Stabilization of Helium in Intense xuv Laser Fields

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We investigate the impact of electron-electron correlation on the ionization dynamics of helium in intense, high-frequency laser fields by solving the time-dependent Schrödinger equation from first principles. Although we observe a decrease in the total ionization yield at high field strengths, the hallmark of atomic stabilization, the repulsion between the electrons has a detrimental effect on the degree of stabilization, in particular for short pulses. Investigation of the ion channel yields reveals that the double ionization process is less prone to two-electron effects, and consequently exhibits the most distinct signature of stabilization. We also find that commonly used one-dimensional models tend to overestimate the effect of correlation.

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The process of multielectron ionization of atomic systems by photon impact is of fundamental interest. Double ionization by a single photon is one of the simplest processes in nature where electron-electron interactions come into play, as the mechanism for double-electron escape involves highly correlated electron motion. In this respect, the three-body breakup of helium serves as a prototype for understanding the role of electron correlations in more complex systems. The investigation of correlated dynamical processes poses a unique challenge to experiment and theory, and has occupied physicists for several decades, exemplified by the case of one-photon double ionization of helium [1–3]. More recently, the two-photon double ionization process has received considerable attention, both theoretically (see, e.g., [4] and references therein), and experimentally, employing state-of-the-art high-order harmonic [5] and free-electron laser (FEL) light sources [6,7].

Developments in FEL technology have enabled the production of laser pulses with unprecedented brilliance, and wavelengths ranging from vacuum ultraviolet to soft x rays [8,9]. The application of these pulses in experimental studies has provided a wealth of information on the multiphoton multiple ionization of complex atoms [10–12] and atom clusters [13]. With new projects being launched targeting the x-ray regime, for instance the European XFEL in Hamburg and the LCLS at SLAC (Stanford, USA), novel opportunities are opening for gaining further insight into complex atomic processes, such as multiphoton ionization and innershell dynamics. A parallel development of increasingly sophisticated theoretical models has taken place [1], and numerical *ab initio* studies providing very accurate descriptions of electron dynamics have been systematically pursued for intense-field multiphoton ionization of helium over a broad range of wavelengths, ranging from optical [14] to extreme ultraviolet (xuv) and x-ray radiation [15,16].

With the extremely high peak intensity expected to be delivered from future FEL technology, the process called atomic stabilization [17,18] might eventually become subject to experimental verification in the xuv regime. Thus far, stabilization has only been observed in Rydberg atoms at optical laser frequencies [19,20]. In addition to being of fundamental interest, stabilization is also relevant for applications of FEL radiation, such as biomolecular imaging with short x-ray pulses [21].

Primarily a high-frequency phenomenon, atomic stabilization is believed to occur when the photon energy exceeds the binding energy of the system [22]. According to theory, stabilization will set in at high laser intensities, and it points to the somewhat counterintuitive finding that the ionization yield may eventually enter a regime of decreasing ionization probability or rate with rising intensity, or alternatively, that the probability levels out at a value lower than one. The problem has been the subject of extensive theoretical study during the last two decades, in particular for one-electron systems (for reviews, see, e.g., [22–24]). Although more complex atoms are expected to reveal a similar behavior, studies of stabilization in multielectron systems are still scarce [22,25]. Calculations performed in one-dimensional (1D) two-electron model systems indicate that correlation effects could reduce the stabilization effect [26,27]. In another model study [28], it was found that the magnetic field component of the laser field could also have a detrimental effect on the degree of stabilization. However, it was recently shown, by solving the three-dimensional time-dependent Schrödinger equation beyond the dipole approximation for atomic hydrogen, that there is an intensity range in which the stabilization still persists [29].

In this Letter, we study the multiphoton ionization of a helium atom interacting with a very intense xuv attosecond pulse, in order to determine the extent to which stabilization occurs in two-electron atoms. In particular, we investigate the role of the electron-electron interaction in the ionization process. For this purpose, we solve the $(5 + 1)$-dimensional time-dependent Schrödinger equation

(TDSE), and as such we do not resort to models of reduced dimensionality. This is an extremely challenging computational task due to the high complexity of the problem, and for this reason we have developed a parallel numerical framework capable of meeting the computational demands. Using our numerical framework, we map out single and double ionization probabilities, as well as total ionization yields, versus the laser intensity for varying pulse durations and laser frequencies. We find that, although stabilization is observed in the helium system, the effect is suppressed due to the correlation, in particular for short pulses, but the suppression is less pronounced than reported in previous 1D model studies [27]. On the other hand, the helium system exhibits significant stabilization in the double ionization channel, and the stabilizing effect is typically much stronger than in corresponding one-electron (hydrogenic) systems. This type of stabilization is much less affected by two-electron effects, and might more easily be subjected to experimental verification.

The helium atom, which is prepared in the ground state, is exposed to a short, intense attosecond laser pulse. The field is linearly polarized and has a sine-squared temporal profile. For the pulses considered here, carrier-envelope phase effects, as well as nondipole effects [28,29] are not important and can be neglected. We consider a range of frequencies exceeding the ionization potentials of both electrons, i.e., $\hbar\omega > 79$ eV. In this regime, the absorption of a single photon is sufficient to doubly ionize the atom, via a combination of knockout and shakeoff processes [30]. However, for a single photon ionization process to occur, the electrons must exchange energy during the action of the pulse, and as such the process represents a clear departure from the independent-electron picture. In contrast, for higher order photon processes, i.e., n-photon ionization processes with $n \geq 2$, electron-electron interactions may play a less important role, and an independent-electron (IE) picture of the ionization process can be meaningful.

In order to solve the TDSE, we expand the wave function in a basis of B splines and coupled spherical harmonics [31]. The resulting set of sparsely coupled ordinary differential equations are quite stiff, and consequently, stable time integration is most readily assured by an implicit method. We have used the Cayley-Hamilton form of the propagator [31]. At each time step a system of linear equations must be solved, and for this we use an iterative preconditioned generalized minimal residual method, which typically converges to machine precision in only a few iterations. The preconditioner is based on an incomplete Gaussian elimination method implemented in the IFPACK toolkit, which is available in the Trilinos library [32].

The aforementioned IE model is constructed in a computationally similar manner. However, the two electrons are treated differently, in that the ''inner'' one moves in a $Z = 2$ Coulombic potential, while for the ''outer'' electron a screened potential is employed [33]. In this manner the single- and double ionization potentials of helium are correctly reproduced.

We extract the total ionization probability by projecting the final wave function onto all bound states of the neutral atom, which are obtained with the shift-invert Arnoldi method [34]. Double ionization probabilities are obtained by projection onto symmetrized products of one-electron Coulomb waves. The IE model predictions for the two-electron ionization probabilities are calculated from the single-electron probabilities for the ''inner'' and ''outer'' electrons. Both numerical schemes are built upon the PYPROP framework [35], and a detailed exposition will be presented in a forthcoming communication.

The question of numerical convergence is a difficult and subtle issue, additionally complicated by our considerations of many different laser parameters. Of main concern here is the size of the basis in terms of angular momenta and B splines included. In addition, the separation of single- and double ionization probabilities requires the use of a radial box large enough to contain the wave packet during the time evolution. By improving the basis and recalculating the results for a selected subset of laser parameters, we conclude upon comparison that the total ionization probabilities are converged to better than 1%. Separating single- and double ionization components presents additional difficulties, as the Coulomb wave projection technique only works in the asymptotic (Coulomb) region [36]; however, we believe that an accuracy of a few percent has been achieved for these quantities. The accuracy of our numerical method may also be ascertained in a different way, by calculating one-photon double ionization cross sections [1,2]. Considering several photon energies in the region 109–272 eV, we compared with experimental values [3], and found the agreement to be better than a few percent in all cases.

Having settled the issue of numerical accuracy, we proceed to the analysis of results from our calculations. Figure 1 shows the total, single- and double ionization probability versus electric field strength E_0, for a 6-cycle (182 as) laser pulse with $\omega = 5$ a.u. (atomic units). The contributions of the one- and two-photon double ionization processes are also shown for comparison. The dynamics is essentially perturbative up to electric fields strengths exceeding 1 a.u. According to lowest order perturbation theory the slope of the curves should be one and two, on a log-log scale, for the one- and two-photon processes, respectively, in excellent agreement with the full calculation for $E_0 \lesssim 1$ a.u. At low field strengths, nonsequential one-photon double ionization dominates, whereas at higher field strengths two-photon sequential and nonsequential ionization take over [15], with the sequential process dominating for longer pulses. Considering the total ionization probability at higher field strengths, we see that it is an increasing function up to some point ($E_0 \sim 17$ a.u.), where it attains a maximum value less than unity, and even becomes a slowly decreasing function with intensity; i.e.,

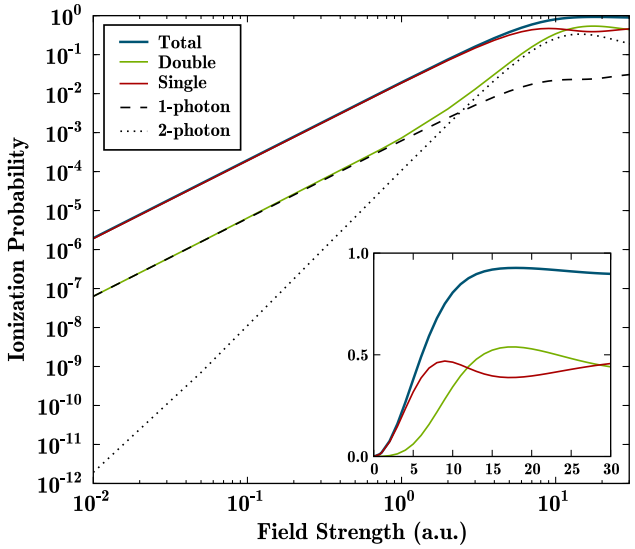

FIG. 1 (color online). Single (red or dark gray line), double (green or light gray line) and total (thick blue line) ionization probability of helium in its ground state versus electric field strength E_0 (in atomic units), for a 6-cycle laser pulse with $\hbar\omega = 136$ eV ($\omega = 5$ a.u.). Inset: same as above, but plotted on a linear scale. Dashed and dotted lines: corresponding one- and two-photon double ionization probabilities.

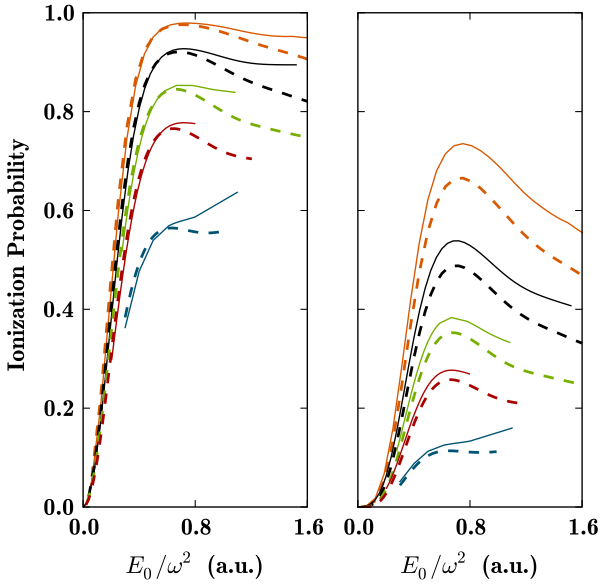

FIG. 2 (color online). Left panel: total ionization probability versus E_0/ω^2 for a 6-cycle laser pulse with central frequencies, 4 (upper orange line), 5 (second black line), 6 (third green line), 7 (fourth red line), and 10 a.u. (lower blue line). Dashed lines: corresponding IE model result. Right panel: double ionization probability.

the ionization process enters the atomic stabilization regime [22–24]. In order to obtain a distinct image of the stabilization region, the ionization yields are plotted on a linear scale in the inset in Fig. 1. We observe that the stabilization is not very efficient and much less pronounced than what is predicted by an independent-electron model (cf. Figure 2). Thus, the electron-electron interaction weakens the stabilization effect.

As expected, the single ionization dominates at lower field strengths. However, at a certain point, i.e., $E_0 \sim 9$ a.u., the single ionization probability reaches a local maximum and thereafter decreases for stronger fields. This apparent stabilization effect should not be mistaken for atomic stabilization. It merely reflects the fact that the population flow into the single ionization channel is smaller than the flow from the single channel into the double ionization channel. This is clearly seen in Fig. 1, as the double ionization probability increases significantly in this region. More interestingly, the single ionization probability reaches a minimum at $E_0 \sim 17$ a.u., and thereafter grows. This increase is accompanied by a corresponding decrease in the double ionization probability. Actually, the double ionization attains a maximum value at the point where the single ionization reaches its minimum value (cf. Figure 1). This is not a coincidence, but a demonstration of the effect that the double ionization channel is blocked. As a matter of fact, the mechanism responsible for this blockade is a true stabilization mechanism.

In Fig. 2 we plot ionization probabilities as a function of E_0/ω^2 for six-cycle pulses at five different laser frequencies, i.e., $\omega = 4$ (upper full curve), 5, 6, 7, and 10 a.u. (lower full curve). In each case, we find that the position at which stabilization sets in is uniquely controlled by the scaling parameter E_0/ω^2; i.e., the electric field corresponding to this position scales like ω^2, in contrast to the $\omega^{3/2}$ scaling law common in one-electron systems [22]. The total ionization results (left panel) show that the electron-electron interaction suppresses stabilization for all considered frequencies, and the effect is even absent for the shortest pulse(s) (highest frequency). In sharp contrast with this, the IE model (dashed lines) predicts significant stabilization in all cases. Even though stabilization is less likely in the total ionization yield, the stabilizing effect in the double ionization channel is striking (right panel), for all but the highest frequency (shortest pulse) considered. The apparent increase of correlation effects at high frequency is tied to the fixed-cycle pulses used, which implies shorter pulses as the frequency is increased. Under these circumstances the correlated nonsequential double ionization process becomes relatively more important. If, for instance, one increases the pulse duration from 6 to 12 cycles for the case with $\omega = 10$ a.u., sequential ionization becomes more likely, and the stabilizing effect is restored.

As suggested in the introduction, 1D models tend to overestimate the importance of correlation effects. For example, the one-dimensional helium model used by Bauer and Ceccherini [27] would predict complete breakdown of stabilization for all the pulses considered in Fig. 2. The explanation of why the diminishment of stabilization is less in 3D than 1D can be found by closely examining the way models of reduced dimensionality are implemented.

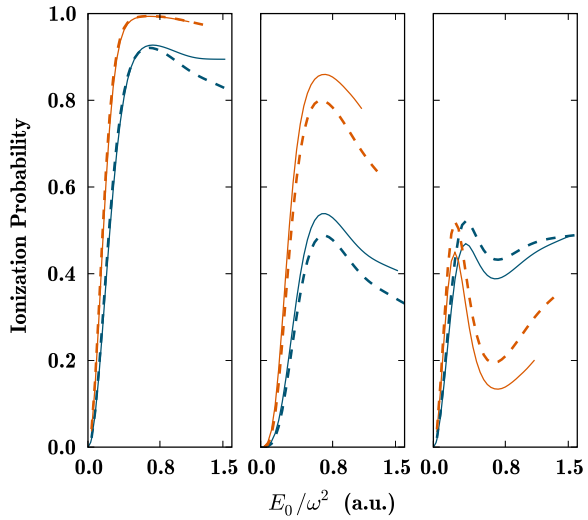

FIG. 3 (color online). Left panel: total ionization probability versus E_0/ω^2 for a 6 (lower blue line) and 12-cycle (upper orange line) laser pulse with $\omega = 5$ a.u. Dashed lines: corresponding IE model result. Middle panel: double ionization probability for the 12 (upper orange line) and 6-cycle (lower blue line) pulse. Right panel: single ionization probability for the 12 (lower orange line) and 6-cycle (upper blue line) pulse.

In such models the Coulomb interaction between particles is usually modeled by introducing a smoothing parameter in the potential, thus removing the singularity and allowing the particles to pass each other. However, one side effect of imposing such smoothing conditions is that the internal kinetic energy in the system is significantly reduced in proportion to the potential energies (repulsion and attraction), leading to an overemphasis on correlation effects.

Finally, we examine the effect of varying the pulse duration. Figure 3 shows total (left panel), double (middle panel), and single (right panel) ionization probabilities for a 12-cycle laser pulse. The 6-cycle result, as well as the IE result (dashed lines), are also shown for comparison. The figure expresses the fact that single ionization is strongly suppressed in favor of double ionization for longer pulses in the stabilization regime. Although atomic stabilization is still observed in the total ionization yield, the effect is very limited. On the other hand, the stabilization in the double ionization channel is still prominent, even for the longer pulse. Another interesting feature of Fig. 3 is that the IE model almost coincides with the full calculation for the total ion yield.

In conclusion, we have employed a recently developed scalable numerical scheme for solving the three-body time-dependent Schrödinger equation to the study of atomic stabilization in helium. We found that electron correlation has a negative impact on the atomic stabilization effectiveness in two-electron atoms, in particular for short pulses, but the effect is markedly less than predicted by 1D models. In some cases we observed a complete breakdown of stabilization, whereas in other situations, the correlation only weakened the stabilizing effect. For longer pulses the electron-electron interaction was found to be less important. Finally, we have shown that, even though stabilization in the total ionization yield is less probable in the two-electron system, the stabilizing effect is still significant in the double ionization channel. This suggests that atomic stabilization is most easily observed by monitoring the production of fully stripped ion fragments.

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