Multiphoton Rabi oscillations between highly excited Stark states of potassium

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(Received 16 April 2011; revised manuscript received 16 September 2011; published 15 November 2011)

We have applied a nonperturbative resonant theory to study the Rabi frequency of microwave multiphoton transitions between two Rydberg states of potassium in a static electric field. The Stark electric dipole moments used to calculate the Rabi frequency are determined by the Stark states' wave functions, which are obtained by the diagonalization method. The frequencies of the Rabi oscillations are in good agreement with either experimental ones or ones calculated by the time-dependent close-coupling method and the Floquet theory. Furthermore, we are able to show that the size of avoided crossings between the $(n+2)s$ and $(n,3)$ states can be predicted from the Stark electric dipole moment and the difference of the two Stark states' energy at a given resonance.

DOI: 10.1103/PhysRevA.84.053414 PACS number(s): 33.40.+f, 32.60.+i, 32.80.Rm, 32.30.Bv

I. INTRODUCTION

In the last few decades, a lot of attention has been paid to the behaviors of Rydberg atoms in external fields, such as the study related to the Stark structure [1], the positions and widths for avoided crossings of Rydberg atoms in electric fields [2,3], microwave ionization [4–6], microwave multiphoton Rabi oscillations [7], and frequency-modulated excitation [8] of Rydberg atoms. The microwave multiphoton resonances between $(n+2)s$ and $(n,3)$ in Rydberg potassium have been observed [9–12]. The initial state referred to is labeled as an s state, despite the fact that the angular momentum quantum number is not a good quantum number in the static electric field. The wave function of this state remains essentially unchanged below the avoided crossings with the Stark manifold. The manifold state that the s state first intersects is labeled by its parabolic quantum number as the $(n,3)$ state because it adiabatically connects to the nf state in the zero field, and the wave function of this state is a superposition of the wave functions of all states with $l \geqslant 3$ due to the external electric field and thus is not a state of definite parity. Consequently, the multiphoton resonances between the Stark states of Rydberg potassium atom occur when the separation between the levels is equal to an integer times of the microwave frequency. In Fig. 1 we show the Stark map of potassium in the vicinity of $n = 19$ ($|m| = 0$). As shown in Fig. 1, the (19,3) states exhibit a linear Stark shift and the $21s$ state has a small quadratic Stark shift. The $21s$ and (19,3) states have an avoided crossing at 303.8 V/cm [13] due to short-range interaction between the Rydberg electric electron and the K^+ core. We use arrows to indicate the static fields at which the three- and five-photon resonances occur for a microwave frequency of 9.1 GHz. The microwave multiphoton process is well understood using Floquet theory and Landau-Zener theory [7,8,10,12,14]. The connection between Landau-Zener transitions in slowly varying field point of view and multiphoton transitions in the photon point of view have been examined.

In general, the major limitation of the conventional Floquet techniques is that they are applicable only to a monochromatic problem where the Hamiltonian is periodic in time. Although such a bottleneck has been circumvented with the development of the so-called many-mode Floquet theorem (MMFT) [15], the analytic treatment of the effective multi-mode Hamiltonian is still a complicated task. The analytical two-mode Floquet description of microwave multiphoton resonances between $(n+2)s$ and $(n,3)$ in Rydberg potassium has been constructed by Lung Ko *et al.* [16]; it is effective only for the case of two harmonically related fields. Furthermore, the Floquet analysis of microwave multiphoton resonance based on a parameterized two-level system requires the size of the avoided crossing and the relative slope [8,14] between the Stark states of $(n+2)s$ and $(n,3)$, which are determined by experiment. Here, the relative slope is the rate of change of energy with the amplitude of the static electric field. In the Floquet analysis, the mean value of the relative slope was used. In fact, the relative slope between the Stark states of $(n+2)s$ and $(n,3)$, which is not a constant, usually varies with respect to the static field. Thus, the Floquet analysis is somewhat a rough approximation for these two-level systems of potassium. Because the multiphoton Rabi frequencies at nonperturbative intensities are especially important for understanding complicated multiphoton process, so it is essential to give a more exact, pure theory description of this problem.

In this paper we apply a nonperturbative resonant theory to study the Rabi frequency of microwave multiphoton transitions between the $(n+2)s$ and $(n,3)$ Stark states of highly excited potassium. By using the well-established B-spline expansion technique [13,17–21] and a parametric model potential, we obtain the Stark states' wave functions by the diagonalization method, and then the Stark dipole matrix elements, which are related to the amplitude of the static electric field are determined at the exact position where we calculate the Rabi frequency. For two-mode multiphoton transitions, the nonperturbative resonant theory only requires that the multiphoton Rabi frequency is far less than the waves' frequencies; it can be easily used for the two-level system driven by either the so-called frequency-modulated field or two harmonically related microwave fields, and two frequency-incommensurate microwave fields. Furthermore, we find a formula for the size of avoided crossings between the $(n+2)s$ and $(n,3)$ states through the comparison between the nonperturbative resonant theory and Floquet theory.

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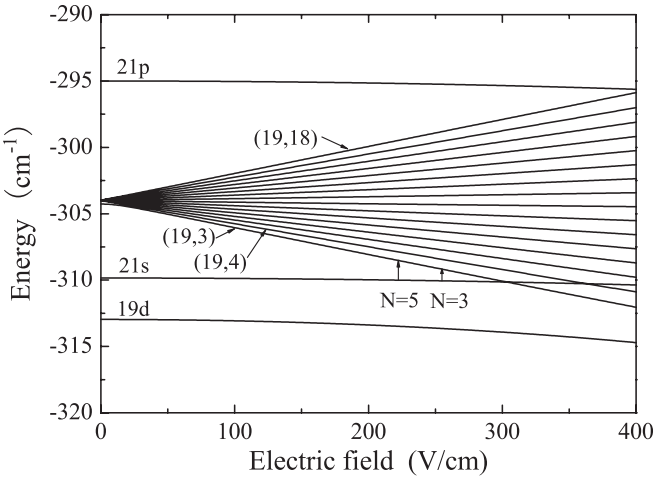

FIG. 1. Stark map of potassium in the vicinity of $n = 19$ ($|m| = 0$). The arrows indicate the positions of the three- and five-photon resonances at 9.1 GHz

II. THEORETICAL METHOD

A. Nonperturbative resonant approach for multiphoton resonant excitation

A general nonperturbative resonant approach (NPRA) to problems involving multiphoton resonant excitation of atoms was introduced by Avetissian *et al.* [22,23]. We consider a two-level system in the presence of a electromagnetic field, the Hamiltonian for the system is presented in the form

$$\widehat{H} = (\varepsilon_1 + V_{11})|1\rangle\langle 1| + (\varepsilon_2 + V_{22})|2\rangle\langle 2| + (V_{12}\,|1\rangle\,\langle 2| + \text{H.c.}) \tag{1}$$

where

$$V_{ij} = -d_{ij} F_{\text{mw}} \cos\omega_{\text{mw}} t \tag{2}$$

and d_{ij} is the matrix element of the electric dipole moment, F_{mw} is the amplitude of the microwave field, ω_{mw} is the angular frequency of the microwave field. The V_{11} and V_{22} account for interaction due to the mean dipole moments and these terms are very important for the multiphoton resonance. For our two-level system, both $(n,3)$ and $(n+2)s$ states are affected by static and microwave fields, so, it is reasonable to contain d_{11}, or equivalently, V_{11} in Eq. (1). The wave function of the system can be written in the form

$$|\psi(t)\rangle = a_1(t) e^{-i(\varepsilon_1 t + \int_0^t V_{11} dt)}\,|1\rangle + a_2(t) e^{-i(\varepsilon_2 t + \int_0^t V_{22} dt)}\,|2\rangle. \tag{3}$$

From the time-dependent Schrödinger equation

$$i\frac{\partial\,|\psi(t)\rangle}{\partial t} = \widehat{H}\,|\psi(t)\rangle \tag{4}$$

we can obtain the equations for the probability amplitudes

$$\begin{aligned} i\frac{da_1(t)}{dt} &= V_{12} e^{-i[(\varepsilon_2-\varepsilon_1)t + \int_0^t V_{22}dt - \int_0^t V_{11}dt]} a_2(t), \\ i\frac{da_2(t)}{dt} &= V_{12}^* e^{-i[(\varepsilon_1-\varepsilon_2)t + \int_0^t V_{11}dt - \int_0^t V_{22}dt]} a_1(t), \end{aligned} \tag{5}$$

rewritten as

$$\begin{aligned} i\frac{da_1(t)}{dt} &= F_{12}(t) a_2(t), \\ i\frac{da_2(t)}{dt} &= F_{12}^*(t) a_1(t), \end{aligned} \tag{6}$$

where

$$F_{12} = V_{12} e^{-i[(\varepsilon_2-\varepsilon_1)t + \int_0^t V_{22}dt - \int_0^t V_{11}dt]} \tag{7}$$

and F_{12}^* denotes the complex conjugation of F_{12}. With the help of expansion in terms of Bessel function

$$e^{ix\sin\alpha}\cos\alpha = \frac{1}{x}\sum_{s=-\infty}^{\infty} s J_s(x) e^{is\alpha}, \tag{8}$$

the function F_{12} can be represented in the following form

$$F_{12} = \sum_{s=-\infty}^{\infty} \eta_{12}(s) e^{i(\varepsilon_1-\varepsilon_2+s\omega_{\text{mw}})t}, \tag{9}$$

where

$$\eta_{12}(s) = -\frac{d_{12}}{d_{22}-d_{11}}(\omega_{\text{mw}} s) J_s\left((d_{22}-d_{11})\frac{F_{\text{mw}}}{\omega_{\text{mw}}}\right) \tag{10}$$

represent the coupling of levels by photons. The probabilities of multiphoton transitions between these levels have maximal values for the resonant transitions

$$\varepsilon_1 - \varepsilon_2 + n\omega_{\text{mw}} \simeq 0, \quad n = \pm 1, \pm 2, \pm 3, \ldots. \tag{11}$$

Hence, the function F_{12} can be represented in the following form

$$F_{12} = [\Omega_{12} + f_{12}]\, e^{i\delta t}, \tag{12}$$

where

$$\Omega_{12} = -\frac{d_{12}}{d_{22}-d_{11}}(\omega_{\text{mw}} n) J_n\left((d_{22}-d_{11})\frac{F_{\text{mw}}}{\omega_{\text{mw}}}\right), \tag{13}$$

$$f_{12} = \sum_{s\neq n} \eta_{12}(s) e^{i[(s-n)\omega_{\text{mw}}]t}, \tag{14}$$

and f_{12} is a rapidly oscillating function on the scale of waves' period. We introduce the resonance detunings

$$\delta = \varepsilon_1 - \varepsilon_2 + n\omega_{\text{mw}}. \tag{15}$$

Following the nonperturbative resonant approach introduced by Avetissian *et al.* [22,23] and separating slow and rapid oscillations, we obtain the following set of equations for the time average probability amplitudes:

$$i\frac{d}{dt}\begin{pmatrix}\overline{a_1(t)}\\ \overline{a_2(t)}\end{pmatrix} = \begin{pmatrix}\Delta & \Omega_{12}\\ \Omega_{12}^* & -\Delta^* - \delta\end{pmatrix}\begin{pmatrix}\overline{a_1(t)}\\ \overline{a_2(t)}\end{pmatrix}. \tag{16}$$

Here, Ω_{12} provides the coupling between state $|1\rangle$ and $|2\rangle$ and causes multiphoton transitions between them. Δ describes the detuning from dynamic Stark shifts. When the resonance condition is satisfied, the resonance detuning δ is neglected. Then the analytical solution of Eq. (16) for the system situated initially in the state $|1\rangle$ is

$$\overline{a_2(t)} = -i\frac{\Omega_{12}}{\sqrt{\Omega_{12}^2 + \Delta^2}} \sin\sqrt{\Omega_{12}^2 + \Delta^2}\, t. \tag{17}$$

For a given microwave field, the population for upper level $|2\rangle$ will oscillate between the two states as a function of time given by the Rabi equation

$$P(t) = \frac{\Omega_{12}^2}{\Omega_{12}^2 + \Delta^2} \sin^2\left(\sqrt{\Omega_{12}^2 + \Delta^2}t\right). \tag{18}$$

When the resonance condition is satisfied, the generalized multiphoton Rabi frequency is $2\Omega_{12}$. Thus, the microwave multiphoton Rabi frequency can be predicted from the Stark electric dipole moments, the amplitude $F_{\rm mw}$, and the frequency $\omega_{\rm mw}$ of the microwave field. Note that according to Eq. (18), the maximum value of $P(t)$ is 1 for resonance. If the maximum value of $P(t)$ is smaller than 1, obviously, the detuning Δ from dynamic Stark shifts cannot be neglected.

The set of Eq. (16) has been derived using the assumption that the amplitudes $a_i(t)$ are slowly varying functions on the scale of the electromagnetic wave's period that put in place the restrictions: $(|\Omega_{12}|,|\Delta|) \ll \omega_{\rm mw}$.

B. Time-dependent close-coupling method

In this paper, we employ the time-dependent close-coupling (TDCC) method used by Zhang *et al.* [14,24] and Jin *et al.* [17] to solve the time-dependent Schrödinger equation. The one-electron Hamiltonian for an alkali-metal atom in a static electric field (the direction of field is taken along the z axis) and a microwave field is as follows

$$H = H_0 + Fz + zF_{\rm mw}\cos\omega_{\rm mw}t, \tag{19}$$

where $H_0 = -\frac{1}{2}\nabla^2 + V(r)$ is the field-free Hamiltonian, F is the amplitude of the static electric field. The wave function $\psi(t)$ of the potassium atom in the two fields obeys Eq. (4). The one-electron potential model developed by Marinescu *et al.* [25] 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 of the valence electron, l, is

$$V_l(r) = -\frac{Z_l(r)}{r} - \frac{\alpha_c}{2r^4}[1 - e^{-(r/r_c)^6}], \tag{20}$$

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

$$Z_l(r) = 1 + (z-1)e^{-a_1 r} - r(a_3 + a_4 r)e^{-a_2 r}, \tag{21}$$

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. α_c is the static dipole polarizability of the positive-ion core. Here r_c, a_1, a_2, a_3, a_4 are the l-dependent parameters, which have been given in Ref. [25]. Due to the central symmetry of the potential, the zero-field wave function has the following form:

$$\Phi_{nlm} = R_{nl}(r)Y_{lm}(\theta,\varphi), \tag{22}$$

where n, l, m are principal, angular, and magnetic quantum numbers, respectively. $Y_{lm}(\theta,\varphi)$ is a spherical harmonic function, and $R_{nl}(r)$ is the radial wave function.

By using the diagonalization method, in which the bases are chosen from B-splines, we obtain the radial wave function $R_{nl}(r)$ in its numerical form. The detailed description of the diagonalization method used in the calculation can be found in Ref. [13]. These radial wave functions have the correct number of nodes, and are orthonormal due to the distinctive characteristics of B-spline functions.

Using the zero-field wave functions Φ_{nlm} as a basis set, the matrix elements of the Hamiltonian $H_1 = H_0 + Fz$ are given as

$$H_{nlm,n'l'm} = \delta_{nlm,n'l'm}E_{nl} + F\langle nlm|z|n'l'm\rangle. \tag{23}$$

By diagonalizing the matrix of H_1, we obtain the eigenvalues E_k and the eigenvectors $\psi_k^{(s)}$ of H_1. $\psi_k^{(s)}$ is referred to as the Stark state wave function, which is a linear combination of the zero-field wave functions Φ_{nlm}, in the following form:

$$\psi_k^{(s)} = \sum_n \sum_{l=|m|}^{n-1} U_{n,l}^{n_1,n_2}(F)\Phi_{nlm}, \tag{24}$$

where $U_{n,l}^{n_1,n_2}(F)$ are the elements of the unitary matrix that projects the Stark states, labeled by the parabolic quantum numbers n_1,n_2, on to the corresponding zero-field states. Within the electric dipole approximation, the Stark electric dipole moment d_{ij} may be written in the following form:

$$\begin{aligned} d_{ij} &= \left\langle \psi_i^{(s)} \middle| P^{(1)} \middle| \psi_j^{(s)} \right\rangle \\ &= \left\langle \psi_i^{(s)} \middle| rC_0^{(1)} \middle| \psi_j^{(s)} \right\rangle. \end{aligned} \tag{25}$$

It necessary to point out that the off-diagonal element about operator r experiences a rapid decrease when the quantum number difference between the two related states increases, so only those eigenvectors whose eigenvalues are close to the states we considered are to be selected as the bases. This fact ensures a finite character for the set, but high accuracy to the calculation. Using $\psi_k^{(s)}$ as the basis set, the time-dependent wave function of the potassium can be written as

$$\psi(t) = \sum_k a_k(t)\psi_k^{(s)}e^{-iE_k t}, \tag{26}$$

where $a_k(t)$ is the coefficient of the expansion. Substituting Eq. (26) into Eq. (4), we obtain a set of coupled equations. Solving these equations by Runge-Kutta algorithm, we can get $a_k(t)$. Then the probability of the outer electron inhabiting in the state k is

$$P_k = |a_k|^2. \tag{27}$$

Using the above formula, we can calculate the state-to-state transition probability, from which the Rabi oscillations can be observed. The long-time average transition probability is expressed as follows:

$$\overline{P} = \frac{1}{\tau}\int_0^\tau P_k(t)dt, \tag{28}$$

where τ is the duration chosen to be long enough for the average. By means of the above formula, we can obtain the multiphoton transition probability and the multiphoton resonance spectra are obtained by scanning the static field.

III. RESULTS AND DISCUSSION

A. Stark electric dipole moments

In numerical calculation, the $21s$ and the (19,3) are the interested states, so the basis set should be in the neighborhood of the two states. For simplicity, the form $(n,l_i \sim l_j)$ denotes

TABLE I. The calculated Stark electric dipole moments, which are given in atomic units.

F(V/cm)	$\|d_{11}\|$	$\|d_{12}\|$	$\|d_{22}\|$
287.37	44.323	8.455	467.534
285.62	44.020	7.629	467.712
287.2	44.293	8.367	467.551
271.2	41.688	4.249	468.937
255.4	39.223	2.867	470.053
239.8	36.809	2.156	471.007
239.7	36.794	2.153	471.013

the contiguous manifold states (n,l_i), (n,l_i+1),..., (n,l_j). The basis set is formed from the zero-field wave functions for (17,2 ∼ 16), (18,2 ∼ 17), (19,0 ∼ 18), (20,0 ∼ 19), (21,0 ∼ 20), (22,0 ∼ 1). The total number of these basis wave functions is 93. By making this approximation, the computing time is saved largely with little accuracy lost. In order to test the convergence of this approximation, we have used a much larger basis set by taking 418 zero-field wave functions for (10,2 ∼ 9), (11,2 ∼ 10), (12,0 ∼ 11), (13,0 ∼ 12),..., (30,0 ∼ 29), (31,0 ∼ 1). The relative deviation of the energy of the 21s Stark state at $F = 280$ V/cm derived from the two basis sets is within 0.0001.

In the calculation, the multiphoton resonance spectra are obtained by scanning the static field from the position of the avoided crossing between the 21s and (19,3) states to zero field for a fixed microwave field, and the positions of the N-photon resonances ($N = 1,2,3,\ldots$) are obtained accurately. The Stark states' wave functions are obtained by diagonalizing the Stark Hamiltonian with the fixed static electric field for a given resonance. Stark electric dipole moments d_{ij} are then determined [1,12]. In Table I, we list the values of the Stark electric dipole moments for the one- to four-photon resonances at 9.1 GHz, and the values of Stark electric dipole moments for the carrier resonance and the third sideband resonance are also listed.

B. Microwave multiphoton Rabi oscillations

The main results in this paper are the microwave multiphoton Rabi frequencies. According to NPRA theory, the coupling between the 21s and (19,3) states due to a microwave field is given by Eq. (13), and the generalized multiphoton Rabi frequency is $2\Omega_{12}$. We can also obtain the multiphoton Rabi frequency directly through numerical experiment by the TDCC method since $P(t)$ is a standard sine or cosine function of time. In addition, the multiphoton Rabi frequency can be derived from Floquet theory [7] as follows:

$$\Omega_{mwN} = \Omega_0 J_N\left(\frac{(k-\alpha F)F_{\rm mw}}{\omega_{\rm mw}}\right), \tag{29}$$

where $\Omega_{{\rm mw}N}$ is the N-photon transition Rabi frequency, Ω_0 is the size of the avoided crossing between the Stark states, J_N is a Bessel function of order N, k is the relative slope between the $(n+2)s$ and $(n,3)$ states or the dipole moment of the $(n,3)$ Stark state [12], and α is the dipole polarizability of the $(n+2)s$ state. For the 21s state, $\alpha/2\pi = 0.195\,{\rm MHz/(V/cm)^2}$ [7]. In the present calculation, the size of the avoided crossing Ω_0 between 21s and (19,3) states is 340.8 MHz and the relative slope of the 21s and (19,3) states is $k/2\pi =$ 604 MHz/(V/cm).

We first calculate the Rabi frequency between 21s and (19,3) states in the microwave field using NPRA theory and Floquet theory. In the calculation, we set $F_{\rm mw} = 7.236$ V/cm, $\omega_{\rm mw}/2\pi = 8$ GHz and adjust the amplitude of the static field, and we observe the single-photon Rabi oscillation with the Rabi frequency $\Omega_{\rm mw1} = 84.96$ MHz. According to Floquet theory and NPRA theory, the Rabi frequency is 83.19 MHz and 85.39 MHz, respectively. If one neglects the quadratic Stark shifts of the 21s state, and uses a value of $k = 549$ MHz/(V/cm) [8] for the relative slope, the Rabi frequency is 81 MHz, which is in good agreement with the experiment [8]. So NPRA theory works well in calculating the Rabi frequency for microwave multiphoton transitions between two Rydberg states of potassium in a static electric field. Note that the one- and two-photon transitions are driven by the microwave fields with very small amplitude, while higher N-photon transitions require a constant additional microwave field amplitude for each additional photon. In fact, stronger microwave fields are required to drive the higher-order transitions, which occur at lower static fields. We set the microwave field frequency $\omega_{\rm mw}/2\pi = 9.1$ GHz, and sweep the static field for a fixed microwave field. Then the positions of the multiphoton resonances are obtained accurately. In Table II, we list the Rabi frequencies of the N-photon resonant excitation for $N = 1,2,3,4$. One can see the good agreement between NPRA theory, Floquet theory, and the numerical results by TDCC except for the 4-photon transition with $F_{\rm mw} =$ 40 V/cm. Note that the TDCC is multistate description, which relies more on the real behavior of the potassium atom, and the results of TDCC are the calculated Rabi frequencies of the multiphoton resonances from 21s to (19,2 ∼ 18) Stark states, the grand total of 18 states that straddle the 21s and (19,3) states are included in the calculations. They are considered as the most accurate ones among these three methods. Obviously, the results by NPRA theory are better than the ones from Floquet theory in comparison with TDCC. Both the NPRA theory and Floquet theory here are dealing with the two-level system, but the NPRA theory is more accurate. The reason is that in the NPRA description, the precise Stark dipole matrix elements that are related to the amplitude of the static electric field are determined at the exact position where we calculate the Rabi frequency, but in the Floquet analysis, the mean value of the relative slope was used.

TABLE II. The calculated Rabi frequencies for the $\omega_{\rm mw}/2\pi =$ 9.1 GHz microwave field, The values of Rabi frequencies are given in units of MHz. N is the number of photons absorbed. The positions of the multiphoton resonances F and the amplitudes of the microwave fields $F_{\rm mw}$ are given in units of V/cm.

	$N=1$	$N=2$	$N=3$	$N=4$	$N=4$
$F_{\rm mw}$	10	35	47	85	40
F	287.2	271.2	255.4	239.8	239.7
TDCC	102.04	135.00	106.96	142.15	33.77
NPRA	102.39	135.83	102.27	143.75	24.62
Floquet	100.02	131.56	98.82	135.99	23.48

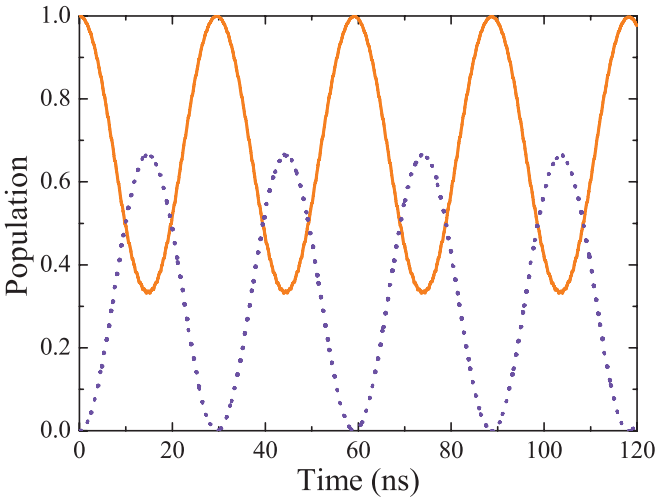

FIG. 2. (Color online) Rabi oscillations [21s, solid orange line; (19,3), dotted violet line] for four-photon resonance at $\omega_{\rm mw}/2\pi = 9.1$ GHz with $F_{\rm mw} = 40$ V/cm.

In Fig. 2 we show the Rabi oscillations for four-photon resonance at $\omega_{\rm mw}/2\pi = 9.1$ GHz with $F_{\rm mw} = 40$ V/cm. Note that the maximum of the population for the upper-level (19,3) state is smaller than 1, and it is equal to 0.67. The steady-state transition probability is one half of the maximum of the population for upper level (19,3) state, equals 0.335, and the resonance is weak enough. When the value $2\Omega_{12} = 24.62$ MHz is substituted into Eq. (18), we obtain the dynamic Stark shift $\Delta = 8.64$ MHz, which is required to meet the resonance condition. When Δ is taken into account, the corrected Rabi frequency is 30 MHz, which approaches to the numerical result 33.77 MHz. If the microwave field amplitude is increased, the multiphoton transition will be brought into resonance by a larger dynamic Stark shift (e.g., the four-photon resonance at $\omega_{\rm mw}/2\pi = 9.1$ GHz with $F_{\rm mw} = 85$ V/cm).

C. Radio frequency multiphoton Rabi oscillations

Nole *et al.* [8] have presented a detailed experimental study of the frequency-modulated excitation between the 21s and (19,3) states. For fast modulation, the two-level atom is driven by both the microwave and rf fields. In the limit where the modulation frequency is large compared to $\Omega_{\rm mw1}$, the rf resonances are well separated. The rf multiphoton Rabi frequency derived from Floquet theory is given by

$$\Omega_{{\rm rf}_N} = \Omega_{\rm mw1} J_N\left(\frac{kF_{\rm rf}}{\omega_{\rm rf}}\right), \tag{30}$$

where J_N is again a Bessel function of order N, but N refers to the number of rf photons being absorbed. $F_{\rm rf}$ is the rf field amplitude, $\omega_{\rm rf}$ is the rf field frequency.

According to NPRA theory [22,23], with the help of expansion in terms of Bessel function

$$e^{i\alpha\sin\omega t} = \sum_{s=-\infty}^{\infty} J_s(\alpha)e^{is\omega t}. \tag{31}$$

The extension to the calculation of the Rabi frequency for a two-color multiphoton process in a two-level system is not difficult, the coupling between the $(n+2)s$ and $(n,3)$ states due to a microwave and rf field is given by:

$$\Omega'_{12} = -\frac{d_{12}}{d_{22}-d_{11}}(\omega_{\rm mw1}n + \omega_{\rm mw2}m) \times J_n\left((d_{22}-d_{11})\frac{F_{\rm mw1}}{\omega_{\rm mw1}}\right) J_m\left((d_{22}-d_{11})\frac{F_{\rm mw2}}{\omega_{\rm mw2}}\right), \tag{32}$$

where $F_{\rm mw1}$ and $F_{\rm mw2}$ are the microwave and rf field amplitudes, and $\omega_{\rm mw1}$ and $\omega_{\rm mw2}$ are the microwave and rf field angular frequencies, respectively. Note that the population for upper-level $|2\rangle$ oscillates between the two states with a Rabi frequency given by $2\Omega'_{12}$.

In the rf multiphoton resonances experiment [8], the microwave amplitude was set to yield a microwave Rabi frequency of $\Omega_{\rm mw1} = 156$ MHz, the rf frequency was set as $\omega_{\rm mw2}/2\pi = 330$ MHz $\approx 2\Omega_{\rm mw1}$, and its amplitude as $F_{\rm mw2} = 4.13$ V/cm. By scanning the static electric field, a well-separated rf multiphoton resonance spectra was obtained. Next, they fixed the static field at an rf resonance and measured Rabi oscillations for various rf field amplitudes. Such measurements along with the theoretical predictions for two rf resonances, the carrier and the third sideband, were carried out.

Setting the microwave amplitude $F_{\rm mw1} = 17.45$ V/cm, frequency $\omega_{\rm mw1}/2\pi = 9$ GHz, and the rf field amplitude $F_{\rm mw2} = 4.13$ V/cm, frequency $\omega_{\rm mw2}/2\pi = 330$ MHz, respectively, which are the same as those in the experiment, then scanning the static field amplitude from 284 V/cm to 289 V/cm, the position of the N-photon rf resonance ($N = 0,1,2,3$) is obtained. The positions for the carrier resonance and the third sideband resonance are 287.37 V/cm and 285.62 V/cm, respectively. Stark electric dipole moments d_{ij} are then determined.

Note that the values of Stark electric dipole moments are given in atomic units. When these values are substituted into Eq. (32), we obtain the rf multiphoton resonance Rabi

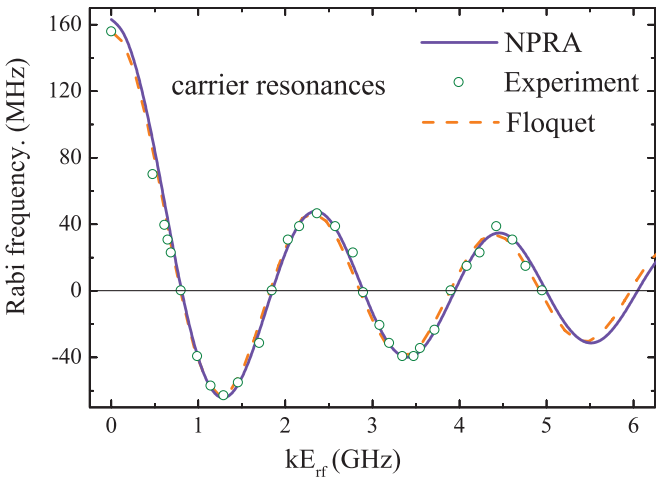

FIG. 3. (Color online) The comparison between the theoretical calculation (NPRA, solid violet line; Floquet, dashed orange line) and the experiment (open circles) for various rf amplitudes with the static field tuned to the carrier resonance. Experimental data are from Ref. [8]. The microwave and rf parameters are given in the text.

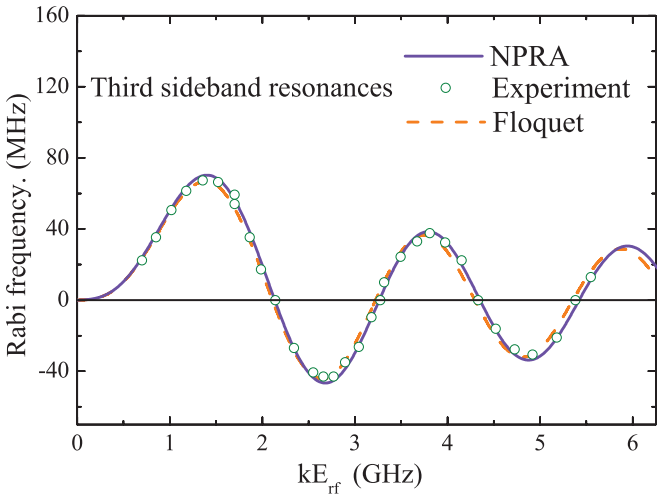

FIG. 4. (Color online) The comparison between the theoretical calculation (NPRA, solid violet line; Floquet, dashed orange line) and the experiment (open circles) for various rf amplitudes with the static field tuned to the third sideband resonance. Experimental data are from Ref. [8]. The microwave and rf parameters are given in the text.

frequencies. Figure 3 shows the results for various rf amplitudes at the carrier resonance while the experimental results and results by Floquet theory are also presented. Figure 4 shows the similar results for the third sideband resonance. In Figs. 3 and 4, we can see the good agreement between NPRA theory, Floquet theory, and the experimental results.

D. The size of avoided crossing

In general, the energy difference of two relevant levels is a function of the electric field. The minimal value of this function is at the point where the position of avoided crossing is defined, and the size of avoided crossing is defined as this minimal value. By scanning the static field [2,13,21], one can calculate the position and the size of a avoided crossing.

From the comparison between the NPRA theory and Floquet theory or the comparison between Eq. (13) and Eq. (29), the size of the avoided crossing between the $(n+2)s$ and $(n,3)$ states can be written as

$$\Omega_0 = 2\left|\frac{d_{12}}{d_{22}-d_{11}}\right|\Delta E, \tag{33}$$

where ΔE is the difference of the two states' energy at the N-photon resonance. Thus, we suggest that the size of avoided crossings between the $(n+2)s$ and $(n,3)$ states can be predicted from the Stark electric dipole moments and the difference of the two Stark states' energy. For the case of the one- to four-photon resonant excitation in Table II, the size of avoided crossings determined by Eq. (33) are 1.356 V/cm, 1.367 V/cm, 1.371 V/cm, 1.362 V/cm, and 1.363 V/cm, respectively, and the mean value is 1.364 V/cm. By scanning the static field, the calculated size of avoided crossings is 1.347 V/cm. Note the frequency sizes are converted to field sizes by using the conversion factors 506 MHz/(V/cm) given by Stoneman *et al.* [2]. Experimental result given by Stoneman *et al.* [2] is 1.4 V/cm, and calculated results of Stoneman *et al.* [2] and Jin *et al.* [13] are 1.3 V/cm and 1.34 V/cm, respectively. Excellent agreement is presented. Note that we use the values of the difference of the two states' energy directly instead of $\omega_{\rm mw}n$. If the steady-state transition probability equals $\frac{1}{2}$, one can use $\omega_{\rm mw}n$ as the value of the difference of the two states' energy at a given resonance, but error from dynamic Stark shifts can not be neglected.

IV. CONCLUSION

In this article we have applied a nonperturbative resonant theory to study the Rabi frequency for microwave multiphoton transitions between two Rydberg states of potassium in a static electric field, and this approach can be easily extended to the calculation of the Rabi frequency for two-color or rf multiphoton process. Our calculated results by NPRA theory have been compared with experimental ones together with theoretical ones by the TDCC method and Floquet theory. It has been shown that the NPRA theory works very well in calculating the multiphoton Rabi frequency. In conclusion, the Rabi frequency of the microwave or rf multiphoton resonance can be predicted from the Stark electric dipole moments, which are determined by the Stark states' wave functions at the exact position where we calculate the Rabi frequency. It must be emphasized that both the NPRA and Floquet theory are used to tackle the problem of a general two-level system in microwave fields. Only the numerical method we used is restricted to the case of alkali-metal atoms. Hence, NPRA theory can be directly used in other two-level systems, meanwhile, we can also conclude that the known multiphoton Rabi frequency dependence of Eq. (13) and Eq. (32) can be employed as a method for measuring transition dipole moments between two highly excited atomic states. Furthermore, the size of avoided crossings between the $(n+2)s$ and $(n,3)$ states can also be obtained from the NPRA theory; further accurate calculation is needed. This issue is currently under investigation.

ACKNOWLEDGMENTS

This work was supported in part by the National Natural Science Foundation of China under Grant No. 11044007 and the Specialized Research Fund for the Doctoral Program of Higher Education of China under Grant No. 20096203110001. Y. L. H. gratefully acknowledges the help of Professor Xiao-Xin Zhou and is grateful to Dr. Cheng Jin and Xiao-Yong Li for the help on the numerical methods and useful discussions.

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