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Wavelength Effects on Strong-Field Single Electron Ionization

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Abstract

In this paper, we perform *ab initio* quantum mechanical calculations on wavelength dependence of strong-field single electron ionization on two model atoms with different initial electron distributions. The numerical results show dramatically different wavelength dependence for different initial electron distributions.

During the past two decades, behaviors of atoms exposed to strong laser fields have been extensively studied and most effects of field-atom interactions can be relatively well understood by the single-active-electron (SAE) approximation [1-3]. Time-dependent quantum mechanical calculations using the SAE approximation have been shown to provide accurate single and sequential ionization rates, above-threshold ionization yield, high-harmonic spectra, and angular distributions for rare gas atoms in strong laser fields [1-3]. In the tunneling regime [4], the SAE-based Ammosov-Delone-Krainov (ADK) tunneling model also provides an accurate fit to single- and sequential multiple-electron ionization rates of rare gas atoms [5]. However, most of the subjects studied extensively in the past are the rare gas atoms that all have similar closed-shell electronic structures.

Molecules can potentially provide important tests of strong field dynamics as even easily studied diatomic molecules have a greater diversity of electronic structures. Despite the success in explaining single- and sequential multi-electron ionization in rare gas atoms, the interpretation of these responses in molecules has encountered difficulties. For example, even the most fundamental single-electron

ionization of diatomic molecules is still not fully understood. One of the well-known examples reflecting the complexity of molecules is the observation that single ionization yield of O_2 is substantially lower than that of the rare gas atom Xe, although O_2 has virtually the same ionization potential as Xe (~ 12 eV) [6,7]. At the first glance, the anomalous behavior of O_2 may be because that O_2 is a molecule and is different from the rare gas atom Xe. However, further experiments show that the low ion signal is not observed in N_2 [7], as compared to its corresponding rare gas atom Ar that has virtually the ionization potential (N_2: I_p~15.58 eV, Ar: I_p~15.76 eV). This indicates that the peculiar behavior of O_2 is not a simple consequence of its being a molecule. Motivated by these experimental findings, extensive theoretical studies [8-10] have been devoted to studying the different ionization behavior between N_2 versus O_2, and the difference between the two molecules is generally attributed to their distinctly different electronic structure as originally proposed in Ref. [7]: N_2 is a closed-shell molecule with its outermost two electron spatial wavefunction symmetrically distributed, while O_2 is an open-shell molecule with its outermost two electron wavefunction anti-symmetrically distributed. Despite the extensive theoretical studies on the suppression of O_2^+ in ultrashort 800-nm pulses, there has been little study on another critically important piece of experimental observation related to the suppression in O_2^+, i.e., wavelength dependence of O_2^+ suppression. It has been reported experimentally that the suppression in O_2^+ is only seen in 800-nm Ti:sapphire laser radiation, but not in 10.6-μm CO_2 laser light [6,11]. Motivated by this wavelength dependence in strong-field single electron ionization, we set out to further investigate this effect theoretically.

In this paper, *ab initio* two-electron quantum-mechanical calculations [12-15] are performed on the two model atoms at different frequencies. Two-electron model atoms are constructed with either a spatially symmetric electronic wavefunction (SSW) or a spatially anti-symmetric electronic wavefunction (SAW) [12,15], which correspond approximately to N_2 and O_2 molecules. The calculations clearly show that the single ionization probability increases in the SSW atom but decreases in the SAW atom as the external light frequency increases. The results of this investigation demonstrate that the behaviors of single-electron ionization of atoms or molecules in strong fields strongly depend on the initial electronic structures.

A direct numerical solution of the full time-dependent Schrödinger equation is a powerful method for investigating atoms in strong laser fields. First introduced by Eberly *et. al.* [16], the *ab initio* quantum mechanical calculations have been commonly performed in 1D using a so-called soft-core potential

[12-18]. Although a 1D model cannot fully characterize the true 3D conditions, the 1D results normally represent relatively well the physics of a 3D system [12-18]. For instance, the soft-core potential eliminates the 1D singularity at the origin and reproduces the 3D like Rydberg series of excited states [16]. Furthermore, the reduced dimension will allow us to handle more complex problems, such as multielectron interactions, using readily accessible computer resources today. Certainly, a cross-dimensional comparison (1D versus 3D) may not reveal all the underlying physics in realistic experimental conditions. Therefore, in this work, instead of comparing a 1D system to the real 3D experimental results, we only compare the ionization dynamics of a 1D SSW model atom to a 1D SAW model atom theoretically. The two model atoms in our study are both in 1D and thus any difference resulting between the two systems is expected to reveal some effects of different initial conditions.

The total Hamiltonian of a two-electron system with full electron-electron interaction and external laser field is, in atomic units (a.u.) [12-15],

$$H = -\frac{1}{2}\frac{\partial^2}{\partial^2 x} - \frac{1}{2}\frac{\partial^2}{\partial^2 y} - \frac{Z}{\sqrt{x^2+a^2}} - \frac{Z}{\sqrt{y^2+a^2}} + \frac{1}{\sqrt{(x-y)^2+a^2}} + E(t)(x+y)$$

where x and y are the coordinates of electron 1 and 2, respectively. Z is the charge of the atomic core. A soft-core potential well is used to characterize the Coulomb interactions with the parameter a controls the width of the potential well [13]. $E(t)$ is the external laser field strength, and the interaction between atom and external field is based on the dipole coupling. The time dependent Schrödinger equation is numerically solved with a split-step Fourier method by evaluating the kinetic-energy action in the Fourier space and the potential-energy action in the real space [13]. The spatial grid spacing and temporal step size are set to be less than 0.4 a.u. and 0.1 a.u., respectively. An absorbing boundary is used to avoid reflections from the boundaries [13,14]. In our calculations, we choose $Z = 2$ and $a = 0.74$ to construct our SSW and SAW model atoms. The ground state energy is determined by evolving a random seed wavefunction in field free conditions along the imaginary time axis with the initial wavefunction either spatially symmetric or anti-symmetric [12]. Figure 1 shows the probability densities of the resulting initial ground state of the SSW and SAW model atoms. In principle, the symmetry of the wavefunction will be conserved when freely propagating in time, however, we still resymmetrize the wavefunction at each time step in order to prevent numerical roundoff error from contaminating the spatial symmetry. In this way, the

ground state energy for the SSW and SAW atoms are found to be -2.9 a.u. and -2.3 a.u., respectively.

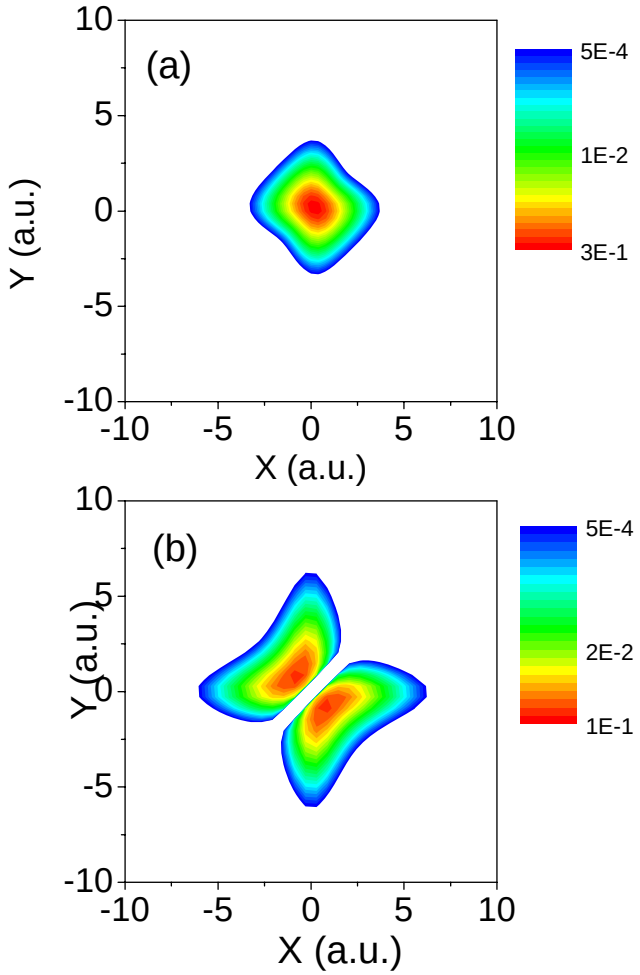

Figure 1. The ground state probability densities of the (a) SSW and (b) SAW model atoms.

In order to keep a moderate computational effort, a sine-squared shaped pulse is used with six cycles at a frequency of $\omega = 0.4$ a.u. and three cycles at $\omega = 0.2$ a.u. This way, the pulse duration at the two frequencies will be identical. Following Refs. [12,14,15], the single and double ionization probabilities are obtained by integrating the electron density in the corresponding spatial regions after the laser pulse, where ionization is defined by an electron being a certain distance, α, away from the core. An ionization distance $\alpha = 5.28$ a.u. is used for both SSW and SAW atoms, so that an electron experiences the same Coulomb attraction during ionization for both model atoms. Similar numerical approach

comparing 1D SSW and SAW model atoms has been used in the past to study the influence of electron wavefunction spatial symmetry on nonsequential double ionization [12,15]. Here, we will focus on studying the wavelength effects on the most fundamental single electron ionization for atoms/molecules with different electron wavefunction distributions.

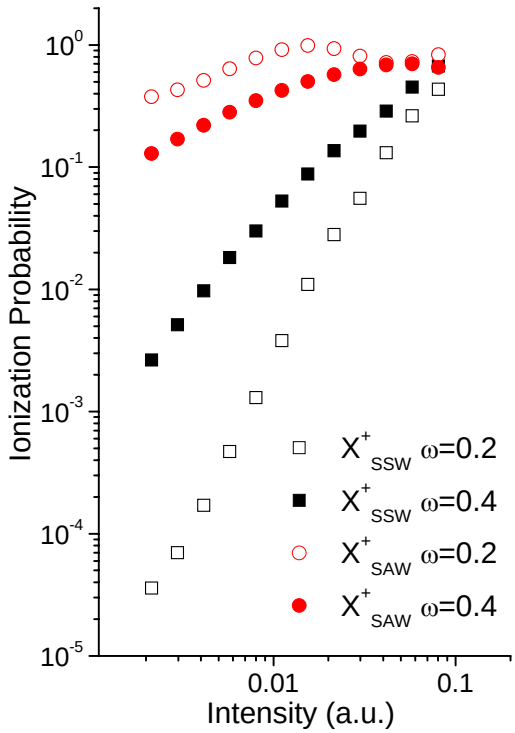

Figure 2. Single ionization probabilities of the SSW and SAW model atoms.

The single ionization probabilities for the SSW and SAW atoms are evaluated at frequencies 0.4 a.u. and 0.2 a.u. and the results are plotted in Fig.2. We first notice that the single ionization probabilities of the SAW atom are higher than the SSW atom. We believe that the higher ionization rate of the SAW atom is because the initial anti-symmetric double-leaf-shaped wavefunction [Fig. 1 (b)] is

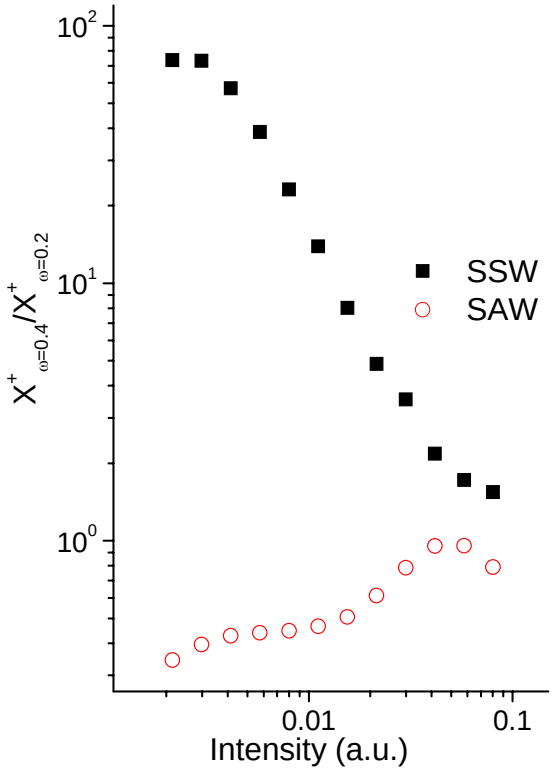

Figure 3. The intensity dependent ratio, $X^+_{\omega=0.4}/X^+_{\omega=0.2}$, for the SSW and SAW model atoms.

easier to reach the ionization boundary than the SSW atom in our 1D model calculations. Therefore, this does not necessarily indicate that the SAW atom is easier to ionize in real experimental conditions. Here, we will only focus on comparing the single ionization probabilities between the SSW atom and the SAW atom at different frequencies. Since the only difference between the two model atoms is their initial electron distribution, we expect that any difference resulting from the calculations is due to the effects of the different initial electron distribution between SSW and SAW. As the frequency ω increases from 0.2 a.u. to 0.4 a.u., we can see from Fig. 2 that ionization probability increases with frequency for the SSW atom. This is relatively straightforward to understand because it needs less photons to ionize an electron as ω increases, and the nonlinear interaction probability is always higher for a lower order effect. For the SAW atom, however, we see a completely opposite ionization behavior as ω increases: single ionization probability of the SAW atom actually decreases as ω

increases from 0.2 to 0.4 a.u., and this actually indicates that the probability of a lower-order (ω = 0.4 a.u.) nonlinear process is less likely to occur than a higher-order (ω = 0.2 a.u.) nonlinear process. This single ionization difference between the SSW and SAW atoms can be seen more clearly from the ratio plot of $X^{+}_{\omega=0.4}/X^{+}_{\omega=0.2}$, as shown in Fig. 3. Below the saturation intensity of 0.02 a.u., we can see from Fig. 3 that the ratio curve is less than unity for the SAW atom but above unity for the SSW atom, indicating that ionization probability increases when ω increases from 0.2 to 0.4 a.u. for the SSW atom but decreases for the SAW atom below their corresponding saturation intensities.

To understand the different single ionization behaviors between the SSW and SAW atoms, we resort models that consider the effects of electron distribution on ionization dynamics. As mentioned earlier, based on the concept that the detailed electronic structures play a key role in molecular ionization [7], several models have show some success in explaining the suppressed ionization in O_2^+ [8-10]. Among these models, a so-called multielectron screening model [8] explicitly explains the critical experimental observation that has not been discussed by the other models, i.e., the suppression in O_2^+ is only seen in 800 nm Ti:sapphire laser radiation, but not in 10.6 μm CO_2 laser light [6,11]. Since the multielectron screening model [8] explicitly takes into account the wavelength effects on electron ionization, we will try to relate our simulation results here in the framework of this model. As discussed in details in Ref. [8], the multielectron screening model predicts that electron screening will become less effective in a SAW atom or molecule, such as O_2, when the external light frequency increases. Based on this argument, we expect that single electron ionization will become harder at a shorter wavelength/higher frequency for a SAW atom or molecule with two electrons residing on the opposite sides of the core. This is precisely what we have observed in Figs. 2 and 3 for the SAW atom ionization when ω increases from 0.2 to 0.4 a.u. However, unlike the SAW atom, the two electrons in the SSW atom are more uniformly distributed and are less distinguishable. As discussed in Ref. [8], a more uniform overall electron distribution is the key for atomic-like ionization behaviors in a molecule, e.g., for the closed-shell molecule N_2. Electron screening strength should not change noticeably with frequency for the SSW atom, and the efficiency of different order of nonlinear processes should play a greater role for the SSW atom. Therefore, we see that ionization probability increases when ω increases from 0.2 to 0.4 a.u. for the SSW atom since the lower order nonlinear process is more efficient (corresponding to 0.4 a.u.).

In summary, we perform a comparative study of wavelength effects on single ionization in two model atoms with different initial electron distributions. The

numerical results show that single ionization probability increases with frequency for the SSW atom but decreases with frequency for the SAW atom. Although our simplified calculations do not necessarily represent exactly a realistic experimental condition, the results of this investigation demonstrate that the behaviors of single-electron ionization in strong fields critically depend on the initial electronic structures.

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