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Study of Microwave Multiphoton Transition of Rydberg Potassium Atom by Using B-Spline*

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Abstract *The B-spline expansion technique and time-dependent two-level approach are applied to study the interaction between the microwave field and potassium atoms in a static electric field. We obtain theoretical multiphoton resonance spectra that can be compared with the experimental data. We also obtain the time evolution of the final state in different microwave fields.*

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1 Introduction

Since the first experiment which was carried out by Bayfield and Koch,[1] a great deal of attention has been paid to Rydberg atoms in strong microwave fields.[2−4] The behaviors of Rydberg hydrogen and alkali-metal atoms in a static electric field and a microwave field, such as microwave ionization and transitions of Rydberg atoms, have been studied extensively.

For alkali-metal atoms, the multiphoton ionization process is now well understood in terms of Landau–Zener theory.[5] In order to further understand the relation between Landau–Zener transition and the multiphoton transition, Bloomfield *et al.*[6] and Stoneman *et al.*[7] investigated the microwave multiphoton transitions between the Rydberg states of potassium atom. The experiment of Bloomfield *et al.*[6] on highly excited potassium atoms focused on the transitions between $(n+2)s$ states and the lowest energy Stark states of the n manifolds for $n = 15 \sim 18$. The process was as follows. First, put potassium atoms in a static electric field and prepare them in the $(n+2)s$ state with a pair of laser pulses. Then, put the excited atoms in a microwave field. By tuning the static electric field from the field at which the pairs of levels nearly cross, to zero field where they are well separated, while microwave field amplitude is kept fixed, they obtained the multiphoton resonance spectra. For the $18s \rightarrow (16,\ 3)$ transition, which is a typical example of a sequence of transitions, they observed the multiphoton resonance spectra.

For the periodic microwave field, the Floquet theory is an effective method to calculate the multiphoton process. Stoneman *et al.*[7] calculated the dynamic Stark shift numerically by means of the Floquet method. Li *et al.*[8] have used the Floquet method plus an atomic model potential to calculate the multiphoton transition probability directly. For the first time the theoretical multiphoton resonance spectra they obtained could be compared with Bloomfield *et al.*'s experimental ones. However, the zero-field radial wavefunctions which they have got in the calculation are not orthogonal and their theoretical approach was very time-consuming. Using the Floquet method, the time evolution of the transition probability is not available because the method is time-independent and it is not possible to see Rabi oscillation in the multiphoton transition.

Recently, B-spline has been widely applied to the calculation of atomic and molecular physics.[9] Especially treating the ground and the lower excited states of atoms in external field, Xi *et al.*, were the first to use the B-spline.[10] They calculated the energy levels of the ground and lower excited states of hydrogen in magnetic field of arbitrary strength. Liu *et al.* computed the spectrum and life-time of the hydrogen circular states in magnetic field by using B-spline method.[11] Rao *et al.* calculated the ground and the lower excited Stark resonances of hydrogen atom in superstrong electric fields in terms of the B-spline method.[12] Zhou *et al.*[13] and Cormier *et al.*[14] used the B-spline to study the high-order harmonic generation and the above-threshold ionization of atoms in intense laser field. To our knowledge, little effort has been made to use B-spline to analyze the behavior of microwave multiphoton transition of the Rydberg potassium atom. In this paper, we present B-spline basis sets expansion for the time-dependent wavefunction of the Rydberg atoms, and apply the two-level approach to study multiphoton transition of potassium atoms in the presence of a static electric field and a microwave field. We do not only derive the multiphoton resonance spectra of the $18s \rightarrow (16,\ 3)$ transitions of potassium atom that are in excellent agreement with the experimental results of Bloomfield *et al.*'s,[6]

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but also we can extract the frequencies of the Rabi oscillations through the time evolution of population of the (16, 3) state.

2 Theory

We consider a potassium atom in a static electric field F_s and a microwave field $F_{mw}\cos(\omega t)$ (the directions of both fields are along the z axis). The time-dependent Schrödinger equation for the outer electron of the atom takes the form (atomic units)

$$\mathrm{i}\frac{\partial}{\partial t}\Psi(t) = H(t)\Psi(t)\,, \tag{1}$$

where $H(t) = H_0 + zF_s + zF_{mw}\cos(\omega t)$ is the Hamiltonian of the system under consideration, and $H_0 = -\nabla^2/2 + V(r)$ is the zero-field Hamiltonian, and F_{mw} is the amplitude of the microwave field.

In order to solve Eq. (1), we use the B-spline basis function and the one-electron model potential given by Marinescu *et al.*[15] to obtain the eigenfunctions of H_0 by means of solving the stationary Schrödinger equation. Next, choosing eigenfunctions of H_0 as the basis, we calculate the eigenstates of $H_1 = H_0 + zF_s$ through diagonalization, i.e., obtain the Stark structure of an alkali-metal atom in the static electric field. In the end, we select two eigenvectors of H_1 of interest as the basis to expand the time-dependent solution of $H(t)$. From solving the two-state close-coupling equations we obtain the multiphoton transition probability and the theoretical multiphoton resonance spectrum.

2.1 *B-spline and the Model Potential*

B splines are piecewise polynomials, which possess the characteristic of both analytical and numerical functions and are thus a very important tool for numerical calculations. Given a knot sequence on the r axis, $\{r_1 \le r_2 \le \cdots \le r_N \le \cdots \le r_{N+k}\}$, B-spline functions of order k is defined as[16]

$$B_{i,1}(r) = \begin{cases} 1\,, & r_i \le r < r_{i+1}\,, \\ 0\,, & \text{otherwise}\,, \end{cases}$$

$$B_{i,k}(r) = \frac{r - r_i}{r_{i+k-1} - r_i}B_{i,k-1}(r) + \frac{r_{i+k} - r}{r_{i+k} - r_{i+1}}B_{i+1,k-1}(r)\,. \tag{2}$$

It is immediately seen that $B_{i,k}$ is piecewise polynomials of order $k-1$ localized within (r_1, r_{N+k}), while $B_{i,k}$ is nonvanishing within (r_i, r_{i+k}). The behavior of the B-spline functions can be readily adjusted with the knot sequence, viz. the choice of knot point r_i, order k, and number of B splines N, which offers a means to optimize the B splines as a basis set to expand the wavefunctions of one or several states concerned.

The one-electron model potential given by Marinescu *et al.*[15] can well describe the motion of the valence electron for the alkali-metal atoms. The form of this potential which depends on the orbital angular momentum l of the valence electron is

$$V_l(r) = -\frac{Z_l(r)}{r} - \frac{\alpha_c}{2r^4}[1 - \mathrm{e}^{-(r/r_c)^6}]\,, \tag{3}$$

where α_c is the static dipole polarizability of the positive-ion core, while the radial charge $Z_l(r)$ is given by

$$Z_l(r) = 1 + (z-1)\,\mathrm{e}^{-a_1 r} - r(a_3 + a_4 r)\,\mathrm{e}^{-a_2 r}\,, \tag{4}$$

where z is the nuclear charge of the neutral atom and r_c is the cutoff radius introduced to truncate the unphysical short-range contribution of the polarization potential near the origin. Here α_c, r_c, a_1, a_2, a_3, a_4 are the parameters which have been given in Ref. [15].

2.2 *Solution of the Stationary Schrödinger Equation*

Due to the central symmetry of the potential, the eigenfunction of H_0 has the following form:

$$\Phi_{nlm} = R_{nl}(r)\mathrm{Y}_{lm}(\theta,\varphi)\,, \tag{5}$$

where n, l, m are principal, angular momentum and magnetic quantum numbers, respectively, $\mathrm{Y}_{lm}(\theta,\varphi)$ is a spherical harmonic function.

The radial wavefunction $R_{nl}(r)$ can be expanded as linear combination of B splines:

$$R_{nl}(r) = \sum_i D_i B_{i,k}(r)\,. \tag{6}$$

Substituting $R_{nl}(r)$ and $V_l(r)$ into the radial Schrödinger equation,

$$\Big[-\frac{1}{r^2}\frac{\mathrm{d}}{\mathrm{d}r}\Big(r^2\frac{\mathrm{d}}{\mathrm{d}r}\Big) + \frac{l(l+1)}{r^2} + V_l(r)\Big]R_{nl}(r) = E_{nl}R_{nl}(r)\,, \tag{7}$$

and then multiplying $B_{j,k}(r)$ from left and integrating with respect to r, we obtain a matrix equation. By means of diagonalizing the equation, the numerical form of $R_{nl}(r)$ can be obtained. These radial wavefunctions have the correct number of nodes.

2.3 *Basis Set*

Using the above zero-field wavefunction $\Phi_{nlm} = |nlm\rangle = \Phi_i$ as a basis set, matrix elements of the Hamiltonian $H_1 = H_0 + zF_s$ have the following form:

$$H_{1nlm,n'l'm} = \delta_{nlm,n'l'm}E_{nl} + F_s\langle nlm|z|n'l'm\rangle\,. \tag{8}$$

By diagonalizing the matrix of H_1, we obtain the eigenvalue E_k and the eigenvector $\psi_k^{(s)}$ of H_1. They obey the following equation:

$$H_1\psi_k^{(s)} = E_k\psi_k^{(s)}\,, \tag{9}$$

where

$$\psi_k^{(s)} = \sum_i C_{ki}\Phi_i\,. \tag{10}$$

2.4 *Time-Dependent Two-Level Approach*

We choose two Stark states: initial state and final state, as the new basis set. The time-dependent wavefunction of the potassium atom in the presence of static electric field together with the microwave field can be written as

$$\Psi(t) = \sum_{k=1}^{2} a_k(t)\psi_k^{(s)}\,\mathrm{e}^{-\mathrm{i}E_k t}\,, \tag{11}$$

where $a_k(t)$ is coefficient of the expansion. It represents the amplitude of the transition probability. Substituting Eq. (11) into Eq. (1), the equations of the two-level approach can be obtained. By solving these equations using the fourth-order Runge–Kutta[17] algorithm, we can get the expansion coefficient $a_k(t)$. Then the probability of the outer electron occupying the state k at time t can be written as

$$P_k(t) = |a_k(t)|^2 \,. \tag{12}$$

The long-time average transition probability is expressed as

$$\bar{P} = \frac{1}{T}\int_0^T P_k(t)\,\mathrm{d}t \,, \tag{13}$$

where T is the duration chosen to be long enough for the average.

By means of the above formula, we can calculate the mulitiphoton transition probability.

3 Results and Discussions

In the calculation, we use the B-spline functions with order $k = 7$ and number $N = 180$. The choice of knot point r_i adopts the proportional spacing in some subsection according to the principle that in the region near the core the knot points are dense, and in the region far away from the core the knot points are sparse. In calculating the eigenvalue E_k and the eigenvector $\psi_k^{(s)}$ of H_1, the basis set constructed from the zero-field wavefunctions should include infinite states in the ideal consideration, but this cannot be realized for a numerical calculation. Usually, one can truncate the basis set to include states near the interested states only. In our case, the 18s and the (16, 3) states are interesting ones. For simplicity, the notation (n, l) represents the state that connects adiabatically to the zero-field state of principal quantum number n and angular quantum number l , and the form $(n, l_i \sim l_j)$ denotes the collection of states (n, l_i), (n, l_i+1), $\cdots$, (n, l_j). The basis set is formed from the zero-field wavefunctions for (14, $3 \sim 13$), (15, $2 \sim 14$), (16, $0 \sim 15$), (17, $0 \sim 16$), (18, $0 \sim 1$), (19, $0 \sim 1$). The total number of these basis wavefunctions is 61. The result of the potassium Stark map in the neighborhood of $n = 15$, $n = 16$ ($|m| = 0$) is shown in Fig. 1. In order to justify the validity of this approximation, we have used a much larger basis set by taking 319 zero-field wavefunctions of $n = 4$ to $n = 25$ manifolds, and the error of the energies of 18s and (16, 3) from the two different basis sets is within 10^{-6} Hartree.

Bloomfield *et al.*[6] obtained the multiphoton resonance spectra by sweeping the static field from the s state crossing field to the zero field for a fixed microwave field amplitude. Their results for the 18s → (16, 3) transitions are shown in Fig. 2. Using the Stark states of 18s and (16, 3) obtained above, with fixed F_s and F_{mw}, we get transition probability $P_k(t)$ through Eq. (1), Eq. (11), and Eq. (12), and by means of Eq. (13) we obtain long-time average transition probability $\bar{P}$. We have studied how $\bar{P}$ varies with the value of F_s for fixed F_{mw}.

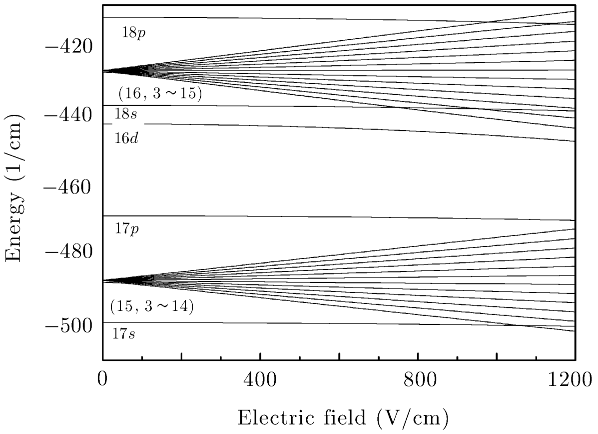

Fig. 1 Stark energy levels of potassium in the vicinity of $n = 15$, $n = 16$ ($|m| = 0$).

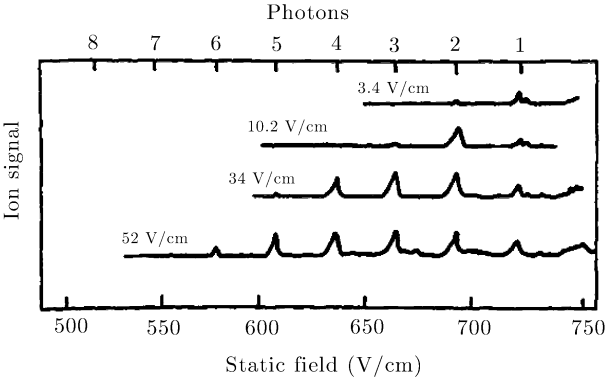

Fig. 2 Experimental multiphoton resonance spectra from the 18s state to the (16, 3) Stark state. Each trace is taken at a fixed microwave field, indicated at the left of the trace. Taken from Ref. [6].

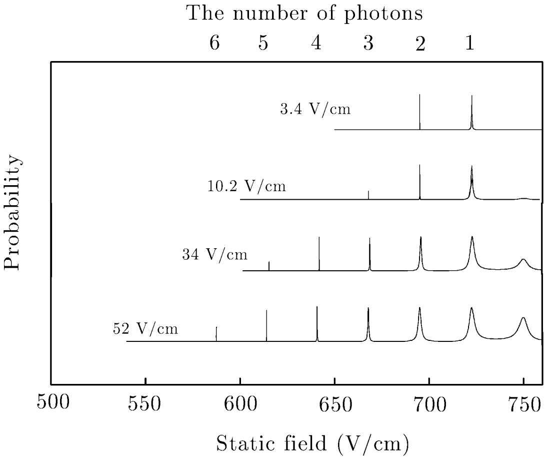

Fig. 3 Theoretical multiphoton resonance spectra from the 18s state to the (16, 3) Stark state. The probabilities of each trace adopt the same scale and are calculated by Eq. (13). Each trace is taken at a fixed microwave field, indicated at the left of the trace.

We show our results of the multiphoton resonance spectra in Fig. 3. In the calculation, the time step is taken as 0.001 ns, which is about one percent of the microwave field period, the duration T in Eq. (13) is at least 500 microwave field periods, and the frequency of the microwave field is taken as $\nu_{mw} = 10.353$ GHz, which is the same as the experiment of Bloomfield *et al.*[6] Meanwhile, in order to testify the reliability of the fourth-order Runge–Kutta algorithm for the long time, using Eq. (12) and choosing $F_s = 650$ V/cm and $F_{mw} = 3.4$ V/cm, we observe the evolution of the total probability of the electron from one to about twenty thousand microwave field periods, and find that the absolute error between the initial period and the final one is within 1×10^{-7}. Clearly, our results are in good agreement with the experimental ones. Comparing with the Floquet results given by Li *et al.*,[8] our results are also in good agreement with theirs. In Table 1, we list the static electric fields at which the multiphoton resonances appear for fixed F_{mw}. Clearly the reduced static electric field decreases with increasing number of photons absorbed for each fixed F_{mw}, but the static electric fields where the resonances occur do not depend much on the strength of F_{mw} for each fixed number of photons absorbed. Considering the Stark shifts of the 18s state and the (16, 3) state, which are shown in Fig. 4, this result is in accord with the photon point of view[7] that multiphoton resonances appear whenever the separation between the levels matches approximately an integer multiple of the energy of a photon.

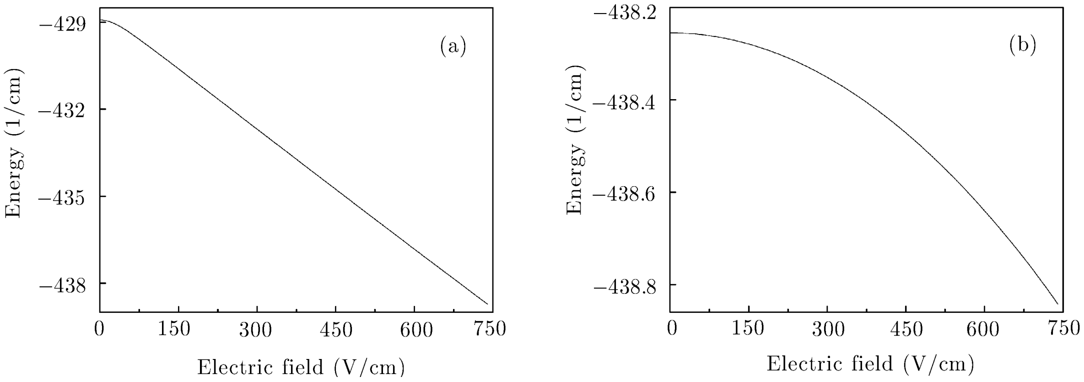

Fig. 4 (a) The calculated Stark shift of potassium atom for the (16, 3) state with static electric field from 0 V/cm to 750 V/cm; (b) The calculated Stark shift of potassium atom in the 18s state with static electric field from 0 V/cm to 750 V/cm.

Table 1 The calculated F_s at which resonances appear for different fixed F_{mw}. The frequency of the microwave field ν_{mw} is 10.353 GHz. The static electric field F_s and the amplitude of the microwave field F_{mw} are given in units of V/cm. N_p is the number of photons absorbed.

F_{mw}	F_s					
	$N_P = 1$	$N_P = 2$	$N_P = 3$	$N_P = 4$	$N_P = 5$	$N_P = 6$
3.4	722.62	695.13				
10.2	722.62	695.13	667.90			
34	722.69	695.13	667.91	640.90	614.09	
52	722.71	695.14	667.91	640.90	614.09	587.48

In order to check the reliability of the two-level approach, we also use 6, 10, 20, 30, and 40 eigenvectors of Stark states as basis sets, respectively, which are chosen in the vicinity of the 18s state and (16, 3) state symmetrically. Using these new basis sets, we calculate the positions of resonance peaks with fixed $F_{mw} = 34$ V/cm. In Table 2 , we list our results with the same F_{mw}. Comparing the results of the two-level approach with the ones of multilevel approach via Table 2, we can see that, for fixed number of photons absorbed, the maximal position error is not lager than 0.1 V/cm. At the same time, we can also see that the positions of resonance peaks obtained by using the multilevel approach with different numbers of basis sets almost have a fixed shift in contrast to the ones obtained by using the two-level approach. This is consistent with the photon point of view,[7] which has been mentioned above. In a word, the influence of the states around the 18s state and the (16, 3) state is very small, i.e., the time-dependent two-level approach warrants enough accuracy and it is an effective method in this case.

Table 2 The calculated positions of the resonance peaks using different basis sets with fixed $F_{mw} =$ 34 V/cm. The frequency of microwave field ν_{mw} is equal to 10.353 GHz. The unit of the static electric field F_s is V/cm. N_p is the number of photons absorbed.

Number of eigenvectors	F_s				
	$N_P = 1$	$N_P = 2$	$N_P = 3$	$N_P = 4$	$N_P = 5$
2	722.69	695.13	667.91	640.90	614.09
6	722.71	695.12	667.89	640.88	614.07
10	722.66	695.12	667.89	640.88	614.07
20	722.61	695.12	667.89	640.88	614.07
30	722.72	695.23	668.00	640.99	614.18
40	722.70	695.22	667.99	640.98	614.17

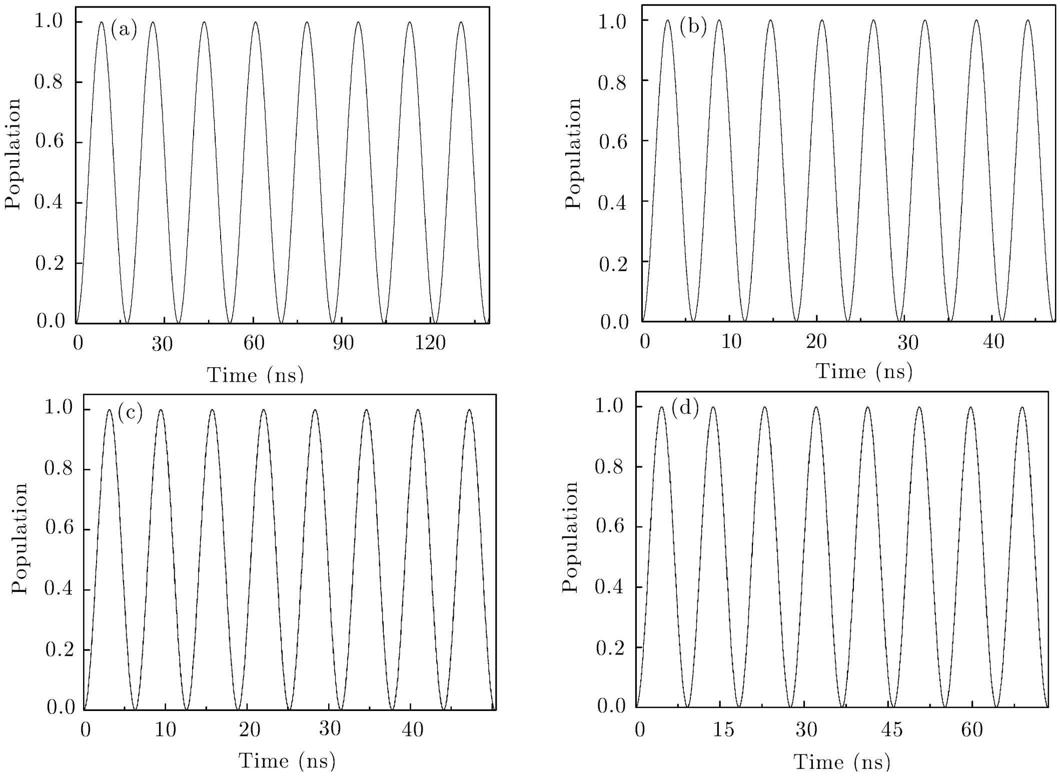

Fig. 5 The population of the (16, 3) Stark state vs. time, in the microwave field with frequency $\nu_{mw} = 10.353$ GHz. F_{mw} is the amplitude of the microwave field, F_s is the static electric field and N_p is the number of photons absorbed. (a) $F_{mw} = 3.4$ V/cm, $F_s = 722.62$ V/cm, and $N_P = 1$; (b) $F_{mw} = 10.2$ V/cm, $F_s = 722.62$ V/cm, and $N_P = 1$; (c) $F_{mw} = 34$ V/cm, $F_s = 695.13$ V/cm, and $N_P = 2$; (d) $F_{mw} = 52$ V/cm, $F_s = 667.91$ V/cm, and $N_P = 3$.

When the theoretical multiphoton resonance spectra are obtained, other related quantities such as the time evolution of the population can also be obtained. In particular, we can observe Rabi oscillations from our calculation for the one-photon, two-photon, three-photon transitions, …, and so on. Such oscillations are shown in Fig. 5, where the parameters F_{mw}, F_s, and N_p are also given. Based on the Floquet theory, Stoneman *et al.*[7] have given an equation

$$\Omega_{mw_q} = \Omega_{\text{static}}\, \mathrm{J}_q\Big(\frac{kF_{mw}}{\nu_{mw}}\Big) \tag{14}$$

to calculate the Rabi frequency. Here Ω_{mw_q} is the q-photon transition Rabi frequency, Ω_{static} is the size of the static field at the avoided crossing, J_q is a Bessel function of order q, F_{mw} is the microwave field amplitude, ν_{mw} is the microwave frequency, and k is the relative slope between the two related states. Based on the calculation of the present paper, the relative slope k between the 18s state and (16, 3) state is 386.23 MHz/(V/cm) and the size of the static field at avoided crossing Ω_{static} between the above two states is 965.18 MHz. In terms of Eq. (14), we calculate that $\Omega_{mw_1} = 66.09$ MHz, $\Omega_{mw_1} = 180.33$ MHz, $\Omega_{mw_2} = 169.36$ MHz, and $\Omega_{mw_3} = 115.36$ MHz when F_{mw} are equal to

3.4 V/cm, 10.2 V/cm, 34 V/cm, and 52 V/cm, respectively. These results are consistent with the oscillations shown in Fig. 5.

4 Conclusion

Our investigations verify that B-spline can be accurate to construct the wavefunction of Rydbery atoms. Using the B-spline expansion method and two-level approach, we present a general method to study the transitions between Rydberg Stark states of potassium atom driven by a microwave field. We have successfully obtained theoretical results, which are consistent with the experimental ones. It must be emphasized that the radial wavefunctions composed of B splines are complete enough and the present method saves significant amount of computing time in comparison with the Floquet method. This method can be widely used to calculate the transition probability and the spectrum of alkali-metal atoms in periodically and nonperiodically driven fields. It can also be used to predict some novel behaviors of alkali-metal atoms in the external fields.

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