of $n = 6$–$15$, $l = 0$–$5$ states in zero field by using B-spline and the calculated results are in good agreement with other theoretical values. We demonstrate that the population can be transferred to a higher state efficiently by exposing Rydberg potassium atoms to specially designed microwave pulses. The results suggest that one can determine the suitable microwave pulse parameters to achieve complete population transfer between the selected states in the multilevel system. The results also suggest that we can use this method to excite the atom to any state and trap it in that state for a long time by using a single specially designed frequency-chirped microwave pulse. In a sense, it is possible to achieve the coherent control of the population transfer of Rydberg potassium atoms by designing appropriate microwave pulses.

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