Fine structures of the harmonic and hyper-Raman spectrum of the hydrogen atom in an intense high-frequency laser pulse

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The photon emission spectrum of the hydrogen atom in an intense high-frequency laser pulse is simulated to show that regular fine structures appear in the two sides of both the odd and even order harmonics. The splitting of the fine structures around the normal harmonic line are determined by the energy differences of the eigenstates of the time averaged Kramers-Henneberger potential with the same parity. The lines around the even order harmonic positions are produced by the hyper-Raman transitions between the dressed atomic states with different parities. The hyper-Raman lines are predicted as long as stabilization occurs and are not so sensitive to the initial state of the atom as those appearing with the low-frequency and less strong laser field. The hyper-Raman lines can be strongly reduced with longer turn-on and turn-off times.

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I. INTRODUCTION

The interaction of an intense laser field with atoms has been an active subject for many years for the nonlinear and nonperturbative phenomena such as the high-order harmonic generation (HHG) [1–3], stabilization [4–6], etc. With the advances of high power free electron lasers (FEL) [7], light sources will soon be available to generate photons whose energy $\hbar\omega_L$ may equal or even exceed the binding energies of the ground state of the atom. In such high-frequency laser fields, atoms may be more stable against ionization, i.e., stabilization occurs and the ionization probability may decrease rather than increase with the rise of laser intensity. Stabilization was first described with the time-independent high-frequency Floquet theory in the Kramers-Henneberger (KH) scheme [5], in which the influence of the external field is taken into account via an effective potential approach with consideration for the rapid oscillations of the external field. By averaging over these oscillations in one optical cycle, this average potential with high laser frequency ω_L is given by [8–10]

$$V_{eff} = \frac{\omega_L}{2\pi}\int_0^{2\pi/\omega_L} dt V[\mathbf{r} + \alpha(t)], \tag{1}$$

where $V(\mathbf{r})$ is the field-free atomic potential. The classical electron displacement in the incident electric field is given by $\alpha(t)$. $\alpha_0 = |\mathbf{E_L}|/\omega_L^2$ denotes the maximum classical displacement of the electron in the incident field. Then V_{eff} has two attraction centers, at $\pm\mathbf{E_L}/\omega_L^2$, for the linearly polarized laser field. For the circularly polarized laser field, V_{eff} is like a circular trough of radius α_0 [6]. KH potential is often used to explain the shape of the wave packet in the intense high-frequency laser field [5,6]. In the present study, it is used to interpret the fine structures in the harmonic spectrum.

HHG, as a possible source of coherent x rays and attosecond extreme ultraviolet (XUV) light, is also an attractive subject. In terms of the three-step model [3,11,12], it is often interpreted that the electron is ionized to the continuum state first by absorbing photons and then comes back again and recombines to the parent ion to emit a photon. The electron tends to come back to the initial state and there are only odd harmonic lines according to the parity conservation. Besides the odd order harmonic lines, it is predicted that there are also hyper-Raman lines [13–16] in the photon emission spectrum. These lines are caused, differently from the harmonic ones, by the transitions between the dressed atomic bound states. They are positioned around the even or odd harmonics depending on the number of the photons absorbed. If the energy difference between the dressed atomic bound states is ω_i, the hyper-Raman lines are then located at frequencies of $\omega_{hR} = \omega_i \pm n\omega_L$ [16]. The hyper-Raman lines have already been predicted theoretically, however, they have never been observed in the photon emission spectrum in an actual experiment. Several explanations have been given for its absence. One of the reasons is that hyper-Raman emissions cannot add coherently because of the lack of phase matching in the forward direction in a microscopic sample. It was also argued that the hyper-Raman lines can only be seen in the central laser pulse [15] and that they are emitted preferentially in a direction orthogonal to the incident laser field and easily escape. It was suggested in Ref. [16] that the production of hyper-Raman lines is restricted: first, the system must be in a linear combination of Floquet states, second, even if the linear combination is produced, the higher lying states should not decay too quickly such that these lines are stable enough to be detected. Many methods has been suggested to satisfy these requirements. For example, the first condition is realized by preparing the initial states as the coherent superposition state or choosing the laser frequency corresponding to the energy difference between two bound states and so on, the second is realized with a not so strong laser field but with a suitable turn-on time and an appropriate constant part. Most of these investigations were made by using a one-dimensional (1D) model potential, including the two-level system and soft Coulomb potential, with only the lines at frequencies $\omega_{hR} = \omega_i \pm 2n\omega_L$ being seen clearly. While the conditions can be more easily realized when stabilization occurs. The bound states are different in the KH potential for

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the intense high-frequency laser field from those in the field-free potential. The linear combination can be realized in the interim of the potential without artificial management and the high lying states will be stable in view of the definition of stabilization. There are not so many requirements for the initial state and the laser pulse. The transitions between bound states is complex and we can see the clear hyper-Raman lines around all orders of harmonics $\omega_{hR}=\omega_i \pm n\omega_L$ when stabilization occurs and the KH potential operates.

II. THEORETICAL METHOD

We make the three-dimensional calculations for the true hydrogen atom. The time-dependent Schrödinger equation that we solve, in atomic units (a.u.), is of the form

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t)=\left(-\frac{1}{2}\nabla^2+\frac{l(l+1)}{2r^2}-\frac{1}{r}+\hat{H}_I(\mathbf{r},t)\right)\psi(\mathbf{r},t), \quad (2)$$

where ψ is the reduced wave function and $\psi=r\phi$. $\hat{H}_I$ is the interaction parts given by

$$\hat{H}_I=-E_L f(t)z\,\sin(\omega_L t), \quad (3)$$

when we only consider the field polarized along the **z** direction. $f(t)$ is the envelope of the laser pulse chosen to have $\sin^2$ for turn-on and $\cos^2$ for turn-off processes and a flat top between them. We expand the wave function in Legendre polynomials in the (r,θ) coordinates as

$$\psi(r_i,x_j,t)=\sum_{l=0}^{L} f_l(r_i,t)P_l(x_j), \quad x_j=\cos\,\theta_j, \quad (4)$$

and then the Fourier transformation is made only with the terms of f_l in the split operator method [17,18]. A smooth cosin mask function confined to a moderately narrow band near the boundaries is adopted in order to prevent reflection of the wave function from the edges of the grid and force the electron density to be zero at these edges. Therefore the norm of the wave function is not conserved. We treat the electron density removed by the mask function as ionization and thus the decrease of the norm is considered to be the ionizing probability [5].

The photon emission spectrum can be determined from the Fourier transform of the time-dependent deduced dipole of the system. Because the radius of the hydrogen nucleus is of the order of 10^{-5} a.u. and the equal-spacing grids are used, the mean acceleration expression needs much more grid points near the nucleus in order to ensure very high accuracy for the wave function. So here we use the velocity form in the momentum space,

$$\dot{d}(t)=\langle\psi(\mathbf{p},t)|p_z|\psi(\mathbf{p},t)\rangle, \quad (5)$$

to get the more accurate spectrum.

III. RESULTS AND DISCUSSIONS

We first take the laser pulse of frequency 1 a.u. for half-photon ionization and the conditions necessary for stabilization can be easily satisfied. Figure 1 is the survival probability for different intensity. Here we take a laser pulse with duration of 100 o.c. for the full evolution of the wave packet in which 5 o.c. are taken for both turn-on and turn-off processes. Stabilization for the true hydrogen atom in the intense high-frequency field exists and affects the relative heights of the harmonic and hyper-Raman lines even though it is not as significant as one often predicted for the low-dimensional model systems.

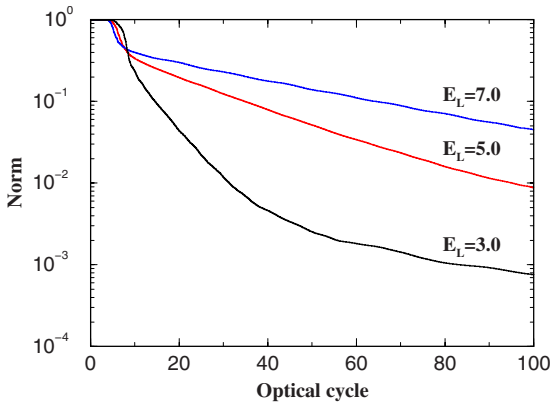

FIG. 1. (Color online) Time-dependent populations of all bound states for hydrogen in a laser pulse of frequency 1 a.u. for E_L =3.0, 5.0, and 7.0 a.u.

The emission spectrum is illustrated in Fig. 2 with a solid line for E_L=5.0 a.u. and a dashed line for E_L=7.0 a.u. One can see the sharp peaks around even and odd times of the photon energy. There are clear and regular splits among the fine peaks. To see the fine structures clearly, the enlarged details around the second and third harmonics are shown in the insets of Fig. 2. For the third one there are peaks symmetrically to the main third harmonic line, while for the second harmonic the central peak does not actually exist, only lines symmetrically appearing in both sides of the central position of the second order harmonic. The same symmetri-

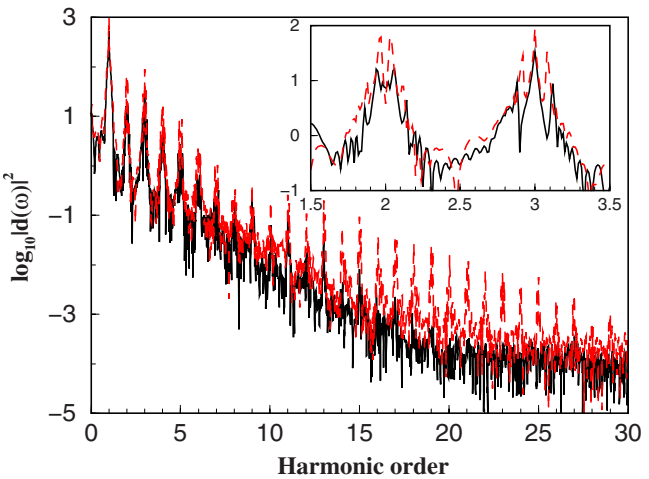

FIG. 2. (Color online) Photon emission spectrum of the hydrogen atom in the laser field of 100 o.c. with 5 o.c. turn-on and turn-off time and frequency 1 a.u., amplitude 5 a.u. (solid line) and 7 a.u. (dashed line). The enlargement of the spectrum around the second and third harmonic order can be seen in the inset.

cal splitting appears for each integer harmonic order at the same relative positions. As mentioned in the Introduction, these lines in the two sides of both the odd and even times of photon energy are hyper-Raman lines.

We can examine the separations between the fine peaks carefully from the insets and find the regularities. First, the relative positions of the fine structures around even and odd harmonic order are different in the same laser field. For the Raman process, the atom first absorbs photons to a virtual energy level from the initial state and immediately transits to another state and emits a photon. If the atom first absorbs even photons, the corresponding hyper-Raman lines should locate at $\omega_{hR}=2n\omega_L \pm \omega_i$, where ω_i is the energy difference between the initial and final states of different parities. If odd photons are absorbed, the hyper-Raman lines will locate at $\omega_{hR}=(2n+1)\omega_L \pm \omega_i$, with ω_i being the energy difference between the initial and final states with the same parity. Second, the detailed splitting peaks with different laser intensity are not located at the same place. This can be interpreted by the changes of the energy eigenvalue of electron with the time averaged KH potential for different α_0. The energy of eigenstates of the KH potential for different α_0 was listed in detail in Ref. [19]. By examining the separations between the fine peaks illustrated in Fig. 2, we can see these two features definitely. For example, the nearest peaks to the third harmonics at 3.12 and 2.88 when $E_L=5.0$ a.u. correspond to the energy difference of 0.12 between the states $1s\sigma_g$ and $2s\sigma_g$. The energy difference of the two bound states is 0.08 for $E_L=7.0$ a.u., corresponding to the peaks locating at 3.08 and 2.92 for the dashed line. Besides, the peaks around the second harmonics at 2.057 and 1.943 when $E_L=5.0$ a.u. correspond to the energy difference of 0.057 between the states $1s\sigma_g$ and $2p\sigma_u$ which have different parity, the other two peaks at 2.145 and 1.855 to the energy difference of states $1s\sigma_g$ and $3p\sigma_u$. The energy differences will decrease by increasing the intensity of the laser field resulting in smaller separations of the fine structure.

In the investigations of the emission spectrum in the infrared laser pulse, the peaks not locating at odd times of photon energy, including intermediate peaks and blueshift or redshift of harmonic peaks, were also seen [20–22]. They were interpreted as the effects of the laser pulse shape. When the peak field amplitude increases during an optical cycle of the laser pulse, the recolliding electron will experience a phase shift relative to the field which produces a blueshift [20,22]. The falling part of the laser pulse corresponds to the redshift of the spectrum. The shift will be more obvious with a faster ramp of the laser pulse. Thus structures between the main peaks can be seen because this effect contributes to a given part of the spectrum repeatedly [21]. So the fine structures can be eliminated by only considering the flat part of the laser field. While in the present case, the major fine structures in the emission spectrum under stabilization conditions can still be seen in our calculations even if we make the Fourier transform only from 5 to 95 o.c., the flat part of the laser pulse as shown in Fig. 3(a). It also indicates that these fines structures are hyper-Raman lines and have a different production mechanism from the harmonics. In Fig. 3(a), there are three sets of data, which are obtained by retaining both the turn-on and turn-off processes, only the turn-on process, and none of the turn-on and turn-off processes, respectively. It can be seen that removing the turn-off process causes negligible changes for the strength of the emission spectrum and that removing both the turn-on and turn-off processes causes considerable changes only for the background between the major structures but not for the strength and the positions of the major peaks.

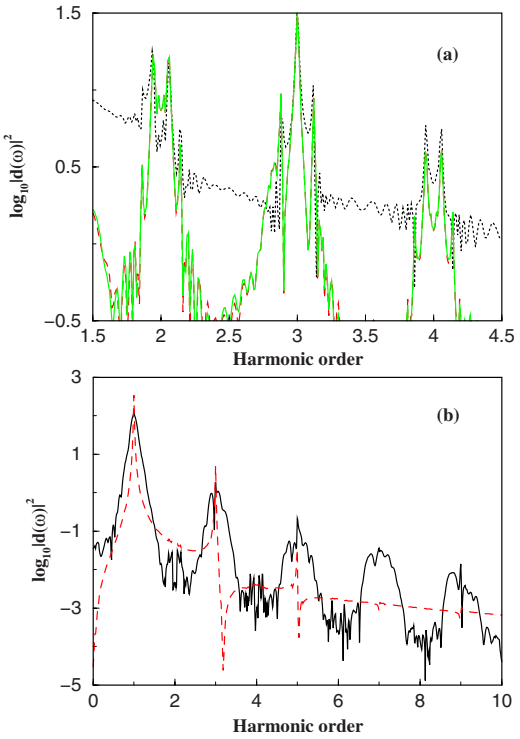

FIG. 3. (Color online) (a) Photon emission spectrum of the hydrogen atom in the laser field of 100 o.c. with 5 o.c. turn-on and turn-off time and frequency 1 a.u., amplitude 5 a.u. Three sets, obtained by retaining both the turn-on and the turn-off processes (solid line), only the turn-on process (dashed line), and none of the processes (doted line), respectively, of the spectrum are presented. (b) The turn-on and turn-off time is increased to be 20 o.c. without changing the whole duration. The spectrum obtained by making Fourier transform from the whole and the flat part of the laser pulse is shown as a solid and dashed line.

Similar results were obtained before by other authors with a laser field of the same frequency and higher intensity [5] except that the regular splits appearing in the present results have been reduced considerably due to the higher laser intensity of $E_L=13.5$ a.u. Therefore the appearance of the even harmonics were interpreted in terms of the nonsymmetric medium which might be caused by the fast ionization process [5]. In Ref. [23], a one-dimensional simulation was carried out to show the influences of the turn-on process. It was seen that the even harmonics strength was lowered with a more slowly turned on pulse. It was interpreted that the fast turn-on means that the evolution process is no longer symmetrical and so a definite parity does no longer exist for the initial and final states; while the splitting of harmonic lines can also be seen in their results even though the author did not pay attention to them. The reduction of the "even" order harmonics with a more slowly turned on pulse can be under-

stood better by considering the characteristics of the hyper-Raman lines. Actually, for the stabilization with high-frequency laser fields, a proper turn-on time is essential to make the population spread across many of the bound eigenstates of the KH potential. The increase of the turn-on time can reduce the spread of the population among the bound states and locate most of the population in the ground state and the transitions between the bound states are restrained significantly. The harmonic intensity changes slightly by increasing the turn-on time, while the hyper-Raman lines are strongly restrained as shown in Fig. 3(b), where the turn-on and turn-off time of the laser pulse is increased to be 20 o.c. with the same total laser duration and $E_L=5.0$ a.u. The present situation is similar to one of the two cases discussed by Pons *et al.* [13] for the influence of the turn-on process on the emission spectrum. Compared to the spectrum in Fig. 3(a), the harmonic lines in Fig. 3(b) are considerably broadened. The broadening is due to the variation of the ground state energy gradually from −0.5 to −0.20196 a.u. during the slow turn-on process of the laser pulse. When the ionized electron from the ground state comes back and recombines with the ion, the energy of the ground dressed state has been enhanced by the laser field resulting in a slight deviation of the emitted photon energy to the left side of the normal position of the odd harmonic lines. The turn-off process causes the opposite shift of the emitted photon. To remove the broadening caused by the turn-on and turn-off processes, narrow harmonic lines can be obtained by taking only the spectrum after the turn-on and before the turn-off of the laser pulse, i.e., the Fourier transform is calculated only from 25 to 75 o.c., the flat part of the laser pulse.

In order to see the dependence of the hyper-Raman process on the frequency and intensity of the laser and find the possible reasons for the absence of the hyper-Raman lines in most existing experiments, we make a few calculations for different laser frequencies with the same $\alpha_0=5.0$ a.u. shown in Fig. 4. As shown in Fig. 4(b), there are only odd harmonics for $\omega_L=0.1$ a.u., which is far below the frequency for the one photon ionization and does not cause stabilization. When stabilization occurs, symmetrical hyper-Raman lines similar to those seen in Fig. 2 can also be found in Fig. 4(c) for $\omega_L=2.0$ a.u. Because the KH potential does not change for the same α_0 and the absolute separations are in reverse proportion to the frequency of the laser pulse, the separations observed in Fig. 4(c) are half of those in Fig. 2 with $E_L=5.0$ a.u. and $\omega_L=1.0$ a.u. For example, the nearest two peaks beside the third harmonics are at 3.06 and 2.94, respectively, in Fig. 4(c). The results of the separation of 0.06 from the normal third order harmonic line multiplied by the photon energy equal also to the energy difference between the states of $1s\sigma_g$ and $2s\sigma_g$.

The hyper-Raman lines can also be seen in Fig. 4(a) for the laser pulse with the often used wavelength of 1064 nm in experiment. The lines are mainly caused by the transitions between $1s$ and $2s$ bound states of the hydrogen atom and locate at $\omega_i \pm (2n+1)\omega_L$ due to parity conservation. These hyper-Raman lines had been predicted in many previous studies, where the ionization is avoided by reducing the intensity of the laser field and the occurrence of the hyper-Raman process depends sensitively on the form of the initial

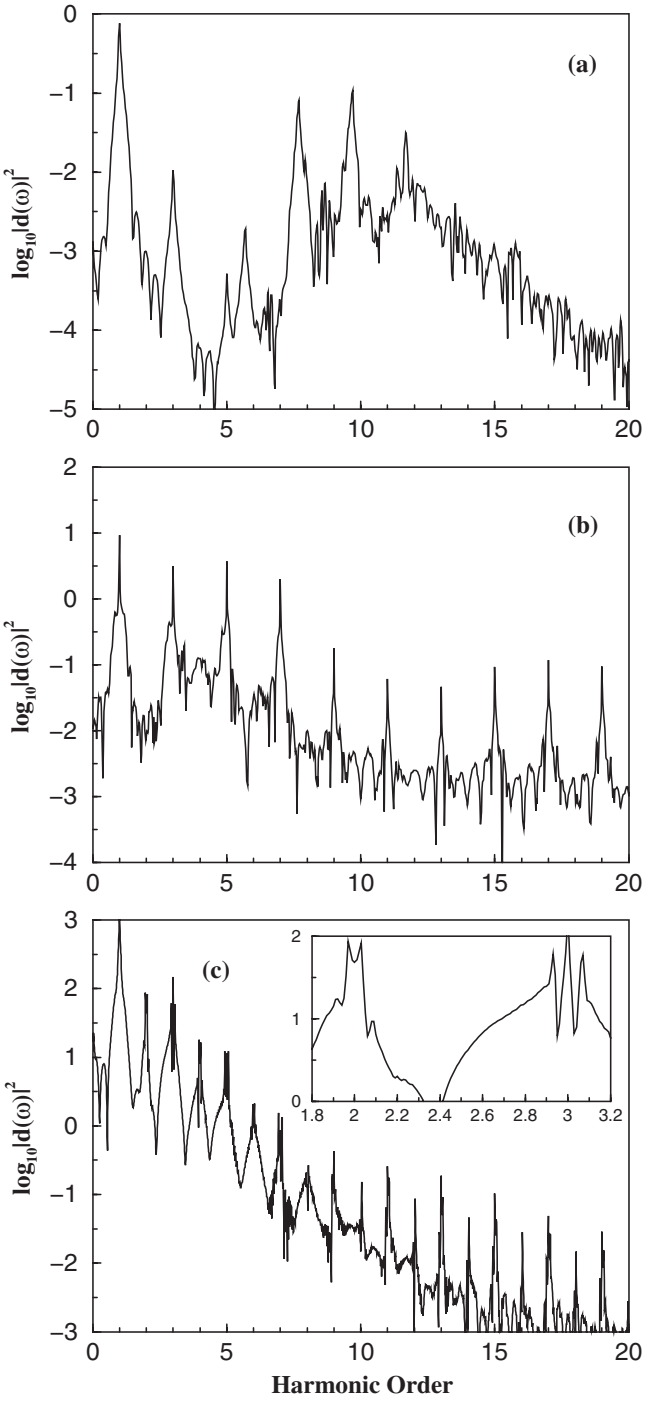

FIG. 4. Photon emission spectrum of the hydrogen atom in the laser field with same $\alpha_0=E_L/\omega_L^2=5.0$ a.u. and different frequency of (a) $\omega_L=0.0428$ a.u., (b) $\omega_L=0.1$ a.u., and (c) $\omega_L=2.0$ a.u.

state. However, with a too low laser intensity both the harmonic and hyper-Raman lines are too weak to detect. Figures 5(a) and 5(b) are the spectra for $\omega_L=0.0428$ and 1.0 a.u. with the initial state being taken to be the $2s$ bound state of hydrogen atom, the emission spectra by choosing the ground state as the initial state are also presented for comparison. The electron is much more easy to be ionized when the $2s$ state is taken as the initial state and the odd harmonic lines are more obvious for the low-frequency laser as shown in

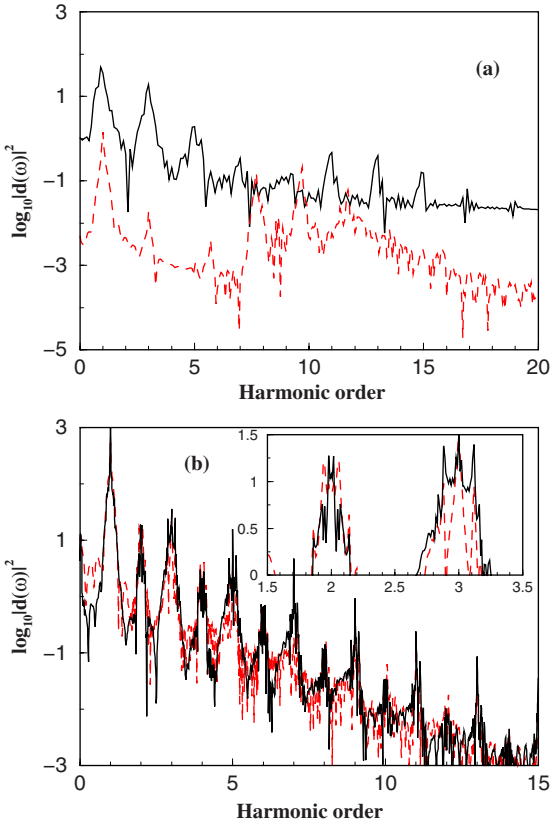

FIG. 5. (Color online) Photon emission spectrum of the hydrogen atom in the laser field with the 2*s* (solid line) and 1*s* (dashed line) bound state as the initial state (a) ω_L=0.0428 a.u., E_L =0.00916 a.u. and (b) ω_L=1.0 a.u., E_L=5.0 a.u.

Fig. 5(a). The potential and eigenstates change so much for the latter case that the preparation of the initial states is inessential in this case. The emission spectrum as well as the position of hyper-Raman lines does not change much. By comparing the two sets results plotted in Fig. 5(a), one can see that with the 1*s* initial state the emission is too weak to detect while with the 2*s* initial state the ionization is too much, resulting in only the harmonic lines. So the absence of the hyper-Raman lines in the existing experiments might be due to the hard satisfied requirements for the initial state for the infrared lasers and under the stabilization conditions with high-frequency lasers it is possible to observe the hyper-Raman process in experiment.

IV. CONCLUSION

The dynamical response and photon emission spectrum of the hydrogen atom in an intense high-frequency laser field are calculated and interpreted in terms of the transitions between the dressed states of the time averaged KH potential. When stabilization occurs, there is considerable possibility for electron oscillating between the bound states of the KH potential. The photon emission spectrum under this condition is complex because of these oscillations and the population spreading across many eigenstates of the KH potential during the turn-on time. The hyper-Raman processes can be strengthened resulting in clear splitting around both even and odd harmonics. It has been deduced that the spaces between the hyper-Raman lines around the even harmonics are determined by the energy difference of eigenstates with different parity and those around the odd harmonics by the energy difference of the same parity because of the parity conservation. The lines can be significantly restrained with a more slowly turned on pulse when the population concentrates on the ground state of the KH potential. So the symmetric hyper-Raman lines can be seen when the one photon energy exceeds the ionization energy of the atom and causes the stabilization. The hyper-Raman lines can also be seen for an infrared laser field if the intensity is too low to ionize the electron, however, their appearance depends sensitively on the initial state.

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