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 pump-probe 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 transparency (EIT) resonance in A-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 non-resonant excitation, and the splitting monotonically increased with the laser intensity. Chang et al. observed 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 Na2. 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 Na2 [12, 13]. Lin et al. indicated probe wavelength and time delay had effect on the AT double splitting and relative peak height in ladder Li2 [14]. Yao et al. observed the AT double splitting in ladder K2, 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-
leng there with the three-level V, A and ladder atomic configuration. Few works are with the ladder K2 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 K2 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 K2 molecule including three electronic states, ground state |X⟩ (|X⟩|Σg⟩), excited state |B⟩ (|B⟩|Σg⟩), and ionic ground state |X+⟩ (|X+⟩|Σg⟩) [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⟩ to |B⟩ is excited by resonant one-photon absorption, and then the populations in |B⟩ are ionized through single-photon absorption.

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

\[ i\hbar \frac{\partial}{\partial t} \Psi = H \Psi \]  

The total molecular-field Hamiltonian H can be written as

\[ H = H_S + H' \]
\[ = T + V + H' \]  

\[ H_S = T + V \]

(2)  

(3)

where \( H_S \) is the Hamiltonian of the K2, T is the operator of the kinetic energy of nuclei, V is the potential energy of the system, and \( H' \) is the interaction between K2 and laser field. For the three-state model, the wave functions can be written in the column vector

\[ \Psi = (\psi_X, \psi_B, \psi_{\text{ion}})^T \]  

(4)

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

\[ \psi_{\text{ion}} = (\psi^{(1)}, \psi^{(2)}, ... , \psi^{(N)})^T \]  

(5)

where N is the number of discrete states of K2 ion.

The kinetic energy T can be expressed as

\[ T = -\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} \begin{pmatrix} 1 & 0 & \vartheta \\ 0 & 1 & \vartheta \\ \vartheta & \vartheta & \Pi \end{pmatrix} \]  

(6)

FIG. 1 Potential energy curves of K2 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, \( \vartheta = (0, 0, 0, \ldots, 0) \) is a zero vector with N-component, \( \vartheta \) is the transverse of the vector \( \vartheta \) and \( \Pi \) is a \( N \times N \) unit matrix and is used to describe the ionization state.

The potential V can be expressed as

\[ V = \begin{pmatrix} V_X & 0 & \vartheta \\ 0 & V_B & \vartheta \\ \vartheta & \vartheta & (V_{\text{ion}} + \xi^{(i)})\Pi \end{pmatrix} \]  

(7)

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

The interaction \( H' \) between K2 molecule and laser field is given by

\[ H' = \begin{pmatrix} 0 & W_{XB} & 0 \\ W_{XB} & 0 & \varpi_{Bi} \\ 0 & \varpi_{Bi} & \varrho \end{pmatrix} \]  

(8)

where \( \varpi=(W^{(1)}, W^{(2)}, \ldots, W^{(N)}) \) is the vector of coupling between \(|B⟩\) and \(|X^+⟩\) 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

\[ W_{XB} = \hbar R_1(R) \cos(\omega_1 t) \]  

(9)

\[ \varpi_{Bi} = \hbar R_2(R) \cos(\omega_2 t) \]  

(10)

\[ R_1 = \frac{1}{\hbar} \mu_{XB}(R) e_1 f(t) \]  

(11)

\[ R_2 = \frac{1}{\hbar} \mu_{Bi}(R) e_2 f(t) \]  

(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 \(|X⟩\rightarrow|B⟩\) and \(|B⟩\rightarrow|X^+⟩\), respectively. \( \mu_{XB}(R) \) and \( \mu_{Bi}(R) \) are the transition dipole moments, \( e_1 \) and \( e_2 \) are...
the amplitudes of laser fields, ω₁ and ω₂ are angular frequencies, f(t) is the Gaussian pulse envelope, τ 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]:

\[ P(\xi^{(i)}) = \lim_{t \to \infty} \int dR |\psi^{(i)}(R, t, \xi^{(i)})|^2 \]  (14)

The transition dipole moments are taken from Refs. [22, 23]. In this work, ξ^{(i)} span over 0–2 eV and N=120. The time-dependent Schrödinger 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 I₁ at I₂=I₉ (I₀=1.0×10¹¹ W/cm²). The peaks of photoelectron spectra show the double splitting with symmetric profiles at I₁≥2I₀, while there is no splitting at I₁≤2I₀. The doublet with symmetric profiles has been observed in three-level ladder Na₂ [11, 12], Li₂ [14], and K₂ 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 so-called 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⟩ is dressed by the laser field and splits into two substrates |α⟩ and |β⟩, which correspond to the two peaks from low to high in photoelectron spectrum respectively. Here, we assume that the energy of |β⟩ state is higher than that of |α⟩ 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], K₂ [15, 16], and Na₂ [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 R₁. The two peaks are symmetric about 0.484 eV, which simply is Eᵥ₀+∑ kωk−Vᵣ(R₀) [11], with Eᵥ₀ being the energy of vibrational ground state, hωk is the photon energy, and Vᵣ(R₀) is the potential of |X⁺⟩ state at equilibrium distance R₀ of the neutral ground state, as indicated with arrows in Fig.1. The positions of two peaks are at 0.484−R₁/2 and 0.484+R₁/2 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 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 Λ-type and ladder atoms [6, 33–35] in on-resonant and off-resonant 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 I₉ at I₂=4I₀. 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/four-level 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 Δ₁=Ω₁−ωₓₓ, where ωₓₓ is the frequency of |X⟩→|B⟩. 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-
The two peaks are at $0.484 + (1/2)\Delta_1$ positions of the doublet. Figure 4(b) shows the peak positions quantifying the magnitude of peak shift and the splitting of double. In Fig.4, an important and interesting feature is that in the photoelectron spectrum, which correspond to the AT doublet is two substates $|\alpha\rangle$ and $|\beta\rangle$. They did not quantify the magnitude of peak shift and the splitting of doublet. Figure 4(b) shows the peak positions vs. pump detuning $\Delta_1$. The positions of the two peaks are at $0.484 + (1/2)\Delta_1$ and $0.484 + (1/2)\Delta_1$, respectively, and these are plotted as solid lines in Fig.4(b). The splitting of the AT doublet is $\sqrt{\Delta_1^2 + R_2^2}$. This magnitude of peaks shift is not shown in earlier results in three-level ladder Li$_2$ 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 resonant wavelength. The threshold points are $\Delta_1 = 0.19$ 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 easily than substrate $|\beta\rangle$, resulting in the enhancement of left peak over right peak. The disappearance of the right/left peak for $\lambda_1 \geq 710$ nm/$\lambda_1 \leq 580$ 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 three-level ladder Li$_2$ 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 peak shifts $\Delta_2$. 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 $0.484 + (1/2)\Delta_1 + \Delta_2$ and $0.484 + (1/2)\Delta_1 + \Delta_2$ 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 $\sqrt{\Delta_1^2 + R_2^2}$.
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 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. 580 nm$\leq$$\lambda$$\leq$710 nm). This can be explained in terms of the asymmetric excitation/population of dressed states. $\Delta_1=0.19$ eV (i.e. $\lambda_2=580$ or 710 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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