Study of frequency-modulated excitation of Rydberg potassium atoms by using B-spline*

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With the B-spline expansion technique and a model potential of the alkali atoms, the properties of frequency-modulated excitation of Rydberg potassium atoms in a static electric field and a microwave field are investigated by using the time-dependent two-level approach. We successfully reproduce the square wave oscillations in the low frequency, the stair step population oscillations in the intermediate frequency, and the multiphoton transitions in the high frequency with respect to the unmodulated Rabi frequency, which have been observed experimentally by Noel *et al.* [Phys. Rev. A **58** 2265 (1998)]. Furthermore, we also numerically obtain the discretized Rabi oscillations predicted in the Landau–Zener accumulation model.

Keywords: B-spline, frequency-modulated field, two-level approach, model potential

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1. Introduction

Since the invention of tunable lasers, the Rydberg atoms in an external field have been intensively studied in both experiment and theory. A number of researches have focused on the behaviors of Rydberg hydrogen and alkali-metal atoms in a static electric field, in a microwave field, and in a laser field, such as energy-level crossing and anticrossing,[1−4] multiphoton transition,[5,6] multiphoton ionization,[7] population transfer,[8] and autoionization.[9] In the absence of frequency modulation, multiphoton transitions (or the standard Rabi oscillations) occur between two Rydberg Stark states driven by a microwave field. And then the presence of a frequency-modulated field makes the time evolution of the Rydberg atoms modified. It has been observed in Ref. [10] that the populations between two Rydberg Stark states present square-wave oscillations with a slow modulation, stair step population oscillations with an intermediate modulation, and multiphoton resonances with a fast modulation. The frequencies of these modulations are studied with respect to the unmodulated Rabi frequency.

To understand those phenomena, the time-dependent multi-level approach has been proposed by Zhang *et al.*,[11] which could almost reproduce all the featured experimental results in Ref. [10]. The approach was further extended to the population trapping with a designed radio-frequency field.[12] However, in the approach of Zhang *et al.*,[11] the Stark states are constructed by using the analytic wave functions obtained by solving the complicated nonlinear algebraic equations, and the numerical calculation is time-consuming, because many Stark states are involved in the close-coupling equations. Therefore, it is desirable to propose an alternative approach, which can avoid the inconvenience in constructing the basis set, but give reasonable good results and cost much less computer time.

Recently, due to the considerable advantages over the earlier basis sets, B-spline has been widely applied to the calculation in atomic and molecular physics.[13] Those works were concerned about the energy levels of the ground and the lower excited states of the hydrogen in a magnetic field of arbitrary strength,[14] the spectra and the life time of the hydrogen circular states in a magnetic field,[15] the Stark shifts and the Stark widths of the highly excited states near the classical ionization threshold of the hydrogen

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atom,[16] the high-order harmonic generation,[17] the above-threshold ionization of atoms in an intense laser field,[18] and the microwave multiphoton transition of the Rydberg potassium atoms.[19]

In this paper, we present the B-spline basis set expansion for the time-dependent wave function of the Rydberg atoms, and apply the time-dependent two-level approach to study the frequency-modulated excitation of the Rydberg Stark states of the potassium atoms in the presence of a microwave field. This approach has been established in our earlier work and shown success in the study of the microwave multiphoton transition of the Rydberg potassium atoms.[4] Our results present the square-wave oscillations in the low-frequency field, stair step population oscillations in the intermediate-frequency field, and multiphoton resonances in the high-frequency field. These calculated results are in good agreement with the experimental measurements of Noel *et al.*[10] and the theoretical calculations of Zhang *et al.*[11] We further carry out the calculation in the intermediate-frequency field with a large amplitude and obtain the discretized Rabi oscillations, which are predicted by the Landau–Zener phase accumulation model.[10]

2. Theory

Apply a static electric field $F_{\rm s}$, a microwave field $F_{\rm mw}\cos\omega_{\rm mw}t$, and a radio-frequency (rf) field $F_{\rm rf}\cos\omega_{\rm rf}t$ to Rydberg atom potassium, the directions of the three fields are along the z axis. The time-dependent Schrödinger equation for the outer electron of the atom is (in atomic units)

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

where $H(t) = H_0 + zF_{\rm s} + zF_{\rm mw}\cos\omega_{\rm mw}t + zF_{\rm rf}\cos\omega_{\rm rf}t$ is the Hamiltonian of the system under consideration, H_0 is the zero-field Hamiltonian, $F_{\rm mw}$ and $F_{\rm rf}$ are the amplitudes of the microwave and the rf fields, respectively.

To solve Eq. (1), we use the B-spline basis function and the one-electron model potential to obtain the eigenfunctions of H_0. And then, by choosing the eigenfunctions of H_0 as the basis, the eigenstates of $H_1 = H_0 + zF_{\rm s}$ can be calculated through diagonalization, i.e., we can obtain the Stark structure of the alkali-metal atom in the static electric field. We then select two eigenvectors of H_1 of interest as the basis set to expand the time-dependent solution of $H(t)$. Finally, by solving the two-state close-coupling equations, we obtain the transition probability.

2.1. The B-spline and the model potential

The B-splines are piecewise polynomials, which possess the characteristics of both analytical and numerical functions. For a given 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 are defined as[20]

$$\begin{aligned} B_{i,1}(r) &= \begin{cases} 1, & r_i \le r < r_{i+1}, \\ 0, & \text{otherwise}, \end{cases} \\ B_{i,k}(r) &= \frac{r - r_i}{r_{i+k-1} - r_i} B_{i,k-1}(r) \\ &\quad + \frac{r_{i+k} - r}{r_{i+k} - r_{i+1}} B_{i+1,k-1}(r). \end{aligned} \tag{2}$$

We can immediately see that $B_{i,k}$ is a piecewise polynomial 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 order k, number N, and knot point r_i, which offers a mean to optimize the B-splines as the basis set to expand the wavefunctions of one or several states concerned. In the process of calculation, we choose order $k = 9$, number $N = 180$, and the knot sequence in the exponential form, i.e.,[13]

$$\begin{aligned} \xi_i &= r_{\min} + (r_{\max} - r_{\min}) \frac{\mathrm{e}^{\gamma\left(\frac{i-1}{n-1}\right)} - 1}{\mathrm{e}^{\gamma} - 1}, \\ & i = 1, 2, \ldots, n. \end{aligned} \tag{3}$$

The one-electron model potential given by Marinescu *et al.*[21] can well describe the motion of the valence electron in the alkali-metal atom. The form of this potential depends on the orbital angular momentum l of the valence electron and is given by

$$V_l(r) = -\frac{Z_l(r)}{r} - \frac{\alpha_{\rm c}}{2r^4}[1 - \mathrm{e}^{-(r/r_{\rm c})^6}], \tag{4}$$

where $\alpha_{\rm c}$ is the static dipole polarizability of the positive-ion core. The radial charge $Z_l(r)$ is given by

$$Z_l(r) = 1 + (Z-1)\,\mathrm{e}^{-a_1 r} - r(a_3 + a_4 r)\,\mathrm{e}^{-a_2 r}, \tag{5}$$

where Z is the nuclear charge of the neutral atom, and $r_{\rm c}$ is the cutoff radius introduced to truncate the unphysical short-range contribution of the polarization potential near the origin. Here $\alpha_{\rm c}$, $r_{\rm c}$, a_1, a_2, a_3, and a_4 are the parameters, which have been given in Ref. [21].

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:

$$\Phi_{nlm} = R_{nl}(r)\mathrm{Y}_{lm}(\theta,\varphi), \tag{6}$$

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

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

$$R_{nl}(r) = \sum_i D_i B_{i,k}(r). \tag{7}$$

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

$$\left[-\frac{1}{r^2}\frac{\mathrm{d}}{\mathrm{d}r}\left(r^2\frac{\mathrm{d}}{\mathrm{d}r}\right) + \frac{l(l+1)}{r^2} + V_l(r)\right] R_{nl}(r) = E_{nl}R_{nl}(r), \tag{8}$$

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. The radial wavefunction will have the correct number of nodes.

2.3. The basis set

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

$$H_{1nlm,n'l'm} = \delta_{nlm,n'l'm}E_{nl} + F_\mathrm{s}\langle nlm|\, z\,|n'l'm\rangle\,. \tag{9}$$

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

$$H_1\psi_k^{(\mathrm{s})} = E_k\psi_k^{(\mathrm{s})}, \tag{10}$$

where

$$\psi_k^{(\mathrm{s})} = \sum_i C_{ki}\Phi_i. \tag{11}$$

2.4. Time-dependent two-level approach

If the two concerned states satisfy the oscillation conditions and the external field is a long pulse, we can safely use the time-dependent two-level approach to study the interaction between the atoms and the external field. In this paper, the two conditions are well satisfied, and we choose two Stark states (one is the initial state and the other is the final state) as the new basis set. The time-dependent wave function of the potassium atom in the presence of the static electric field together with the microwave and the rf fields can be written as

$$\Psi(t) = \sum_{k=1}^{2} a_k(t)\psi_k^{(\mathrm{s})}\,\mathrm{e}^{-\mathrm{i}E_k t}, \tag{12}$$

where $a_k(t)$ is the coefficient of the expansion, which represents the amplitude of the transition probability. Substituting Eq. (12) into Eq. (1) and solving these equations using the fourth-order Runge–Kutta[22] algorithm, we can obtain the expansion coefficient $a_k(t)$. Then the probability of the outer electron occupying state k at time t can be written as

$$P_k(t) = |a_k(t)|^2\,. \tag{13}$$

Using the above formula, we can calculate the multi-photon transition probability.

3. Results and discussion

For simplicity, we denote one Stark state as (n,k), where n and k are adiabatically connected with the quantum numbers n and l in the zero field, and the form $(n, k_i \sim k_j)$ denotes a collection of states (n,k_i), (n,k_i+1), …, (n,k_j). In this paper, we focus on the two Stark states 21s and 19f, and the basis set is formed by the zero-field wave functions of $(16, 2\sim 15)$, $(17, 1\sim 16)$, $(18, 0\sim 17)$, $(19, 0\sim 18)$, $(20, 0\sim 19)$, $(21, 0\sim 20)$, $(22, 0\sim 1)$, and $(23, 0)$. The total number of these basis wave functions is 111. In principle, the basis set should include infinite states, but this cannot be carried out in a real calculation. We have checked that the truncated basis in our calculation is large enough.

To check the accuracy of the time-dependent two-level approach, in which the transition caused by the external fields is limited within two Stark states only and the transition probability to the other states is very small compared to that of the concerning states, we calculate the positions of multi-photon resonance peaks by scanning a static electric with a microwave field. There already have been some experimental and theoretical works about the microwave multi-photon transition in a static electric field, see Refs. [5], [6], and [19]. We compare the calculations based on the two-level approach with Stark states 21s and $(19, 3)$ and the multi-level approach with the Stark states

in the vicinity of the 21s state and the $(19, 3)$ state symmetrically. The 10, 20, 30, 40, 50, and 60 Stark states are included in the multi-level approach. In the calculation, the amplitude and the frequency of the microwave field are $F_{\rm mw} = 7.23646$ V/cm and $\omega_{\rm mw} = 8.0$ GHz, respectively. And then, we sweep the static electric field from the s state crossing field to the zero field with a step of 0.01 V/cm. Finally, we obtain the one-photon resonance peaks under the static electric field $F_{\rm s}$, which are shown in Table 1. We can see that the influence of the states around the 21s state and the $(19, 3)$ state is very small, i.e., the time-dependent two-level approach provides enough accuracy and is an effective method in this case. Moreover, we also have checked the convergence of the two-level approach with respect to the multi-level approach with an rf modulated field (not shown here).

We have checked the accuracy of the two-level approach, and it can be used to study the Rydberg Stark atoms in a microwave field with another frequency-modulated field.

Table 1. Resonance peaks calculated by using the time-dependent multi-level approach. The n is the number of the Stark states in the basis set.

n	2	10	20	30	40	50	60
$F_{\rm s}$/V·cm^{-1}	289.38	289.38	289.38	289.38	289.38	289.38	289.39

3.1. Low-frequency field

The low frequency (or slow modulation) is the frequency far smaller than the Rabi frequency of the one-photon transition. In this paper, we choose the amplitude and the frequency of the microwave field as $F_{\rm mw} = 7.23646$ V/cm and $\omega_{\rm mw} = 8.0$ GHz, respectively. These parameters are the same as those in the experiment[10] and Zhang *et al.*'s calculation.[11] Using the two-level approach and scanning the static field, we obtain the one-photon transition resonance peak at $F_{\rm s} = 289.38$ V/cm, and the Rabi frequency is $\Omega_{\rm mw_1} = 83.9$ MHz according to Eq. (13). Then we add an rf field, and set $F_{\rm rf} = 0.6$ V/cm, and $\omega_{\rm rf} = 10.25$ MHz ($\omega_{\rm rf} \approx \frac{1}{8}\Omega_{\rm mw_1}$), which is the same as that in the experiment.[10] We obtain the population of the excited states using Eq. (13). In Fig. 1, we show our calculated square-wave oscillations in terms of microwave pulse width together with the experimental results,[10] Zhang *et al.*'s[11] calculation results, and the results of the two-state approximation based on the theory of dressed or Floquet Stark states.[5,10]

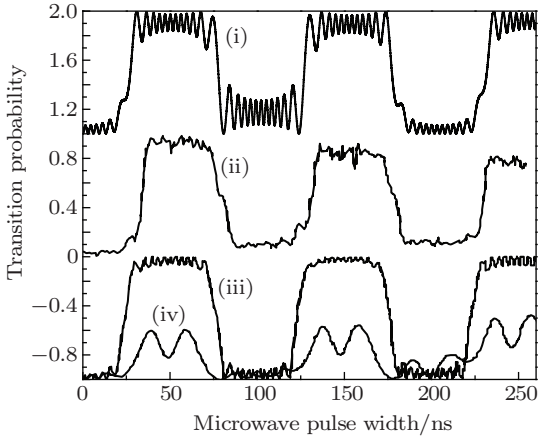


Fig. 1. Square wave oscillations in low-frequency field. Curve (i) is our calculated result; curve (ii) is the experimental result;[10] curve (iii) is the results of Zhang *et al.*;[11] and curve (iv) presents the results of the dressed theory based on Eq. (1) of Ref. [10].

As showing in curve (iv), the results of the two-state dressed theory cannot describe the square oscillations, and disagree with the experiment. It is obvious that our results [see curve (i)] are in good agreement with the experimental measurements[10] [see curve (ii)] and Zhang *et al.*'s calculation results[11] [see curve (iii)]. The characteristic of the square oscillations in the experiment has been reproduced in our theory. It can be understood as follows. When we add a static electric field to the one-photon microwave resonance under a modulated field, which begins as a cosine and whose amplitude is large initially, because of the Stark effect, this modulated field makes the system off resonance, thus the system populate in the lower state 21s initially. And then the modulated field $F_{\rm rf}\cos\omega_{\rm rf}t$ approaches zero, the two-level system makes a transition from 21s to 19f. Later, the system will be off the resonance as the cosine becomes nonzero, the population will remain in the upper states, and so on. After a careful examination, there are some structures both on the square-wave crest and in the trough of the square wave. These structures seem like experimental noise in the experimental results, but they are quiet regular both in our calculation and Zhang *et al.*'s. In comparison with

the microwave frequency, the modulated frequency is very low. In a small time interval, $F_{\rm rf}\cos\omega_{\rm rf}t$ can be considered as a constant, and the rf field can be regarded as a static electric field. Due to the presence of this affixation field, the Stark shift will change, and the Rabi oscillation will also change accordingly. In Fig. 1, we also see that the amplitudes of the oscillations on the square-wave crest and in the trough of the square wave of our results are bigger than those in Zhang *et al's*. Actually, these structures can be effectively controlled by changing the amplitude of the rf field to modify the Stark shift in our calculation.

3.2. Intermediate-frequency field

The intermediate frequency (or intermediate modulation) is the frequency that can be comparable with the Rabi frequency. The time evolution of the Rydberg Stark atom exhibits stair step population oscillations with an intermediate modulation in the experiment.[10] In Ref. [10], only the microwave Rabi frequency $\Omega_{\rm mw_1} = 108$ MHz and the frequency $\omega_{\rm rf} = 52$ MHz of the modulated-frequency field were given. But there was no information about the amplitude and the frequency of the microwave field. In Fig. 2, curve (i) shows the experimentally measured Rabi oscillation in the absence of the frequency modulation, curves (ii) and (iii) are the time evolutions of the stair step populations in the presence of the modulated-frequency field with the frequency of $\omega_{\rm rf} = 52$ MHz $\approx \frac{1}{2}\Omega_{\rm mw_1}$ and the amplitudes of 0.92 V/cm and 0.78 V/cm, respectively. We can see the constructive interference in curve (ii) and the destructive interference in curve (iii). To compare with the experiment, we set the amplitude of the static field as $F_{\rm s} = 287.62$ V/cm, and the amplitude and the frequency of the microwave field as $F_{\rm mw} = 11.07$ V/cm and $\omega_{\rm mw} = 9$ GHz, respectively. We obtain the frequency of the one-photon microwave transition, $\Omega_{\rm mw_1} = 111$ MHz. The amplitude and the frequency of the rf field are the same as those in the experiment. In Fig. 2, curves (iv), (v), and (vi) are our calculated results, and they are similar to curves (i), (ii), and (iii), respectively. It shows that our results agree well with the experimental ones.

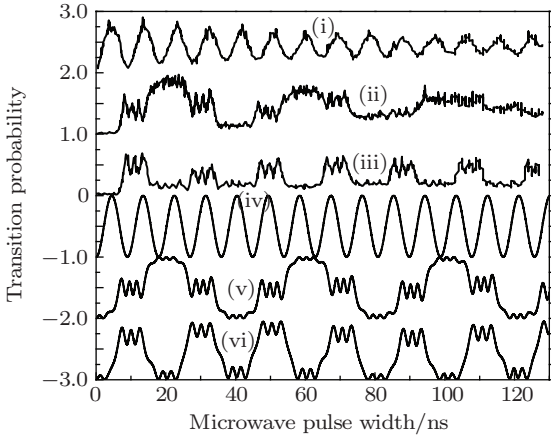


Fig. 2. Curves (i), (ii), and (iii) are the experimental results.[10] Curve (i) is the Rabi oscillation with the frequency of 108 MHz when the frequency modulation is turned off. Curves (ii) and (iii) are the time evolutions of the two-level atom system in the presence of frequency modulation with the frequency of 52 MHz and the amplitudes of 0.92 V/cm and 0.78 V/cm, respectively. Curves (iv), (v), and (vi) are our theoretical results corresponding to curves (i), (ii), and (iii), respectively.

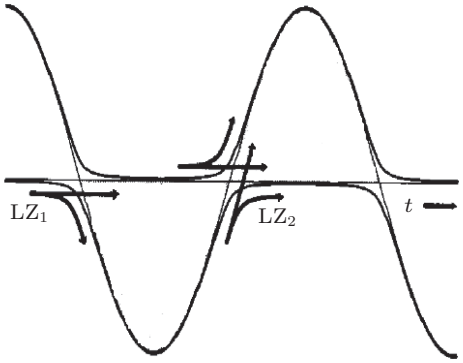


Fig. 3. Schematic representation of the Landau–Zener phase accumulation model,[10] where LZ_1 and LZ_2 are the Landau–Zener paths at the first and the second avoided crossings, respectively.

In terms of the Landau–Zener phase accumulation model,[10] we can understand the above results. As shown in Fig. 3, each microwave dressed state as a function of time in the sinusoidal modulating field is considered. When the rf field changes to zero, the multiphoton transition condition is satisfied, and there is a nonzero transition amplitude to the other state. The system is now in a superposition of the two dressed states. Therefore, when the avoided crossing is traversed again, there will be interference between the two transition amplitudes. The interference depends on the relative phase accumulated by the two states during their evolutions in between the crossings. Different rf amplitudes and frequencies would result in different half and single cycle transition probabilities. According to the Landau–Zener phase accumulation model,[10] when the rf frequency is $\Omega_{\rm rf} = \frac{1}{2}\Omega_{\rm mw_1}$, the amplitude of the rf field is set to be large, the discretized Rabi oscillations will appear, which have not

been investigated in the experiment because of the limitation of the apparatus. In theory, we use the same parameters of the microwave field above, and then set the amplitude and the frequency of the rf field as $F_{\rm rf} = 10.0$ V/cm and $\Omega_{\rm rf} = 54$ MHz, respectively, we obtain the discretized Rabi oscillations shown in Fig. 4.

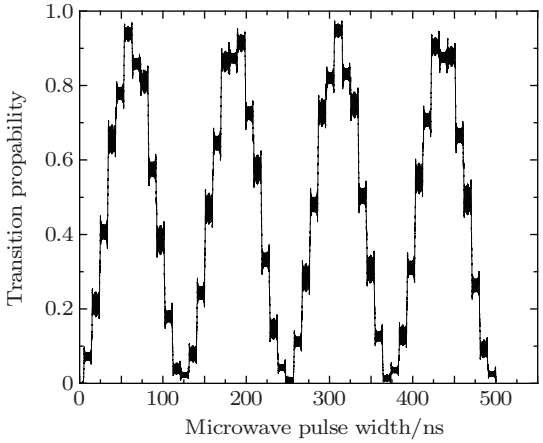


Fig. 4. Discretized Rabi oscillations with the modulated field of the frequency $\Omega_{\rm rf} = 54$ MHz, and the amplitude $F_{\rm rf} = 10.0$ V/cm.

3.3. High-frequency field

The high frequency (or fast modulation) is the modulated frequency, which is much higher than the Rabi frequency. To obtain the rf multiphoton transition, we first turn off the rf field and set the microwave amplitude at $F_{\rm wm} = 17.45$ V/cm and the microwave frequency at $\omega_{\rm mw} = 9$ GHz. Next, by scanning the static field, we obtain the position of the one-photon resonance when $F_{\rm wm} = 287.61$ V/cm and the microwave Rabi frequency $\Omega_{\rm mw_1} = 159$ MHz. And then, turn on the rf field and set the frequency at $\omega_{\rm rf} = 330$ MHz, which is about $2\Omega_{\rm mw_1}$, and the amplitude at $F_{\rm rf} = 4.13$ V/cm. Finally, we obtain the multiphoton resonances by scanning the electric field from 284.5 V/cm to 289.5 V/cm. In Fig. 5, we show our theoretical results (upper curve) and the experimental results (lower curve). We notice that our results agree well with the experimental ones.

From Fig. 5, we can see that the widths of the odd rf resonances are wider than the even ones because of the variation in the rf Rabi frequencies defined as[10]

$$\Omega_{{\rm rf}_q} = \Omega_{\rm mw_1} {\rm J}_q\left(\frac{SE_{\rm rf}}{\omega_{\rm rf}}\right), \tag{14}$$

where $\Omega_{{\rm rf}_q}$ is the q-photon transition Rabi frequency, ${\rm J}_q$ is a Bessel function of order q, and S is the relative slope between the two related states. From this equation, we can learn that for one particular amplitude of the rf field, there are narrow resonances when $\Omega_{{\rm rf}_1}$ and $\Omega_{{\rm rf}_3}$ are small, and large resonances when $\Omega_{{\rm rf}_0}$ and $\Omega_{{\rm rf}_2}$ are large.

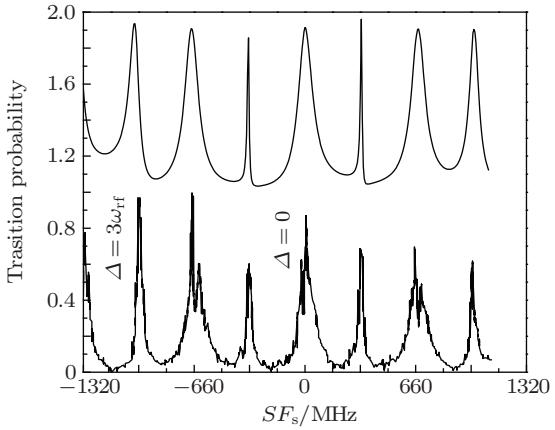


Fig. 5. Multiphoton resonance spectra with a modulated field ($F_{\rm rf} = 4.13$ V/cm and $\omega_{\rm rf} = 330$ MHz). The abscissa axis is labeled by $SF_{\rm s}$, where $F_{\rm s}$ is the static field amplitude. The upper curve is our calculation result, and the lower curve is the experimental multi-photon resonance spectrum from Ref. [10]. The carrier resonance is labeled as $\Delta = 0$ and the third sideband is labeled as $\Delta = 3\omega_{\rm rf}$.[10]

4. Conclusion

In this paper, we have presented a time-dependent two-level approach employing the B-spline as the basis set and a more accurate potential model of the alkali atoms to study the prosperities of the Rydberg atoms in a frequency modulation field together with a static electric field and a microwave filed. As an alternative of the multi-level approach proposed by Zhang *et al.*,[11] our method also provides a quantitative description of the experimental results in Ref. [10]. Meanwhile, the Stark state constructed using the B-spline as the basis set is accurate and easily calculated numerically, and the close-coupling equations can be solved quickly using the time-dependent two-level approach. Based on our model, we have calculated the square oscillations in the low-frequency field, the stair step population oscillations and the discretized Rabi oscillations in the intermediate-frequency field, and the multiphoton transitions in the high-frequency field. Our calculated results are in good agreement with the experimental ones[10] and Zhang *et al.*'s calculations.[11]

Our method has been successfully applied to study the characteristics of the Rydberg atoms in a frequency modulated field. Its accuracy has been

tested by comparing with the experimental measurements and the other theoretical calculations in this paper. This method has also been applied to study the microwave multiphoton transition[19] and the multiphoton Rabi oscillations[23] of the Rydberg potassium atom by our group. The accuracy of the Stark state involved in our model has also been tested by our group[4] through the positions and the widths of the anticrossings for potassium Rydberg Stark states against the experimental results. All of these works have shown that the B-spline expansion technique is powerful. And our model can be used to predict some novel behaviors of the alkali-metal atoms in the external fields, such as static electric fields, microwave fields, and radio-frequency fields.

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