Autler–Townes splitting in photoelectron spectra of K_2 molecule*

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(Received 11 March 2011; revised manuscript received 25 April 2011)

We investigate Autler–Townes splitting in the photoelectron spectra of K_2 molecule driven by pump-probe pulses via employing the time-dependent wave packet approach. It is found that the magnitude of Aulter–Townes splitting varies with the wavelength of the intense laser pulse. In particular, the phenomenon of Aulter–Townes splitting vanishes for the far-off resonance of the pump pulse. Also, the split peaks of Autler–Townes in the case of resonant pump pulse give us an approach to directly obtaining the transition dipole moment of a molecule.

Keywords: Autler–Townes splitting, photoelectron spectra, K_2 molecule, wave packet

PACS: 33.60.+q, 42.50.Hz, 33.20.Xx **DOI:** 10.1088/1674-1056/21/2/023302

1. Introduction

Femtosecond pulses have opened the possibility to create, observe, and control the internal dynamics of molecules.[1−5] The various properties of molecules, for example, molecular-bond softening and stabilization,[6−8] above-threshold ionization and dissociation,[9,10] Rabi oscillations,[11,12] light-induced potentials,[13−16] etc. have been investigated extensively in intense fields. The well-known Autler–Townes (AT) splitting[17] was observed first in an atomic beam of sodium[18] and an He–Ne discharge laser,[19] separately. This phenomenon is then studied in atoms,[20−24] single molecules,[25,26] and quantum dots.[27−31] Unlike atoms, molecules are characterized by additional degrees of freedom, such as vibrations and rotations. The effects of these further degrees of freedom are equivalent to a system including many rovibronic states. Typically, the density of vibrational quantum states increases rapidly with the electronic energy, the observation of AT splitting is therefore more difficult for molecular system and so far, only a few experimental[32−34] and theoretical[35,36] studies have addressed the phenomenon in molecular system. According to early findings, the AT splitting effect in molecular system plays a profound role in observing and controlling the dynamic properties of molecules, such as molecule alignment and the configuration of potential energy surface in a strong field.

As is well known, the influence of molecular vibration is an important factor in the AT splitting, especially, if the vibrational wave packet dynamics becomes important in molecular system, the AT splitting effect would be washed out.[35,36] This means that the molecules, targeted for investigating the AT effect in molecular system, have some "special properties", such as, small shift of the potential between the excited state and the ground state, low vibrational energy on the excited electronic state, etc. After considering those points, K_2 molecule is an excellent prototype molecule to present the AT splitting effect in a molecular system.

In this paper, we theoretically investigate the nature of the AT splitting in the photoelectron spectra of K_2 molecule ad we discuss the effects of the pump laser intensity and/or wavelength on AT splitting for three cases: resonant pulse, near resonant pulse, and far-off resonant pulse. The corresponding features are different at different wavelengths of pump pulses.

The rest of the present paper is organized as follows. In Section 2, we show the model of K_2 molecule and the method in our theoretical investigation. In Section 3, we present our numerical results of AT splitting in photoelectron spectra of K_2 molecule. The conclusion and the outlook are briefly summarized in Section 4.

*Project supported by the National Natural Science Foundation of China (Grant Nos. 91021009 and 10874102) and the Research Fund for the Doctoral Program of Higher Education, China (Grant No. 200804220004).

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2. Model and theoretical framework

In this paper, we model the system of K_2 molecule by three states: the ground state $|X\rangle^1\Sigma_g^+$, excited state $|B\rangle^1\Pi_g$, and ionization state $|X^+\rangle^2\Sigma_g^+$. A similar model of K_2 molecule is also employed in the K_2 experiment of Ref. [37]. In our treatment, we consider the case that pump and probe laser fields are simultaneously applied to the K_2 molecule. The pump field couples with the ground state $|X\rangle^1\Sigma_g^+$ and excited state $|B\rangle^1\Pi_g$ under the dipole moment approximation. The probe field is used to detect the emitted photoelectron from the ionization state $|X^+\rangle^2\Sigma_g^+$. We ignore the rotational degree of freedom in our study, since the rotational motion of the K_2 molecule can be essentially frozen on the femtosecond time scale.

The K_2 molecule system satisfies the time-dependent Schrödinger equation

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\Psi = \mathscr{H}\Psi. \tag{1}$$

The Hamiltonian $\mathscr{H}$ of the system can, in the Born–Oppenheimer approximation, can be written as

$$\begin{aligned}\mathscr{H} &= \mathscr{H}_s + \mathscr{H}' \\ &= \mathscr{T} + \mathscr{V} + \mathscr{H}',\end{aligned} \tag{2}$$

where $\mathscr{T}$ is the operator of the kinetic energy of nucleus, $\mathscr{V}$ is the potential energy of the system and $\mathscr{H}'$ is the interaction between K_2 molecule and pump and probe fields. For the three-state model, the wave functions can be written in the column vector as

$$\Psi = (\psi_X, \psi_B, \boldsymbol{\psi}_{\mathrm{ion}})^{\mathrm{T}}, \tag{3}$$

where ψ_X, ψ_B and $\boldsymbol{\psi}_{\mathrm{ion}}$ are the wave functions for the ground state $|X\rangle$, the excited state $|B\rangle$ and the ionization state, respectively. The ionization state is a continuum state and it is discretized into a band of quasicontinuum states in terms of the energies of emitted photoelectron for the numerical calculations. The ionization state $\boldsymbol{\psi}_{\mathrm{ion}}$ can be further expressed as

$$\boldsymbol{\psi}_{\mathrm{ion}} = (\psi^{(1)}, \psi^{(2)}, \ldots, \psi^{(\mathscr{N})})^{\mathrm{T}}, \tag{4}$$

where $\mathscr{N}$ represents the number of discrete states of K_2 ion.

The kinetic energy $\mathscr{T}$ can be expressed as

$$\mathscr{T} = -\frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial R^2}\mathscr{I}, \tag{5}$$

where μ is the reduced mass of the nucleus, R is the distance between the nuclei in K_2 molecule.

$$\mathscr{I} = \begin{pmatrix} 1 & 0 & \mathfrak{O} \\ 0 & 1 & \mathfrak{O} \\ \widetilde{\mathfrak{O}} & \widetilde{\mathfrak{O}} & \mathbb{I} \end{pmatrix},$$

where $\mathfrak{O} = (0, 0, \ldots, 0)$ is a zero vector with $\mathscr{N}$-component, $\tilde{\mathfrak{O}}$ is the transverse of the vector $\mathfrak{O}$, and $\mathbb{I}$ is an $\mathscr{N} \times \mathscr{N}$ unit matrix.

The potential $\mathscr{V}$ can be expressed as

$$\mathscr{V} = \begin{pmatrix} V_X & 0 & \mathfrak{O} \\ 0 & V_B & \mathfrak{O} \\ \widetilde{\mathfrak{O}} & \widetilde{\mathfrak{O}} & \mathbb{V} \end{pmatrix}, \tag{6}$$

where V_X and V_B refer to the potential curves of ground state $|X\rangle$ and excited state $|B\rangle$, respectively. $\mathbb{V}$ is an $\mathscr{N} \times \mathscr{N}$ matrix, which is the discretized states of K_2 ionization state $|X^+\rangle$. It can be written as

$$\mathbb{V} = V_{X^+}\mathbb{I} + \begin{pmatrix} \mathscr{E}^{(1)} & 0 & \cdots & 0 \\ 0 & \mathscr{E}^{(2)} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \mathscr{E}^{(\mathscr{N})} \end{pmatrix}, \tag{7}$$

where $\mathscr{E}^{(i)} = (i-1)\Delta\mathscr{E}$ $(i = 1, 2, \ldots, \mathscr{N})$ is the energies of the emitted photoelectron.

The interaction $\mathscr{H}'$ between K_2 molecule and pump and probe fields is given by

$$\mathscr{H}' = \begin{pmatrix} 0 & W_{XB} & 0 \\ W_{XB} & 0 & \mathbb{W}_{Bi} \\ 0 & \widetilde{\mathbb{W}}_{Bi} & \mathbb{O} \end{pmatrix}, \tag{8}$$

where $\mathbb{W}_{Bi} = \left(W_{Bi}^{(1)}, W_{Bi}^{(2)}, \ldots, W_{Bi}^{(\mathscr{N})}\right)$ is the vector of coupling between $|B\rangle$ and $|X^+\rangle$ states via probe field with $\mathscr{N}$-component, and $\mathbb{O}$ is the $\mathscr{N} \times \mathscr{N}$ *zero* matrix. The coupling between two states via external laser field is given as follows:

$$\begin{aligned} W_{XB} &= \hbar\Omega_{XB}\cos(\omega_{\mathrm{pump}}t), \\ W_{Bi} &= \hbar\Omega_{Bi}\cos(\omega_{\mathrm{probe}}t), \end{aligned} \tag{9}$$

where $\Omega_{XB} = (1/\hbar)\boldsymbol{\mu}_{XB}(R)\cdot\mathscr{E}_{\mathrm{pump}}f_{\mathrm{pump}}(t)$ and $\Omega_{Bi} = (1/\hbar)\boldsymbol{\mu}_{Bi}(R)\cdot\mathscr{E}_{\mathrm{probe}}f_{\mathrm{probe}}(t)$ are the Rabi frequencies of pump and probe fields, respectively; $\boldsymbol{\mu}_{XB}(R)$ $(\boldsymbol{\mu}_{Bi}(R))$ is the transition dipole moment between $|X\rangle$ and $|B\rangle$ states ($|B\rangle$ and $|X^+\rangle$ states); $\mathscr{E}_{\mathrm{pump}}$ and $\mathscr{E}_{\mathrm{probe}}$ are the amplitudes of pump and probe fields; ω_{pump} and ω_{probe} are their angular frequencies. The envelopes of pulses $f_{\mathrm{pump}}(t)$ and

$f_{\rm probe}(t)$ are taken to be in the Gaussian form as follows:

$$f_{\rm pump}(t) = {\rm e}^{-4\ln 2(t/\tau_{\rm pump})^2},$$
$$f_{\rm probe}(t) = {\rm e}^{-4\ln 2(t/\tau_{\rm probe})^2}, \qquad (10)$$

where $\tau_{\rm pump}$ and $\tau_{\rm probe}$ are the widths of pump and probe pulses.

The time-dependent population in each electronic state is obtained from the numerical calculation of

$$P_i(t) = \int {\rm d}R|\psi_i(R,t)|^2, \qquad (i = X, B, {\rm ion}). \quad (11)$$

The photoelectron spectrum, as a function of the electronic energy $\mathscr{E}^{(j)}$, can be expressed as[35,36,38,39]

$$P(\mathscr{E}^{(j)}) = \lim_{t\to\infty}\int {\rm d}R|\psi^{(j)}(R,t,\mathscr{E}^{(j)})|^2,$$
$$(j = 1, 2, \ldots, \mathscr{N}). \qquad (12)$$

The potential energy $\mathscr{V}$ of K_2 molecule is taken from Refs. [40]–[42] and its schematic diagram is shown in Fig. 1. These potentials of K_2 molecule are in good agreement with relevant spectroscopic constants of experimental results.[43,44]

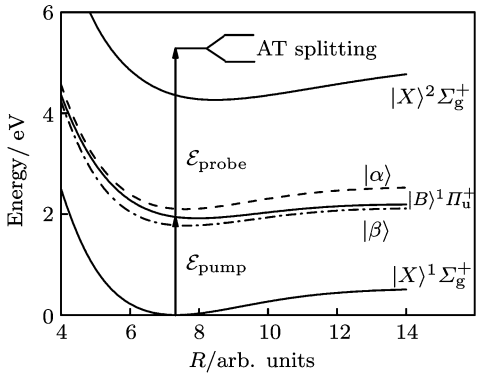


Fig. 1. Potential energy curves of K_2 molecule[38−40] employed in this work.

For the case of the ultrafast femtosecond pulse in this paper, we can employ the Condon approximation.[35,36] The transition dipole moments of K_2 molecule are taken from Refs. [37], [45], and [46]. In our calculations, the energies of the emitted photoelectron $\mathscr{E}^{(j)}$ span over $0 \sim 1.0$ eV and the number of discrete states of K_2 ion, $\mathscr{N}$, equals 100.

3. Results and discussion

The time-dependent Schrödinger equation Eq. (1) is exactly solved by "split-operator Fourier" method.[46−52] Initially, the K_2 molecule populates on its vibrational ground state of $|X\rangle$ state. The pump field excites the molecule onto $|B\rangle$ state. In order to avoid the influence of molecular vibration on the AT splitting of K_2 molecule, the ultrashort laser pulse of width 30 fs is taken.[36]

In Fig. 2, we show the populations on electronic states, each as a function of evolutionary time and their corresponding photoelectron spectra for the case of four different intensities of pump laser field. The laser wavelength of pump pulse $\lambda_{\rm pump}$ is taken to be the energy difference between $|X\rangle$ and $|B\rangle$ states at the equilibrium position R_0 of $|X\rangle$ state, namely, $\hbar\omega_0 = V_B(R_0) - V_X(R_0)$ or the wavelength $\lambda_0 = 640$ nm. Because the K_2 molecule is excited by the ultrafast femtosecond pulse, the nucleus motion is almost frozen during the interaction with external fields and the transition between electronic states takes place mainly in the region around R_0. The left column of Fig. 2 (namely, panels (a), (b), (c) and (d)) shows the populations and the right column (namely, panels (e), (f), (g) and (h)) displays their corresponding photoelectron spectra. In our calculations, the intensities of pump pulse are chosen to be I_0 for panels (a), (e), $2.25I_0$ for panels (b), (f), $4I_0$ for panels (c), (g), and $6.25I_0$ for panels (d), (h), which correspond to

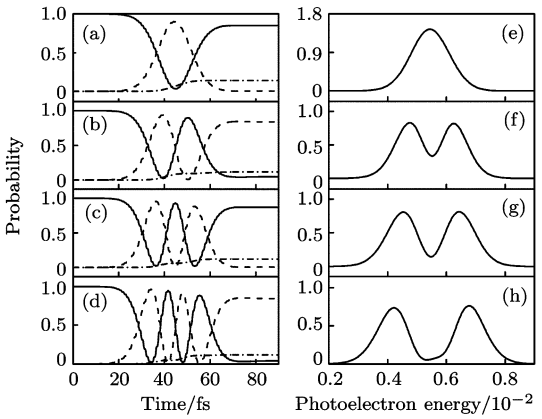


Fig. 2. Populations of each electronic state (left column) and the photoelectron spectra (right column) of K_2 molecule at the fixed pump and probe laser wavelengths and widths of Gaussian pulses. The panels (a) and (e) are for pump laser intensity I_0, panels (b) and (f) for $2.25I_0$, panels (c) and (g) for $4I_0$, and panels (d) and (h) for $6.25I_0$. The parameters are as follows: the pump laser intensity $I_0 = 1.3\times10^{11}$ W/cm^2, $\lambda_{\rm pump} = 640$ nm, $\tau_{\rm pump} = 30$ fs and the probe laser intensity is $0.1I_0$, $\lambda_{\rm probe} = 450$ nm, $\tau_{\rm probe} = 30$ fs. The populations on state $|X\rangle$ are denoted with solid lines, state $|B\rangle$ with dash lines and state $|X^+\rangle$ with dash-dot lines.

pulse area $\Theta = 2\pi$, 3π, 4π, and 5π, respectively. It should be noted that all parameters are taken to be the same as those in the experiment of Ref. [37] except

for the intensities and widths of the pump and probe pulses. The values of these parameters are listed in the caption of Fig. 2.

From the left panels of Fig. 2, we can see that the population on each electronic state shows Rabi oscillation during the interaction between the K_2 molecule and external fields. This can be interpreted as wave packets trapped in the adiabatic potentials created by the laser pulse.[53] Under the excitation of the ultrashort and intense pump pulse laser, which is resonant with the two states $|X\rangle$ and $|B\rangle$ of K_2 molecule, the wave packets jump rapidly back and forth between the $|X\rangle$ and $|B\rangle$ states during the field-molecule interaction. The period of Rabi oscillations depends on pulse area Θ. The complete oscillations between $|X\rangle$ and $|B\rangle$ states are achieved when the pulse area Θ is equal to 2π and 4π separately and the population on the ground state $|X\rangle$ is depleted when the pulse area Θ is equal to 3π and 5π separately.

The right panels of Fig. 2 show their corresponding photoelectron spectra for the intensities of $1.0I_0$, $2.25I_0$, $4I_0$, $6.25I_0$, respectively. As the intensities of the pump pulse increase, the peaks of photoelectron spectra show the AT splitting with symmetric profiles. When this happens, from the dressed state theory,[54] the excited state $|B\rangle$ is dressed by the pump laser field and splits into two substates $|\alpha\rangle$ and $|\beta\rangle$ (we assume that the energy of $|\alpha\rangle$ state is higher than that of $|\beta\rangle$ state. This is shown in Fig. 1). From Fig. 2, we can know that the intensity of pump pulse determines the magnitude of AT splitting. Also, with the increase of laser intensity, the two symmetric profiles move further away from each other.

According to the dressed state theory, the separation between two splitting peaks Δ_S, for a system interaction with an intense field, is given by[55]

$$\Delta_S = (\Delta_1^2 + \Omega_{XB}^2)^{1/2}, \tag{13}$$

where $\Delta_1 = \omega_{\text{pump}} - \omega_0$ is the detuning frequency of laser field.

In the case of $\Delta_1 = 0$, we have

$$\Delta_S = \Omega_{XB}. \tag{14}$$

Figure 3 displays the separation Δ_S between two AT splitting peaks, each as a function of pump laser intensity I at resonant wavelength $\lambda_{\text{pump}} = 640$ nm. The solid squares denote our exact numerical results and the solid line is for the results of Eq. (14). From the figure, we can obtain that our numerical results are in good agreement with those from dressed state Eq. (14) and the amplitude of AT splitting depends linearly on $\sqrt{I}$ for the case of $\omega_{\text{pump}} = \omega_0$. It should be noted that the slope of the $\Delta_S \sim \Omega_{XB}$ line in Fig. 3 can be used to infer the transition dipole moment μ_{XB} of K_2 molecule.

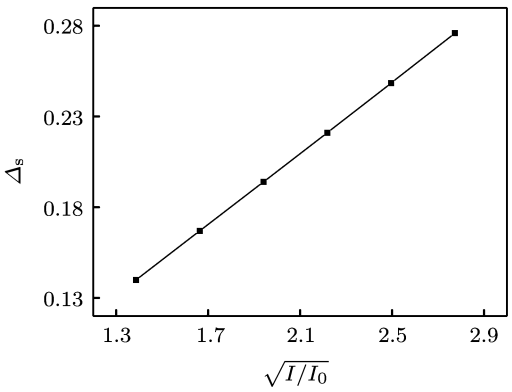


Fig. 3. The amplitude of AT splitting varying with the square root of pump laser intensity I at fixed wavelength $\lambda_{\text{pump}} = 640$ nm. The solid squares denote our exact numerical results and the solid line refers to the results of Eq. (14). The other parameters of pump and probe laser fields are the same as those in Fig. 2.

We now consider the case that K_2 molecule is excited by the non-resonant (near resonant) pump pulse, or $\omega_{\text{pump}} \neq \omega_0$. Figure 4 demonstrates the photoelectron spectra for the different wavelengths of pump pulse at an intensity of $4I_0$. From the figure, we know that the wavelength of the pump pulse determines not only the amplitude of AT splitting, but also the enhancement of peaks. Asymmetric profiles arise when the pump pulse is slightly detuned from resonance and the splitting peaks shift about $\frac{1}{2}\Delta_1$ compared with that of resonant pulse $\lambda_{\text{pump}} = 640$ nm. At $\lambda_{\text{pump}} = 620$ nm, the right peak is enhanced and moves toward higher energy, which corresponds to $|\alpha\rangle$ substate. However, at $\lambda_{\text{pump}} = 660$ nm the left peak is enhanced and moves toward lower energy, which corresponds to $|\beta\rangle$ substate. Physically, because of the influence of laser field, the "bare" state $|B\rangle$ with pump pulse produces "dressed states" $|\alpha\rangle$ and $|\beta\rangle$. In this case, the energies of $|\alpha\rangle$ and $|\beta\rangle$ states and the transitions between these two states depend on the wavelength of pump pulse. The peaks of photoelectron spectra reflect the energies and transitions of them. In the case of $\lambda_{\text{pump}} = 640$ nm, the dressed states $|\alpha\rangle$ and $|\beta\rangle$ will be equally excited because of their symmetries, so the two symmetric peaks are obtained (see the middle panel of Fig. 4). However, if

the wavelength of pump pulse scans from 620 nm to 660 nm, the dressed states $|\alpha\rangle$ and $|\beta\rangle$ will be shifted and excited differently, the two asymmetric peaks are obtained at near resonant pump pulse and the AT splitting peaks shift toward lower energy with the increase of laser wavelength. This process is shown in Fig. 4.

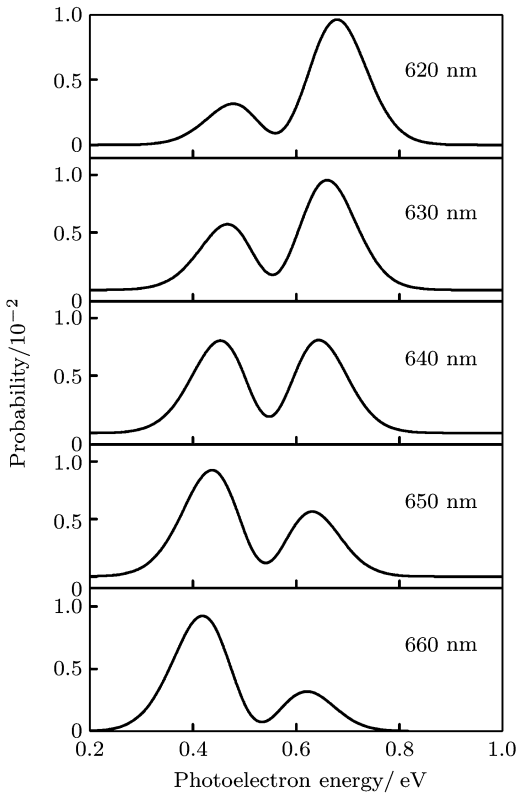


Fig. 4. Variations of AT splitting with the wavelength of pump pulse at fixed intensity $4I_0$. The other parameters of pump and probe laser fields are the same as those in Fig. 2.

Finally, we discuss the case that K_2 molecule is excited by the far-off resonant pump pulse, namely $|\omega_{\rm pump} - \omega_0| > \Omega_{\rm pump}$. In this case, the AT splitting disappears and only a single peak appears in the photoelectron spectrum of K_2 molecule. According to dressed state theory, we can deduce that the population on the excited state is distributed only on one substate: $|\alpha\rangle$ or $|\beta\rangle$. Our calculation shows that the population on the $|\alpha\rangle$ substate is zero for $\lambda_{\rm pump} \geq 720$ nm in this work. In other words, the excited state $|B\rangle$ is shifted under the strong far-off resonant laser field. The shift of $|B\rangle$ state can be interpreted as ac-Stark effect.[46,56] If $\omega_{\rm pump} > \omega_0$, ac-Stark effect leads to upshift of $|B\rangle$ state. The reverse situation happens when $\omega_{\rm pump} < \omega_0$. The extent of shift depends linearly on pump laser intensity I. Figure 5 shows the photoelectron spectrum as a function of the pump laser intensity I and photoelectron energy for the fixed wavelength $\lambda_{\rm pump} = 740$ nm. The asterisks in Fig. 5 denote our exact numerical results and they represent the energies corresponding to the maximum probabilities of photoelectron spectrum at intensity I. The black line refers to the result of linear fitness. We can see that the expected linear behaviours are shown to be unlike the case of resonant excitation $\omega_{\rm pump} = \omega_0$ which is proportional to $\sqrt{I}$.

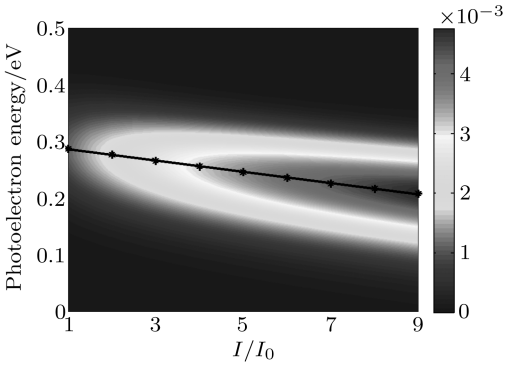


Fig. 5. Photoelectron spectrum of K_2 molecule as a function of the pump laser intensity I and photoelectron energy for the fixed wavelength $\lambda_{\rm pump} = 740$ nm. The other parameters of pump and probe laser fields are the same as those in Fig. 2.

4. Conclusion and outlook

In this paper, we theoretically present the nature of AT splitting in K_2 molecular system by employing the time-dependent wave packet approach. With the ultrashort and intense pump laser field, the populations on each electronic state show Rabi oscillations during the excitation process. For the resonant pump pulse, a symmetric profile is shown and the amplitude of AT splitting is determined by the pump laser intensity. For the near resonant pump pulse, the asymmetric profiles are shown, the laser wavelength determines the positions of AT peaks and the enhancement of the peaks. When the pump pulse is far-off resonance, the AT splitting vanishes and only a single peak appears in the photoelectron spectrum of K_2 molecule. The shift of the single peak is proportional to pump laser intensity I, unlike the AT splitting which is proportional to $\sqrt{I}$ in the case of resonant excitation $\omega_{\rm pump} = \omega_0$.

Since the photoelectron spectra of AT splitting map the information about dressed states, we are able to manipulate the energies and populations of dressed states with the laser intensity and wavelength. This could be used to control quantum dynamics and chemical reaction. In the view of applications to quan-

tum control, selectivity and efficient population transfer to the specific target states are of primary importance in physics, biology, and chemistry. These can be achieved by tuning the dressed state energy into resonance with the target state. The laser wavelength can be used to manipulate the populations on the dressed states whereas the intensity of the strong pulse is used to determine the dressed states splitting. This dynamic information can be obtained accurately by the observation of the photoelectron spectrum. With the help of a strong ultrashort laser pulse in the femtosecond domain, the course of reaction can be controlled into distinctive reaction channels by changing the dressed potential surfaces. Even faster switching among the different final states is achieved by the control of the electronic coherence via selective population of dressed states. The strong non-resonant laser field introducing ac-Stark effect can be applied to the control of molecular dynamics via the time-dependent modification of an effective Hamiltonian. The insights into the physical mechanism of dressed states permit us to design specific strong field pulse in order to maximize the laser control efficiency, a larger molecular system with more electronic states is currently studied in our group to realize the laser control.

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