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High-order harmonic generation spectrum of an excited one-dimensional Coulomb atom in an intense laser field*

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Response of the wave packet of a one-dimensional Coulomb atom to an intense laser field is calculated using the symmetrized split operator fast Fourier method. The high-order harmonic generation (HHG) of the initial state separately being the ground and excited states is presented. When the hardness parameter α in the soft Coulomb potential $V(x) = -1/\sqrt{x^2+\alpha}$ is chosen to be small enough, the so-called hard Coulomb potential $V(x) = -1/|x|$ can be obtained. It is well known that the hard one-dimensional Coulomb atom has an unstable ground state with an energy eigenvalue of ~ 0.5 and it has no states corresponding to physical states in the true atoms, and has the first and second excited states being degenerate. The parity effects on the HHG can be seen from the first and second excited states of the hard one-dimensional Coulomb atom. The HHG spectra of the excited states from both the soft and hard Coulomb atom models are shown to have more complex structures and to be much stronger than the corresponding HHG spectrum of the ground state of the soft Coulomb model with $\alpha = 2$ in the same laser field. Laser-induced non-resonant one-photon emission is also observed.

Keywords: high order harmonic generation, intense laser field, one-dimensional Coulomb atom
PACC: 3280K, 3115, 4265K

The interaction of an intense laser field with an atom has been an active subject for many years. Many theoretical researches have been carried out based on the one-dimensional (1D) nonsingular atomic potential in the form of

$$V(x) = -1/\sqrt{x^2+\alpha}\,. \tag{1}$$

Such a model was suggested some years ago[1,2] and is usually called the soft Coulomb field, where the hardness parameter α is used to avoid numerical problems associated with the singularity of the potential at the origin $x = 0$. When dealing with the one-dimensional singular Coulomb field, the following potential that is usually called hard Coulomb potential is used:[3]

$$V(x) = -1/|x|. \tag{2}$$

The value of α can also be changed to adjust the depth of potential and often chosen to be 1.0[3−5] or 2.0.[6] However, little attention has been paid to the dependence of the derived conclusion on the α value and also to the similarities and dissimilarities between the soft and hard Coulomb fields. It has also been predicted most recently that excited states response quite differently from the ground state.[7,8] In the present study, the action of the model of soft Coulomb atom in an intense laser pulse is calculated by using a symmetrized split operator fast Fourier method. Change of the generation spectrum, in particular the high-order harmonic generation (HHG) spectrum, with different α value will be given to show the correspondence of the model atom states on the states of the real atoms. By choosing a very small α, HHG spectrum of the hard Coulomb atom will also be calculated to show the parity effect and the non-resonant one-photon emission.

The 1D Coulomb model's field-free Hamiltonian is

$$\hat{H}_0(x) = -\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{\sqrt{x^2+\alpha}}\,. \tag{3}$$

Generally the corresponding Schrodinger equation does not have analytic solutions, but we can obtain the eigenvalues and corresponding eigenfunctions numerically. For $\alpha = 2$, the energy eigenvalue of ground state is -0.5 equal to that of a real hydro-

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gen atom. The eigenvalue of the ground state is going down monotonously with the value of the parameter α becoming smaller. When α is small enough, the Coulomb model can be regarded as the hard Coulomb model,[9] whose Hamiltonian is

$$\hat{H}_0(x) = \lim_{\alpha\to 0}\Big(-\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{\sqrt{x^2+\alpha}}\Big) = -\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{|x|}. \quad (4)$$

Schwengelbeck and Faisal[9] and Loudon[10] have discussed the potential in the expression (4). In contrast to the soft Coulomb model, the hard Coulomb model allows degeneracy of the eigenfunctions having even($\sigma = 0$) and odd ($\sigma = 1$) parities

$$\phi_n^\sigma(\alpha = 0, x) = \sqrt{2/n^3}|x|(\mathrm{sgn}(x))^\sigma \times \exp\Big(-\frac{-|x|}{n}\Big) F\Big(1-n, 2, \frac{2|x|}{n}\Big), \quad (5)$$

where $n = 1, 2, \ldots, \infty$ and the eigenvalues are

$$\epsilon_n(\alpha = 0) = -\frac{1}{2n^2}, \quad (6)$$

the lowest energy eigenvalue is not degenerate and depends logarithmically on α as follows:

$$\epsilon_0(\alpha) \approx -2\Big[\ln\Big[\frac{1}{\alpha}\Big]\Big]^2, \quad \text{for} \quad \alpha \ll 1 \quad (7)$$

and

$$\epsilon_0(\alpha \to 0) \to -\infty, \quad (8)$$

implying an infinite binding energy, which is not a physical solution.

The interaction Hamiltonian

$$\hat{H}_\mathrm{I}(x,t) = -xE(t) \quad (9)$$

describes the influence of the laser field $E(t) = E_0 f(t)\sin(\omega t)$ in the dipole approximation, where E_0 denotes the strength, $f(t)$ the envelope, and ω the frequency of the laser pulse. Then our goal is to solve the time-dependent Schrödinger equation (TDSE)

$$\mathrm{i}\frac{\partial}{\partial t}\psi(x,t) = [\hat{H}_0(x) + \hat{H}_\mathrm{I}(x,t)]\psi(x,t)\,. \quad (10)$$

The calculations reported here are carried out by using the symmetrized split operator fast Fourier transform method (SSOFFT).[11−14] Given the initial wavefunction at $t = t_0$, $\psi(x, t_0)$, its time evolution on the potential surface $V(x)$ can be expressed as

$$\psi(x, t_0 + \delta t) = \mathrm{e}^{-\mathrm{i}[\hat{H}_0 + \hat{H}_\mathrm{I}]\delta t}\psi(x, t_0), \quad (11)$$

where δt must be small enough and satisfy[12,13,15]

$$\delta t < \pi/\delta V_\mathrm{max}\,, \quad (12)$$

where δV_max is the maximum excursion of the potential. To prevent the reflection of the wavefunction from the edge of the grid, we use a smooth cosine mask function confined to a moderately narrow band near the boundaries after each time evolution step to force the electron density to be zero on the edges of grid.

In the interaction of an intense laser field with atoms, many nonlinear phenomena have been observed, HHG is a typical one.[16−20] The HHG spectra has the following feature: there is a sharp fall from the driving frequency to a plateau, followed by a cut-off. For the HHG process, the harmonic position can be predicted by the quasiclassical three-step model,[21,22] namely

$$n_\text{cut-off} \simeq (I_\mathrm{p} + 3.17U_\mathrm{p})/\omega\,, \quad (13)$$

where I_p is the ionization potential of the atom and U_p the ponderomotive energy of the electron in the incident field. Furthermore we choose the electric field envelope as follows:

$$f(t) = \begin{cases} \Big(\sin\dfrac{\pi t}{6T}\Big)^2, & 0 < t \le 3T, \\ 1, & 3T < t \le T_\mathrm{max}, \end{cases} \quad (14)$$

where T denotes the optical cycles and here we calculate 10 optical cycles in total. The harmonic spectrum is calculated from the Fourier transform of the time-dependent acceleration,

$$|a(\omega)|^2 = \Big|\frac{1}{T_\mathrm{max}}\int_0^{T_\mathrm{max}} a(t)\mathrm{e}^{-\mathrm{i}\omega t}\mathrm{d}t\Big|^2, \quad (15)$$

where the field-induced acceleration $a(t)$ is obtained from the time-dependent wavefunction $\psi(x,t)$,

$$a(t) = \langle\psi(x,t)|\frac{\partial}{\partial x}V(x)|\psi(x,t)\rangle. \quad (16)$$

In the present calculation, the frequency and strength of the laser field is fixed at $\omega = 0.06$ a.u., and $E_0 = 0.08$ a.u. respectively. According to the expression (13), we have

$$n_\text{cut-off} = 23.48 + I_\mathrm{p}/0.06. \quad (17)$$

So when the laser field is fixed, the harmonic position is determined only by the ionization potential. In other words, as the hardness parameter α becomes larger, the cut-off position of harmonics should decrease. Figure 1 shows a comparison of the HHG spectrum with $\alpha = 0.5, 1.0, 2.0$, with the corresponding energy of the ground state $\epsilon_0 = -0.8911, -0.6699, -0.5$ a.u. respectively. According to expression (17) the cut-off harmonic positions are

$n_{\text{cut-off}} \simeq 39, 35, 33$ respectively. Clearly the calculations agree basically with the quasiclassical predictions. Figure 2 shows the time-dependent survival probability of the ground state in the three situations. We can see that the atom with larger α and smaller ionization potential is easier to ionize. From Figs.1 and 2, we can also find that the strength of the HHG depends on the space confinement of the initial wavefunction. The strength of the HHG increases with the space extension of the initial wavefunction before the atom is completely ionized after the laser pulse. In other words, in the situation of soft Coulomb potential, strength of the HHG increases with the increase of α before a specific value at which the atom is completely ionized after the laser pulse.

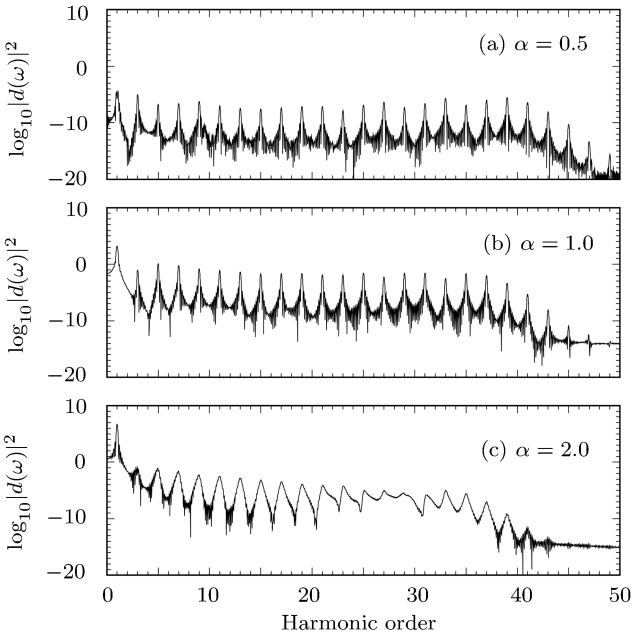


Fig.1. The HHG spectrum of the soft Coulomb potential with $\alpha = 0.5, 1.0$ and 2.0 in the same laser field.

As is well known, the ground state is confined within a smaller space with a less hardness parameter α. And for the hard Coulomb potential, we have $\phi_0(x) \to \delta(x)$ as $\alpha \to 0$, and then the Coulomb potential can be regarded as the hard Coulomb potential. This ground state has no real corresponding physical states. Here we choose $\alpha = 10^{-4}$ to approximate the states of the hard Coulomb potential, for which the eigenvalue of the ground state ($n = 0$) is -13.5088 a.u. calculated by using the shooting method. The energy eigenvalues are also determined by the spectrum method.[23] The spectrum method is a method of numerically solving the Schrödinger eigenvalue problem. This method first requires to compute the correlation function $\wp(t) = \langle\psi(x,0)|\psi(x,t)\rangle$, where $\psi(x,0)$ is the initial wavefunction and $\psi(x,t)$ represents the evolution of the wavefunction, by calculating the time-dependent Schrödinger equation without laser field. Then the value of $\wp(E)$, Fourier transform of $\wp(t)$, displays a set of sharp maximum points for $E = E_n$, where E_n are the energy eigenvalues. This energy spectrum for $\alpha = 10^{-4}$ is shown in Fig.3. We can see that the ground ($n = 0$) state almost cannot be found in the spectrum due to its very confined space extension.

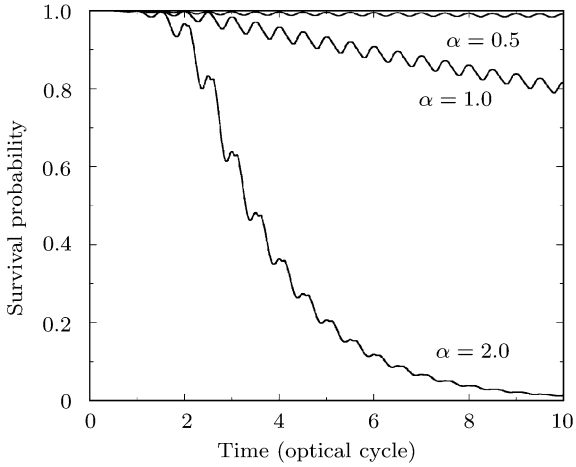


Fig.2. The survival probability of the ground state of the soft Coulomb potential with $\alpha = 0.5, 1.0, 2.0$ in the same laser field as that in Fig.1.

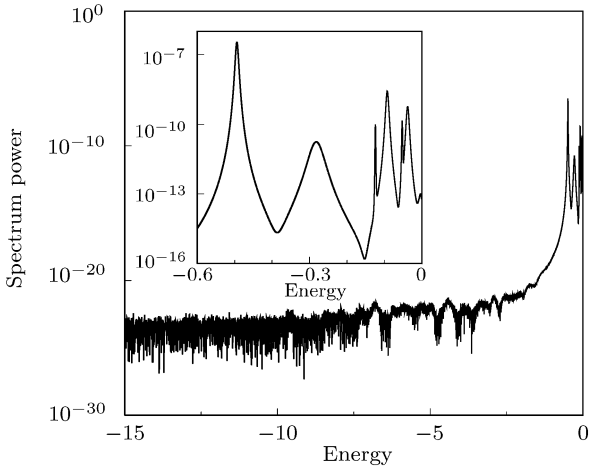


Fig.3. The energy spectrum of the hard Coulomb potential with $\alpha = 10^{-4}$.

We choose the initial wavefunction to be the first and second excited states whose energy eigenvalues are the same, i.e. -0.5 a.u., but their parities are different as shown in Fig.4. We take these two states as the analogous states of the 1D model hard Coulomb to those of a true hydrogen atom, and try to find

the parity effects on the dynamical response of the atoms to the laser pulse. The HHG spectra of the first and second excited states in an intense laser field are shown in Fig.5 with solid and dotted lines, respectively. The cut-off positions of both the two lines are near to 31 determined by the ionization potentials. However, the details of the spectrum display apparent differences. We can also conclude that the HHG spectrum is similar to that of the soft model atom when $\alpha = 2.0$ (see the dashed line in the same figure with the same cut-off position). As mentioned in Ref.[9], the hard Coulomb model has less stability than the soft Coulomb model, so the strength of the HHG spectrum of the hard Coulomb potential is higher than that of the soft one with more ionization probability. Nevertheless, the calculations with hard Coulomb model is more time consuming because the time step δt should be smaller than that of soft Coulomb model according to Eq.(12).

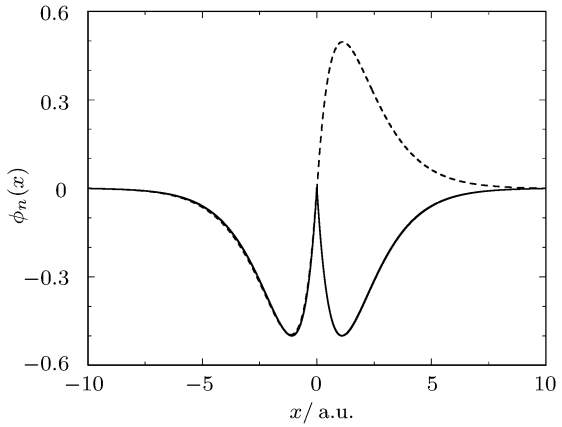


Fig.4. The wavefunctions of the double-degenerate first and second excited states of the hard Coulomb atom.

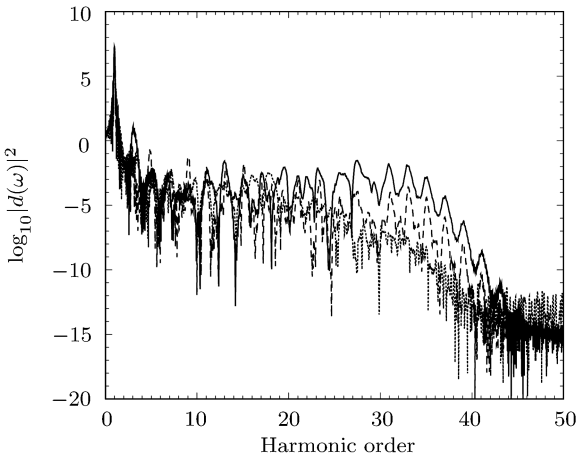


Fig.5. The HHG spectrum of hard Coulomb potential with the initial state being the $n = 1$ state (solid line), and $n = 2$ state (dotted line). The dashed line denotes the HHG spectrum of soft Coulomb potential $-1/\sqrt{x^2+2}$.

The laser-induced non-resonant one-photon emission is also observed in the HHG spectrum. A modulation has been carried out for the HHG of the laser-driven first excited state of the soft Coulomb atom with $\alpha = 0.01$. The spectrum is given in Fig.6, in which except for the normal HHG before 43, a strong emission is found around 55 corresponding to the energy difference between the first excited state and the ground state.

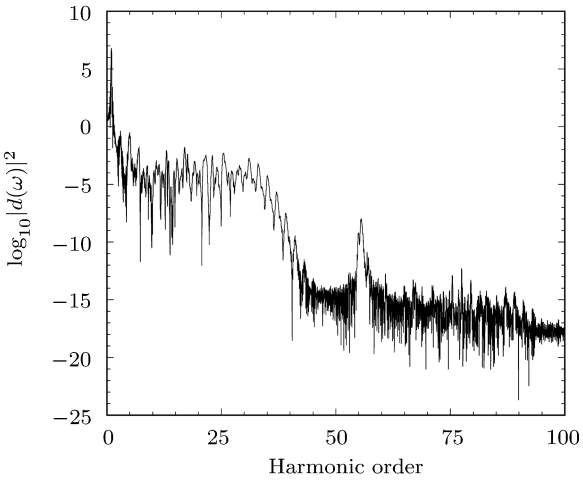


Fig.6. The HHG spectrum of the soft Coulomb potential with $\alpha = 0.01$. The first excited state is chosen to be the initial state to show the laser-induced non-resonant one-photon emission between the first excited state and the ground state.

To summarize, we have calculated the interaction of strong laser field, separately, with the so-called hard and soft Coulomb potential model atoms. By changing the value of the hardness parameter α of the soft model atom, from the different generations of the HHG we have deduced that the wavefunctions and the eigenvalues of the first excited state of the hard Coulomb model atom are physically analogous to those of the real 3D hydrogen atom. When α is growing, the energy levels split and the degeneration is eliminated. The characteristic of soft Coulomb potential can be seen. According to the quasiclassical three-step results, the cut-off position of HHG spectrum changes, with hardness parameter α of the soft Coulomb model atom. The soft Coulomb model is more stable with smaller values of α before the ground state appears to be unstable when the values of α becomes too small. When we select $\alpha = 2.0$ the soft Coulomb model results are similar to those of the first excited state of the hard Coulomb potential, but demands less calculating time step, so it is more practical in calculation.

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