The influence of spatial symmetry on the dynamics of strong-field = ionization

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Abstract

The effect of spatial symmetry on strong field ionization is = investigated with 1-D two-electron *ab initio* quantum mechanical calculations. Numerical results clearly show that the nonsequential double ionization = rate in atoms with spatially symmetric wavefunctions is higher than in atoms = with anti-symmetric wavefunctions. Moreover, single ionization is = demonstrate, for the first time, to be correlated to double ionization indicating that strong field ionization is in fact a highly dynamic process.

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Many phenomena in strong field laser physics can be well described by = the single active electron (SAE) approximation when single-electron = excitation and ionization dominate. Time dependent quantum mechanical calculations using the SAE approximation have been shown to provide accurate single ionization rates, above-threshold ionization spectra and angular distributions for rare gas atoms in strong laser fields [1–4]. However, the SAE approximation clearly fails = when multielectron effects are not negligible. In helium, for example, even though the SAE based calculation can give a highly accurate fit to the single ion yield, it systematically underestimates the double ion yield. This anomalous double ionization is commonly referred to as = non-sequential double ionization (NSDI) [4–6]. Two theoretical = models have been proposed to explain the experimental observations in the = earlier studies of NSDI. The so-called "shake-off" model suggests that as one electron is ionized, another electron could be "shaken off" into the continuum due to the rearrangement caused by the sudden removal of the = first electron [5]. Unfortunately, this "shake-off" model is still = not fully tested due to the extreme demands of a realistic calculation. The "rescattering" model states that an electron can collide with its = parent ion core one half cycle after being ionized by a laser field, and this = e-2e rescattering could lead to enhanced double ionization [7]. = However, the rescattering model fails to predict the observed NSDI threshold and magnitude [4].

More recently, we reported the experimental investigation of single and double ionization of diatomic molecules in strong laser fields = [8]. In this study, the diatomic molecule N_2 behaves like a = structureless atom for both single and double ionization. However, O_2 shows = clearly nonatom-like behavior, having both single and NSDI rates below those of = an atom with the same ionization potential. Electronic structure is = believed to play a key role leading to these different ionization behaviors since = N_2 is a closed-shell molecule in a singlet state, while O_2 is an open-shell molecule in a triplet state. Perhaps more significantly, the outermost two electron wavefunction is spatially symmetric in N_2 = and anti-symmetric in O_2. Motivated by these distinctly different ionization behaviors between N_2 and O_2, we began to further investigate the underlying physics behind these observations = theoretically.

In this paper, model atoms are constructed with either a spatially = symmetric electronic wavefunction (SSW) or a spatially anti-symmetric wavefunction (SAW) which correspond approximately to N_2 and O_2 molecules. = For the first time, an exact two-electron quantum mechanical calculation is performed in one spatial dimension (1D) on both SSW and SAW atoms. The calculation clearly shows that the atom with a SSW has a higher NSDI = rate than the one with a SAW. Furthermore, the SSW and SAW atoms show = different structures in the single ionization probability as a function of = intensity indicating that the single ionization depends on the wavefunction = symmetry, as well. The results of this investigation demonstrate that even the = single electron motion in strong fields cannot be fully described by the SAE approximation.

A direct numerical solution of the full time-dependent Schrodinger = equation is a powerful method to investigate atoms in strong laser fields. For a two-electron system, models are often used to study NSDI in a 1D or 3D atomic system [9,10], but an exact treatment is always = desirable in order to reveal the underlying physics without any assumptions. = However, the *ab initio* calculations are mostly limited to 1D systems due to = the heavy computational demands. Although a 1D model cannot fully = characterize the true 3D conditions, the 1D results normally represent relatively = well the physics of a 3D system. For instance, a commonly used soft-core potential eliminates the 1D singularity at the origin and reproduces the = 3D like Rydberg series of excited states [11]. There are certainly = some drawbacks to the 1D treatment, such as the lack of angular = distributions, no level degeneracies and the inability to treat circularly polarized = fields.

Using 1D *ab initio* multielectron calculations to simulate 3D = experimental results has been rigorously pursued in the previous studies = [12,13].=20 However, these cross dimension comparisons may not be able to reveal all = the underlying physics in realistic experimental conditions. Instead of = comparing a 1D system to the real 3D experimental results, we take a new = approach by comparing the ionization dynamics of a 1D SSW atom to a 1D = SAW atom. Certainly, this comparison will not lead to a direct = explanation of our experimental results in Ref. [8]. However, = the two model atoms in our study are both in 1D and thus, any difference = resulting between the two systems is expected to reveal the effects of = different initial conditions. In fact, this comparison allows us to = concentrate on the study of only the spatial symmetry effects of = wavefunction without involving the experimental details and, therefore, = the simplified 1D model and idealized laser parameters (discussed later) = we are going to use in our calculation become relatively unimportant in = this purely theoretical (instead of phenomenological) investigation.

The total Hamiltonian of a two-electron system with full electron-electron interaction and external laser field is, in atomic = units (a.u.),=20

$$H = 3D - \frac{1}{2}\frac{\partial^2}{\partial =^2 x} - \frac{1}{2}\frac{\partial^2}{\partial =^2 y} - \frac{Z}{\sqrt{x^2+a^2}} - \frac{Z}{\sqrt{y^2+a^2}} + \frac{1}{\sqrt{(x-y)^2+a^2}} - E(t)(x+y) \quad (1)$$

where x and y are the coordinates of electron 1 and 2 respectively. = Z is the charge of the atomic core, and the parameter a controls the = width of the potential well. $E(t)$ is the external laser field strength, and = the interaction between the atom and external field is based on the dipole coupling. Most of the existing computational studies are based on the = helium atom with the SSW as the ground state wavefunction. Even though we are performing a more general study, in order to establish some comparison between the existing results and our SSW atom, we also construct a = helium atom by choosing Z =3D 2 and a =3D 0.74. The ground state energy is determined by evolving a seed wavefunction in field free conditions = along the imaginary time axis with the initial wavefunction either spatially symmetric or anti-symmetric [14]. 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 is found to be 2.905 a.u. for the SSW atom = and 2.307 a.u. for the SAW atom. It should be noted that the SAW atom does = not exactly represent the character of the O_2 molecule since the = outermost two electrons reside on two degenerate energy levels in O_2, but = they still both have the SAW. The electron density of the SSW and SAW atoms = after the imaginary time evolution is plotted in Fig. 1. We can see that the electron density is uniformly distributed in the SSW atom but has a = valley along the x =3D y direction in the SAW atom.

The computational method used here for obtaining the exact single and = double ionization probability is similar to the one described in Ref. = [13]. In order to keep a moderate computational effort, a six-cycle = sin-squared shaped pulse with a relatively high frequency, ω =3D 0.4 a.u., = is used. 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 at least 6 a.u. away = from the core. This method has been verified to agree with the one of = projecting the wavefunction onto ionic bound states [9]. After the first electron is ionized, a one-electron He^+ ion is formed and can be studied by solving the 1D Schrodinger equation,=20

$$i\frac{\partial}{\partial t}\Psi(x,t) = 3D(-\frac{1}{2}\frac{\partial =^2}{\partial^2 x} - \frac{Z}{\sqrt{x^2+a^2}} - xE(t))\Psi(x,t) = \tag{2}$$

with Z =3D 2 yielding the ionization probability, P_{1-2}. The = product of P_1P_{1-2} will therefore be the ionization probability of = sequential double ionization, where P_1 is the single ionization probability of = the original atom. The NSDI component can be obtained by subtracting the sequential component from the total double ionization yield.

The single ionization probabilities of both SSW and SAW atoms are also obtained within the SAE approximation by solving Eq.(2) with Z = =3D 1 and choosing the parameter a so that the ground state of a single = active electron matches the deduced single ionization energy. We adjust a = instead of the charge Z to obtain the correct ionization energy because a = mainly affects the short-range potential, while changing Z will produce a long-range effect which might overestimate the interaction when the = electron is far away from the core region. However, the difference for the = ionization probability between adjusting a and Z was found to be small = [13].

The single, sequential and total double ion signal based on the exact treatment and SAE calculations for both SSW and SAW atoms are plotted in Figs. 2 and 3. We can see, from the figures, that total double ion production is higher than the sequential component in both SSW and SAW = atoms indicating that there must be a non-sequential component in both cases. However, the difference between the total and sequential component is = more significant in the SSW atom. This is seen more clearly in the plot of = the non-sequential component vs. total double ionization production in Fig. = 4. Below the intensity of 0.08 a.u. where the single ion yield starts to saturate, there is more NS component in the double ionization for the = SSW atom than the SAW atom, which is consistent with our experimental = observation that N_2 has a higher NSDI rate than O_2 in terms of spatial = symmetry of wavefunction [8]. Note that even though the ionization potential is = different between our SSW and SAW atom, the NSDI=20 rates in both cases are divided by the total double ionization rates. = This normalization process makes the comparison valid despite the = difference in ionization potentials of two model atoms.

The intensity dependence of the ratio X^{2+}/X^{+} has been used to = indicate NSDI [4,8], where X^{+} and X^{2+} are the yields for single and double ionization of same atom or molecule X. If X^{2+} is produced in = a sequential process, X^{+} will be the intermediate species and the = ratio X^{2+}/X^{+} will reflect just the ionization rate from X^{+} to = X^{2+} which will have a strong intensity dependence. However, if this ratio is only weakly dependent on intensity, it shows that the precursor to = X^{2+} is not X^{+}, and the ionization involves the NS process. In our = previous experimental report, the weak intensity dependent ratio of = N_2^{2+}/N_2^{+} is about ten times higher than O_2^{2+}/O_2^{+} [8]. In Fig. 5, the ratio curves are also plotted with the numerical results obtained here. Due to the reduced-dimension and relatively high laser frequency, we do not observe a significant flattening of ratio = curves, which indicates NSDI. However, a clear shoulder is seen in the SSW atom = in the intensity range of 0.04 - 0.07 a.u. but not in the SAW atom. This = again reinforces our argument that NSDI is more significant in a SSW molecule = than in a SAW molecule. Caution needs to be taken in comparing the mechanism = between the model atoms here and the molecules in our previous = experimental investigation since molecules are often complicated by its = extra degrees of freedoms, such as enhanced ionization at critical = internuclear separation (R_c) [15]. However, it has been = demonstrated that the ionization in N_2 and O_2 in our previous = experiment in fact occurs before R_c is reached [16]. = Therefore, the ionization in the molecules we studied can be viewed as = happening instantaneously, especially for single ionization and NSDI. = This leads the molecular nature in our experiment relatively = unimportant, rather the spatial symmetry of wavefunction is in fact the = key factor leading to the different ionization behaviors in N_2 and = O_2.=20

Figs. 2 and 3 show that the single ionization probability obtained = through the exact treatment in both the SSW and SAW atoms increases with = intensity up to a saturation point, where the double ionization has a comparable strength with the single ion. When the intensity is further increased, = the single ionization curves start to decrease because the sequential = production of the double ionization begins to deplete the single ion yield. Below = the saturation intensity, the single ionization probability increases with intensity smoothly in the SAW atom while some structure is present in = the SSW curve, especially a dip in the ion curve in the range of 0.04 - 0.07 a.u. This dip also appeared in the calculations of Ref. [13] but = its significance was not pointed out: the intensity at which the dip appears = in the ion curve of the SSW atom is exactly the same as where the non-sequential component is most significant in the double ionization. Recently, the degree of electron correlation is also found to be a = maximum where there is a shoulder in the double ion yield [17]. This is = in fact reasonable because when NSDI is prominent, the two electrons share their energy allowing them to leave the core simultaneously which could = also suppress the ionization of a single electron. This is further = corroborated by the SAE based calculation, since there is no consideration for the dynamics of the second electron on the first ionized electron, and no structure is present in any part of the single ionization curve of = either the SSW or SAW atom. The dip is also not simply a depletion effect due = to the high double ionization rate as the double ionization does not make = up for the suppressed single electron ionization. Therefore, the results of = the calculation clearly indicate that single-electron ionization is not an isolated process, rather it is highly related to the dynamics of the = second electron motion.

Finally, the SAE based calculation gives about the same degree of single ionization as the exact treatment in the SSW atom, while underestimating = the ionization strength in the SAW atom. Thus, the observed suppression of single ionization in O_2 is not reproduced in our calculation of the = SAW atom, which might be because the model atom we are using here does not = have quite the same electronic structure as O_2. It is also inappropriate = to draw a conclusion that there will be any enhancement in the single ionization of the SAW atom since the calculation performed here does not represent realistic experimental conditions. If we again only make a relative comparison between the results of SAE and exact treatments, it should be noticed that the exact treatment does not give as high an ionization signal in the SSW atom as in the SAW atom with respect to the = SAE results. This may indicate that the overall single ionization strength = in the SSW atom with the exact treatment is suppressed compared to that in = the SAW atom. If this is the case, it again reinforces the possibility that = the single ionization is highly related to NSDI because NSDI is more = significant in the SSW atom than in the SAW atom over the whole intensity range = below the saturation point, and this can lead an overall suppression in the = single ionization production in the SSW atom, like the case in which a dip = appears in the single ion curve at the intensity where the NSDI is most = significant.

In summary, a comparison study was performed, for the first = time, between 1D SSW and SAW model atoms in strong laser fields using = *ab initio* quantum mechanical calculations. The numerical results = show that the NSDI rate in the SSW atom is higher than in the SAW atom = in 1D and multiphoton ionization regime=20 (due to the relative high laser frequency [18]) over the whole intensity range below the saturation point. This spatial = wavefunction symmetry has the same effects on NSDI as in our previous = experimental observation in N_2 and O_2 in tunneling regime = [8,18]. Therefore, the spatial symmetry of the initial = electronic wavefunction and the electron correlation effects due to this = symmetry seem to play a key role in influencing the NSDI in both the = multiphoton and tunneling regimes. Furthermore, the different single = ionization behavior between the SSW and SAW atoms indicates that even = the single ionization is not an independent process, instead it is = related to the dynamics of the second electron motion. Despite the fact = that the calculation is performed only in 1D systems and with the = idealized laser parameters in the multiphoton regime, the differences = observed in this calculation between the SSW and SAW atoms have to be = attributed to the spatial symmetry since every aspect of the two systems = are the same except the spatial symmetry of the initial wavefunction. = Further experimental and numerical investigations in both tunneling and = multiphoton regimes are necessary to fully explore the wavefunction = symmetry effects=20 we present in this paper. However, our results pinpoint, for the first = time, the influence of the spatial symmetry of the electronic = wavefunction on the dynamics of single and double ionization of atoms = and molecules in strong laser fields. Moreover, it provides insights in = understanding the wavefunction symmetry aspect of electron correlation = effects on strong-field multielectron phenomena = [14,19,20].=20

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FIGURE CAPTIONS

1. The electron density of the initial state for with a spatially symmetric electronic wavefunction (a) and a spatially anti-symmetric electronic wavefunction (b).

2. Single and double (total and sequential) ionization probability = for a spatially symmetric initial state with exact and SAE calculations.

3. Single and double (total and sequential) ionization probability = for spatially anti-symmetric initial state with exact and SAE calculations.

4. The fraction of the nonsequential component in the total double ionization for both the spatially symmetric and spatially anti-symmetric initial states.

5. The ratio curves of X^{2+}/X^{+} for both the spatially = symmetric and spatially anti-symmetric initial states.