# ARTICLE Influence of Femtosecond Laser Parameters on Autler-Townes Splitting of Three-Level Ladder Molecules

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We theoretically investigate the Autler-Townes (AT) splitting in the photoelectron spectra of three-level ladder  $K_2$  molecule driven by a pump-probe pulse via employing the timedependent wave packet approach. The dependence of AT splitting on two laser intensities and wavelengths are studied in detail. We firstly quantify these effects on peak shift and AT separation. The photoelectron spectra show double splitting with symmetric profiles, but with asymmetric profiles when the wavelength is changed. The magnitude of AT splitting increases with the pump laser intensity, but does not vary with probe intensity. The shifts of the absorption peaks and the splitting between AT doublet are predicted by using an analytical fitting function when the intensity/wavelength of one of the two fields is changed. These novel results are of importance for the molecular spectroscopy and may further stimulate the first principles theoretical studies analytically.

Key words: Autler-Townes splitting, Photoelectron spectra, Three-level ladder K<sub>2</sub> molecule

#### I. INTRODUCTION

Autler-Townes (AT) splitting occurs due to nonlinear interactions between light and matter driven by one or more strong variable radiation fields [1]. AT splitting is found to be sensitive to laser intensity [2-18], laser wavelength [2-6, 10, 14-17], pulse width [11], pulse envelope [13, 17, 18], and time delay [7-9, 14]. It has been studied theoretically and experimentally in multi-level atomic/molecular system of various configurations.

The three-level system interacting with two laser fields is the most extensively studied subject in quantum optics and laser spectroscopy. The AT double splitting can generally be observed by using a pumpprobe laser configuration, and can be interpreted by the dressed-state theory. Du *et al.* [2] and Wilson *et* al. [3] presented the dependence of the AT effect on either coupling field detuning or microwave field detuning in electromagnetically induced transparent (EIT) resonance in  $\Lambda$ -type atoms/molecules. Zhu *et al.* suggested pump wavelength affected the positions and splitting of the AT doublet of spontaneous emission spectrum for the V-type/ladder atoms [4]. Peng and Zheng investigated the coherent optical spectroscopy for single quantum dots under the pump-probe driving in V-type system, showed that the pump intensity and detuning

affected the splitting, position and relative height of AT double peaks [5]. Papademetriou *et al.* suggested that the AT absorption spectrum of ladder Na became asymmetric, the position and splitting varied with the pump detuning [6]. Wollenhaupt et al. observed experimentally the AT double splitting in photoelectron spectra of ladder K, and studied the effect of pump intensity and time delay on AT doublet [7-9]. They suggested that the asymmetry of the splitting was due to the nonresonant excitation, and the splitting monotonically increased with the laser intensity. Chang et al. showed the evolution of the EIT doublet with pump wavelength in ladder Cs [10]. They proposed that increasing pump detuning enlarged the spacing of the EIT doublet, vary the light shift of the center position, and altered the relative position of the doublet. Sun and Lou studied theoretically the effect of pump intensity and pulse width on the AT double splitting in ladder Na<sub>2</sub>. The longer pulse can induce the asymmetry of doublet [11]. Yuan et al. studied the effects of molecular rotation and alignment on the AT double splitting in ladder  $Na_2$  [12, 13]. Liu et al. indicated probe wavelength and time delay had effect on the AT double splitting and relative peak height in ladder Li<sub>2</sub> [14]. Yao et al. observed the AT double splitting in ladder K<sub>2</sub>, and suggested the pump intensity and wavelength determined the magnitude of splitting and the enhancement of peaks, but they did not quantify the effect [15, 16].

Most experimental and theoretical studies on the dependence of AT splitting on laser intensity and wave-

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leng thare with the three-level V,  $\Lambda$  and ladder atomic configuration. Few works are with the ladder K<sub>2</sub> molecular configuration. In this work, we present new data on intensity/wavelength dependence of the AT splitting in the photoelectron spectra of three-level ladder K<sub>2</sub> via the time-dependent quantum wave packet method. The corresponding features are different and interesting for three cases: resonant pulse, near resonant pulse, and far-off resonant pulse. We firstly quantify these effects on peak shift and AT separation.

## **II. THEORY**

We model the three-level ladder K<sub>2</sub> molecule including three electronic states, ground state  $|X\rangle (|X\rangle^{1}\Sigma_{\rm g}^{+})$ , excited state  $|B\rangle (|B\rangle^{1}\Sigma_{\rm g}^{+})$ , and ionic ground state  $|X^{+}\rangle$  $(|X^{+}\rangle^{2}\Sigma_{\rm g}^{+})$  [15, 16, 19]. We ignore the rotational degree of freedom in the study. The potential energy curves are shown in Fig.1, which are taken form Refs.[20-22]. We only consider the case in which two pulses only couple the states indicated in Fig.1. The transition from  $|X\rangle$ to  $|B\rangle$  is excited by resonant one-photon absorption, and then the populations in  $|B\rangle$  are ionized through single-photon absorption.

In the Born-Oppenheimer frame, the wavefunctions  $\Psi$  can be obtained by solving the time-dependent Schrödinger equation

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

The total molecular-field Hamiltonian H can be written as

$$H = H_S + H' \tag{2}$$

$$= T + V + H^{2} \tag{2}$$

$$H_S = T + V \tag{3}$$

where  $H_S$  is the Hamiltonian of the K<sub>2</sub>, T is the operator of the kinetic energy of nuclei, V is the potential energy of the system, and H' is the interaction between K<sub>2</sub> and laser field. For the three-state model, the wave functions can be written in the column vector

$$\Psi = (\psi_X, \psi_B, \psi_{\text{ion}})^T \tag{4}$$

where  $\psi_X$ ,  $\psi_B$ , and  $\psi_{\text{ion}}$  are the wave functions for  $|X\rangle$ ,  $|B\rangle$ , and  $|X^+\rangle$ , respectively.  $|X^+\rangle$  is a continuum state and is discretized into a band of quasicontinuum states. The  $\psi_{\text{ion}}$  can be further expressed as

$$\psi_{\text{ion}} = (\psi^{(1)}, \psi^{(2)}, ..., \psi^{(N)})^T \tag{5}$$

where N is the number of discrete states of  $K_2$  ion.

The kinetic energy T can be expressed as

$$T = -\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} \begin{pmatrix} 1 & 0 & \vartheta \\ 0 & 1 & \vartheta \\ \tilde{\vartheta} & \tilde{\vartheta} & \text{II} \end{pmatrix}$$
(6)

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FIG. 1 Potential energy curves of  $K_2$  molecule used in the work. The arrows indicate the excitation energy of 1.937 eV (640 nm) and 2.695 eV (460 nm), respectively.

where  $\mu$  is the reduced mass of the nuclei, R is the internuclear distance,  $\vartheta = (0, 0, \ldots, 0)$  is a zero vector with N-component,  $\tilde{\vartheta}$  is the transverse of the vector  $\vartheta$  and II is a  $N \times N$  unit matrix and is used to describe the ionization state.

The potential V can be expressed as

$$V = \begin{pmatrix} V_X & 0 & \vartheta \\ 0 & V_B & \vartheta \\ \tilde{\vartheta} & \tilde{\vartheta} & (V_{\rm ion} + \xi^{(i)}) \Pi \end{pmatrix}$$
(7)

where  $V_X$ ,  $V_B$ , and  $V_{\text{ion}}$  are the potential curves of  $|X\rangle$ ,  $|B\rangle$ , and  $|X^+\rangle$ , respectively.  $\xi^{(i)}=(i-1)\Delta\xi$  $(i=1,2,\ldots,N)$  is the energy of the emitted photoelectron.

The interaction H' between  $K_2$  molecule and laser field is given by

$$H' = \begin{pmatrix} 0 & W_{XB} & 0\\ W_{XB} & 0 & \varpi_{\mathrm{Bi}}\\ 0 & \varpi_{\mathrm{Bi}} & \varrho \end{pmatrix}$$
(8)

where  $\varpi = (W_{2i}^{(1)}, W_{2i}^{(2)}, \dots, W_{2i}^{(N)})$  is the vector of coupling between  $|B\rangle$  and  $|X^+\rangle$  via laser field with *N*-component, and  $\varrho$  is the  $N \times N$  zero matrix. The coupling between two states via external laser field is given as follows

$$W_{XB} = \hbar R_1(R) \cos(\omega_1 t) \tag{9}$$

$$\varpi_{\rm Bi} = \hbar R_2(R) \cos(\omega_2 t) \tag{10}$$

$$R_1 = \frac{1}{\hbar} \mu_{XB}(R) e_1 f(t) \tag{11}$$

$$R_2 = \frac{1}{\hbar} \mu_{\rm Bi}(R) e_2 f(t) \tag{12}$$

$$f(t) = \exp\left[-4\ln 2\left(\frac{t}{\tau}\right)^2\right]$$
(13)

where  $R_1$  and  $R_2$  are the Rabi frequencies of laser fields for  $|X\rangle \rightarrow |B\rangle$  and  $|B\rangle \rightarrow |X^+\rangle$ , respectively.  $\mu_{XB}(R)$  and  $\mu_{\text{Bi}}(R)$  are the transition dipole moments,  $e_1$  and  $e_2$  are

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the amplitudes of laser fields,  $\omega_1$  and  $\omega_2$  are angular frequencies, f(t) is the Gaussian pulse envelope,  $\tau$  is the full width at half maximum (FWHM) of pulse, and it is 30 fs in our calculations.

The energy-resolved photoelectron spectrum is defined as [11-18]:

$$P(\xi^{(i)}) = \lim_{t \to \infty} \int \mathrm{d}R |\psi^{(i)}(R, t, \xi^{(i)})|^2$$
(14)

The transition dipole moments are taken from Refs.[22, 23]. In this work,  $\xi^{(i)}$  span over 0–2 eV and N=120. The time-dependent Schrodinger equation (Eq.(1)) is solved by "split-operator Fourier" methods exactly [25–28].

#### **III. RESULTS AND DISCUSSION**

The effect of pump laser intensity on AT splitting is considered in resonant region. Figure 2(a) shows the photoelectron spectra as a function of pump laser intensity  $I_1$  at  $I_2=4I_0$  ( $I_0=1.0\times10^{11}$  W/cm<sup>2</sup>). The peaks of photoelectron spectra show the double splitting with symmetric profiles at  $I_1 \ge 2I_0$ , while there is no splitting at  $I_1 \leq 2I_0$ . The doublet with symmetric profiles has been observed in three-level ladder  $Na_2$  [11, 12],  $Li_2$  [14], and  $K_2$  molecules [15, 16]. This is similar to the expected two-peak emission/absorption (dynamic splitting) in four-level atomic system with various configurations interacting with three driving fields [28–32]. The splitting pattern of photoelectron spectrum is socalled AT splitting. In terms of the explanation of ac-Stark splitting in dressed state picture, the two-peak structure arises from the sufficient Rabi oscillation in the resonant region in the ionization process [11-16]. From the dressed state theory, the excited state  $|B\rangle$  is dressed by the laser field and splits into two substrates  $|\alpha\rangle$  and  $|\beta\rangle$ , which correspond to the two peaks from low to high in photoelectron spectrum respectively. Here, we assume that the energy of  $|\beta\rangle$  state is higher than that of  $|\alpha\rangle$  state. Another important feature in Fig.2(a) is that the magnitude of the double splitting increases with the increase of pump laser intensities. This is similar to the observations in three-level K [7-9], Rb [2], K<sub>2</sub> [15, 16], and Na<sub>2</sub> [11, 12]. A similar feature is shown in the absorption spectrum for four-level atoms/molecules interacting with three driving fields [17, 18, 28–32].

Figure 2(b) shows the peak positions as a function of pump Rabi frequency  $R_1$ . The two peaks are symmetric about 0.484 eV, which simply is  $E_{v0} + \sum \hbar \omega_k - V_I(R_0)$ [11], with  $E_{v0}$  being the energy of vibrational ground state,  $\hbar \omega_k$  is the photon energy, and  $V_I(R_0)$  is the potential of  $|X^+\rangle$  state at equilibrium distance  $R_0$  of the neutral ground state, as indicated with arrows in Fig.1. The positions of two peaks are at 0.484- $R_1/2$ and 0.484+ $R_1/2$  and these are plotted as solid lines in Fig.2(b), which are in good agreement with the numerical results. From Fig.2(a), it is noted that as the pump



FIG. 2 (a) The photoelectron spectra for various pump laser intensities  $I_1$ . (b) Peak positions vs. pump Rabi frequency  $R_1$ . Also shown is the result using the dressedstate formalism (solid lines). The parameters are as follows:  $I_0=1.0\times10^{11}$  W/cm<sup>2</sup>,  $I_2=4I_0$ ,  $\lambda_1=640$  nm,  $\lambda_2=460$  nm,  $\tau=30$  fs.

laser intensity increases, more higher-order peaks begin emerging. The spectrum consists of pairs of peaks. The emergence of more peaks with increasing laser intensity has been observed previously in three-level  $\Lambda$ -type and ladder atoms [6, 33–35] in on-resonant and outresonant case. In the work, we focus on the main two peaks in the photoelectron spectra.

The effect of the probe intensity on AT splitting is considered. Figure 3 shows the photoelectron spectra for several probe laser intensities  $I_2$  at  $I_2=4I_0$ . The photoelectron spectra show a double splitting with symmetric profiles. The probe intensity affects only the peaks heights, this is similar to the observation in three/fourlevel system [5, 32].

The effect of the pump wavelength on AT splitting is considered. Figure 4(a) demonstrates the photoelectron spectra for different pump wavelengths (540–790 nm) while keeping the probe wavelength consistent. The pump field detuning is given by  $\Delta_1 = \omega_1 - \omega_{BX}$ , where  $\omega_{BX}$  is the frequency of  $|X\rangle \rightarrow |B\rangle$ . From Fig.4(a), it is noted that the spectrum exhibits a double splitting with symmetric profiles when the pump field has a zero detuning, while asymmetric profiles arise as the pump is moved off resonance wavelength. The asym-



FIG. 3 The photoelectron spectra for various probe intensities of  $0.06I_0$ ,  $2I_0$ ,  $4I_0$ ,  $5I_0$ ,  $8I_0$ ,  $10I_0$ ,  $15I_0$ ,  $20I_0$ . The parameters are as follows:  $I_2=4I_0$ ,  $\lambda_1=640$  nm,  $\lambda_2=460$  nm,  $\tau=30$  fs.

metry of the splitting is due to the nonresonant excitation. This asymmetry has been observed in double splitting in three-level K atoms [7-9] and K<sub>2</sub> molecules [15, 16] and other atoms [4-6, 10]. The double peaks shift toward the lower energy with increasing pump wavelengths. This is because the longer wavelength indicates the lower photon energy, resulting in lower photoelectron energy. This energy shift trend with laser wavelength is qualitatively identical with the observation in three/four-level atoms/molecules [3-5, 7, 7]17, 28].This is also identical with the observation in three-level  $K_2$  molecule [15, 16]. They did not quantify the magnitude of peak shift and the split-Figure 4(b) shows the peak poting of doublet. pump detuning  $\Delta_1$ . The positions of sitions vs. the two peaks are at  $0.484 + (1/2)\Delta_1 - (1/2)\sqrt{\Delta_1^2 + R_1^2}$ and  $0.484 + (1/2)\Delta_1 + (1/2)\sqrt{\Delta_1^2 + R_1^2}$ , respectively, and these are plotted as solid lines in Fig.4(b). The splitting of the AT doublet is  $\sqrt{\Delta_1^2 + R_1^2}$ . This magnitude of peaks shift is not shown in earlier results in three-level ladder  $K_2$  system, but in other configurations [2-5, 36, 37].

In Fig.4, an important and interesting feature is that the double structure is transformed into one peak progressively as the pump is detuned from the resonance wavelength. The threshold points are  $\Delta_1 = \pm 0.19 \text{ eV}$ (*i.e.*  $\lambda_1 = 580$  or 710 nm) for two-to-one transformation. The peaks disappearance can be explained in terms of the asymmetric excitation/population of dressed states [5, 15, 16]. For the resonant case, the dressed states  $|\alpha\rangle$ and  $|\beta\rangle$  will be equally excited because of their symmetries, so the two symmetric peaks are observed. However, if the pump wavelength scans from 590 nm to 700 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. For example, as the pump wavelength increases from resonance, the longer wavelength indicates the lower photon energy, the dress state  $|\alpha\rangle$  will be populated more eas-



FIG. 4 (a) The photoelectron spectra for various pump laser wavelengths  $\lambda_1$  (540–790 nm). (b) Peak positions vs. pump detuning  $\Delta_1$ . The parameters are as follows:  $I_1=I_2=4I_0$ ,  $\lambda_2=460$  nm,  $\tau=30$  fs.

ily than substate  $|\beta\rangle$ , resulting in the enhancement of left peak over right peak. The disappearance of the right/left peak for  $\lambda_1 \ge 710 \text{ nm}/\lambda_1 \le 580 \text{ nm}$  indicates that the population on the  $|\beta\rangle/|\alpha\rangle$  substate is zero.

The effect of the probe wavelength on AT splitting is considered. Figure 5(a) demonstrates the photoelectron spectra for different probe wavelengths (400-520 nm) while keeping the pump resonant. The probe field detuning is given by  $\Delta_2 = \omega_2 - \omega_{460}$ . The probe wavelength affects only the peak positions. This is different from the suggestion that the probe wavelength has effects on the separation and peak height of the doublet in threelevel ladder Li<sub>2</sub> molecule [29]. Figure 5(b) shows the peak positions vs. probe detuning  $\Delta_2$ . The linear fits are  $0.391+0.997\Delta_2$  and  $0.578+0.997\Delta_2$  for two peaks respectively. This indicates both peaks shift  $\Delta_2$ . The splitting (0.198 eV) between the two peaks do not vary with probe wavelength.

Considering the effects of pump/probe intensity and wavelength on the AT splitting discussed, two peaks are at  $0.484+(1/2)\Delta_1+\Delta_2-(1/2)\sqrt{\Delta_1^2+R_1^2}$  and  $0.484+(1/2)\Delta_1+\Delta_2+(1/2)\sqrt{\Delta_1^2+R_1^2}$  from low to high in the photoelectron spectrum, which correspond to two substates  $|\alpha\rangle$  and  $|\beta\rangle$  respectively. The splitting of the AT doublet is  $\sqrt{\Delta_1^2+R_1^2}$ .

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FIG. 5 (a) The photoelectron spectra for various probe laser wavelengths  $\lambda_2$  (400-520 nm). (b) Peak positions vs. probe detuning  $\Delta_2$ . The parameters are as follows:  $I_1=I_2=4I_0$ ,  $\lambda_1=640$  nm,  $\tau=30$  fs.

### **IV. CONCLUSION**

We investigated the dependence of AT splitting on pump/probe laser intensity and wavelength in the photoelectron spectra of the three-level ladder  $K_2$  molecule by the time-dependent wave packet approach. The corresponding features are discussed for: resonant pulse, near resonant pulse, and far-off resonant pulse.

In pump resonant region, for fixed intensity of probe field, the photoelectron spectra show the double splitting with symmetric profiles (in peak height and splitting of two peaks), and the AT splitting increases with pump intensity. The two peaks approach a linear splitting proportional to  $R_1/2$ . The probe intensity affects only the peaks heights.

In the pump nonresonant region, for the fixed intensities of two fields, the photoelectron spectra show the double splitting with asymmetric profiles (in peak height), the splitting  $(\sqrt{\Delta_1^2 + R_1^2})$  between the doublet varies with pump wavelength. The two peaks shift to lower energy with different shift as pump wavelength increases. As the pump moves off resonance wavelength, the double structure is transformed into one peak progressively in near resonant region (*i.e.*) Yue-hua Li et al.

580 nm $\leq \lambda \leq$ 710 nm). This can be explained in terms of the asymmetric excitation/population of dressed states.  $\Delta_1=0.19 \text{ eV}$  (*i.e.*  $\lambda_2=580 \text{ or } 710 \text{ nm}$ ) is the threshold point of near resonant region and far-off resonant region.

The probe wavelength affects only the peak positions. Both peaks shift to lower energy with the same shift  $\Delta_2$ . The splitting (0.198 eV) between the two sideband peaks in doublet do not change with probe wavelength.

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