Non-sequential double ionization of molecules in a strong laser field

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We consider the final stage of double ionization of O\textsubscript{2} molecules by short linearly polarized laser pulses. The saddles of the effective adiabatic potential energy close to which simultaneous escape of electrons from a molecule takes place are identified. The analysis of the saddles and numerical simulations of the ionization indicate that to observe clear signatures of simultaneous electron escape in double ionization of O\textsubscript{2} molecules stronger and much shorter laser pulses than those used in the recent experiment [E. Eremina, et al, Phys. Rev. Lett. 92, 173001 (2004)] should be applied.

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I. INTRODUCTION

Experimental studies of a non-sequential double (or multiple) ionization is possible thanks the use of high intensity ultrashort-pulse lasers.\textsuperscript{[1, 2]} On the other hand theoretical description of such multi-electron dynamics is far from complete and suggests that interactions between electrons become important in considerations.\textsuperscript{[1, 2, 3, 4]} In this paper we present description of the non-sequential double ionization of molecules based on a classical model for electrons in a combined Coulomb and external field following the approach developed in\textsuperscript{[5, 6, 7, 8, 9]} for the multiple ionization of atoms.

Recent experimental investigations aimed on the nonsequential double ionization of diatomic molecules\textsuperscript{[10, 11, 12, 13]} showed that there are differences between molecular species. For instance, in the case of N\textsubscript{2} it seems that electrons escape with similar momenta along field polarization axis more often than in the case of O\textsubscript{2}\textsuperscript{[13, 14]}. Such a correlation in momenta of escaping electrons is viewed as characteristic feature of the non-sequential double ionization. In our previous publication\textsuperscript{[14]} we have analyzed non-sequential double ionization of molecules and presented numerical results mostly for double ionization of N\textsubscript{2} molecules. Here we complete the analysis by investigating how the distributions change in the case of O\textsubscript{2} molecules for different parameters of the system.

II. RESULTS

In our considerations we assume the re-scattering scenario\textsuperscript{[14, 15, 16]}. Namely, in the first step one electron tunnels out through the Stark saddle and then is returned back to the nuclei. And so at the expense of the energy brought back by the returning electron a highly excited state of a molecule is formed. Finally, such a highly excited compound state decays in several ways through a single, double or multiple ionization. We start our analysis after the formation of the excited complex that is to say we assume that we have an initial state of two highly excited electrons close to the molecular core in the presence of linearly polarized laser field. For short laser pulses molecules have not enough time to change their orientation\textsuperscript{[14, 15, 17]} and thus we presume that the motion of the molecular core is frozen. Hence, the Hamiltonian reads (in atomic units, which are used throughout):

\begin{equation}
H = \frac{p_{i}^{2} + p_{\theta}^{2}}{2} + V, \tag{1}
\end{equation}

where the potential,

\begin{equation}
V = -\sum_{i=1}^{2} \left[ \frac{1}{\sqrt{(x_{i} + \frac{d}{2} \sin \theta)^{2} + y_{i}^{2} + (z_{i} + \frac{d}{2} \cos \theta)^{2}}} \right. \\
+ \frac{1}{\sqrt{(x_{i} - \frac{d}{2} \sin \theta)^{2} + y_{i}^{2} + (z_{i} - \frac{d}{2} \cos \theta)^{2}}} \\
+ \frac{1}{|r_{1} - r_{2}|} + (z_{1} + z_{2})F(t). \tag{2}
\end{equation}

The potential consists of the potential energies associated with interactions of the electrons with the nuclei (the entire structure of the molecular core is approximated by two positively charge nuclei), with each other and with the external field. Origin of coordinate system is placed in the center of mass of the nuclei and without loss of generality it is assumed that the molecule lies in the $x$-$z$-plane. Then $x_{i}$, $y_{i}$ and $z_{i}$ indicate position of the electrons, $d$, the distance between nuclei, and $\theta$, the angle between the molecular axis and the $z$-axis (the polarization axis), respectively. The electric field strength $F(t)$ has an oscillatory component times the envelope from the pulse, namely:

\begin{equation}
F(t) = F \cos(\omega t + \phi) \sin^{2}(\pi t/T_{d}), \tag{3}
\end{equation}

with $F$, $\omega$, $\phi$ and $T_{d}$ the peak amplitude, frequency, initial phase of the field and pulse duration, respectively,

\begin{equation}
T_{d} = \frac{2\pi}{\omega}, \tag{4}
\end{equation}

where $n$ is number of cycles in the pulse.
Within this model the only difference between different diatomic species lies in the distance, \(d\), between the nuclei. In the present publication we restrict ourselves to the oxygen molecule, hence \(d = 2.28\) a.u. in the following.

To identify channels for simultaneous electron escape we use an adiabatic approximation, keeping the field fixed, for the reason that the classical motion of the electrons is fast compared to the field oscillations (later, in the numerical simulations, we use the potential \(U\) without any adiabatic assumption). The channels for the non-sequential ionization correspond to saddles of the potential \(U\). That is, when electrons pass close to a saddle they may leave a molecule simultaneously because one of unstable directions of a saddle allows both electrons to increase distance from a molecule.

![Diagram](image)

**FIG. 1:** Energy of the saddle (top panel) and cross section exponent (bottom panel) as a function of orientation angle, \(\theta\), for \(O_2\) molecule \((d = 2.28\) a.u.). Broken line corresponds to the saddle in the \(yz\)-plane, whereas solid line corresponds to the saddle in the \(xz\)-plane. The field strength is \(F(t) = 0.075\) a.u.

For a general orientation of a molecule with respect to the field polarization axis we have found two saddles for electron escape. The first is situated in a plane defined by the molecular and field axes while the other in the perpendicular plane. A few stable and several unstable directions are revealed by the local stability analysis of the saddles. One unstable direction corresponds to the non-sequential double ionization, while the other unstable directions reflect interactions that will push electrons away from the non-sequential double ionization path leading to the single ionization.

The Lyapunov exponents that characterize the different directions allow one to obtain the cross section behavior for non-sequential double ionization events. Similarly to the double ionization without a field, analyzed many years ago by Wannier \(18, 19, 20, 21\), the competition between the various unstable directions gives rise to an algebraic variation of the cross section with energy close to the threshold, namely,

\[
\sigma(E) \propto (E - V_S)^\alpha,
\]

where \(V_S\) is the saddle energy and the exponent contains the Lyapunov exponents,

\[
\alpha = \sum_i \lambda_i / \lambda_r;
\]

\(\lambda_r\) is the Lyapunov exponent of the unstable direction corresponding to the non-sequential double ionization path, and \(\lambda_i\) are the Lyapunov exponents of all other unstable directions of the saddle \(8, 22\).

In Fig. 1 variations of the parameters of the saddles with the orientation \(\theta\) for the internuclear distance corresponding to \(O_2\) molecule are presented. Fig. 1 shows that the energies and exponents do not change very much with the orientation of the molecule (especially in the case of the saddle located in a plane defined by the field and molecular axes). Thus, we may conclude that from the point of view of the local analysis of the non-sequential ionization, decay of the highly excited complex does not depend strongly on the orientation of the molecule with respect to the field polarization axis.

Thus far we have analyzed the potential within the adiabatic approximation. Now we present results of numerical simulations of decay of a highly excited two electron compound state taking into account full time dependence of the potential \(U\). Before the double ionization escape both electrons pass close to the nuclei where they interact strongly with each other and with the nuclei. Thus we may assume that all memory of the previous motion (i.e. tunneling of the first electron, its evolution in the combined Coulomb and laser fields and the rescattering process) is lost. Then it is plausible to assume that the compound state which decays to a doubly charged molecule can be classically modeled by a statistical distribution for two electrons close to the nuclei. Detailed discussion on numerical calculations and initial conditions is presented in our previous publication \(14\).

In Fig. 2 distributions of the final electron momenta parallel to the field axis for the \(O_2\) molecule oriented along the field axis are presented. Different initial energy \(E\) and different pulse durations are considered. For very short laser pulses \((n = 2\) cycles\) the distributions are localized along the diagonals indicating that the electrons escape predominately by passing close to the saddles analyzed previously. For longer pulses and for energy
FIG. 2: Final distribution of the parallel electron momenta for different initial energy, \( E \) and different pulse length (i.e. number of cycles in the pulse, \( n \)) in the double ionization of the \( \text{O}_2 \) molecule. The field strength is \( F = 0.075 \text{ a.u.} \), the frequency \( \omega = 0.057 \text{ a.u.} \), the internuclear distance \( d = 2.28 \text{ a.u.} \) and the orientation parallel to the field, \( \theta = 0 \). The plots differ in initial energy \( E \) and the number of field cycles \( n \): (a) \( E = -0.05 \text{ a.u.}, n = 2 \); (b) \( E = -0.05 \text{ a.u.}, n = 26 \); (c) \( E = -0.9 \text{ a.u.}, n = 2 \) and (d) \( E = -0.9 \text{ a.u.}, n = 26 \).

\( E = -0.9 \text{ a.u.} \) the distribution drastically changes its character. Namely, the first and fourth quadrants of the panel become strongly populated implying that a number of sequential decays significantly increases. The reason for this is that, after re-scattering when a highly excited two electron complex is created, there are two dominant scenarios within the first half cycle of the field: a non-sequential double escape or a single ionization. During the next cycles, unless the molecule is already doubly ionized, we are left with a singly ionized molecule which may survive to the end of the pulse or the second electron escapes and that corresponds to a sequential double ionization. As the pulse becomes longer, the number of such outcomes increases, even to the point of overwhelming the number of direct, non-sequential double ionization cases. Hence, even though the re-scattering scenario is involved in the double ionization process, for sufficiently long pulses the momentum distributions will not show signatures of the non-sequential electron escape. For the initial energy \( E \) much higher than the minimal energy of a saddle (\( V_S \approx -1.27 \text{ a.u.} \)) the probability of non-sequential ionization is bigger and even if the pulse duration is quite long (e.g. \( n = 26 \) in panel (b) of Fig. 2) the signatures of non-sequential process remain.

Alteration in the orientation of the molecule does not lead to significant changes in the momentum distributions as one can see by comparing Fig. 3 and Fig. 2. For the field amplitude used in the simulations the saddles for non-sequential process are far away from the nuclei and the positions and other parameters of the saddles change only slightly with the vary of orientation \( \theta \) — compare Fig. 1.

Figs. 2-3 should be compared with the corresponding figures for nitrogen molecule presented in Ref. [14] — there we chose energies \( E = -0.3 \text{ a.u.} \) and \( E = -0.6 \text{ a.u.} \) (also the field peak amplitude was slightly smaller, i.e. \( F = 0.07 \text{ a.u.} \)). Comparing the data we may conclude that high initial energy leads to much clearer signatures of simultaneous electron escape (even for long pulses) than low energy. Initial energy is determined in the
can be reached by increase of the amplitude with simultaneous reduction of the pulse duration — very short laser pulses and the field stronger than that used in Ref. 13 should allow for clear observation of simultaneous electron escape in double ionization of O₂ molecules.

Our analysis is restricted to the final stage of the non-sequential double ionization of O₂ molecules, where we choose initial conditions microcanonically. Early stages of the excitation process and the nature of the compound state (i.e. the symmetry of the valence orbital may affect the excitation process) before the final decay towards double ionization may have influence on the initial conditions. That, in turn, may modify the final parallel momenta distributions. This aspect needs further investigation.

III. SUMMARY

We have performed a purely classical analysis of the final stage of the non-sequential double ionization of O₂ molecules in the strong laser field where a molecular core is approximated by two positively charged nuclei and the re-scattering scenario is assumed. Our analysis, together with the previous studies devoted mostly to double ionization of N₂ molecules [14], allow us to draw following conclusions: i) From the point of view of classical analysis, within the considered model, there is no difference between nitrogen and oxygen molecules in the sense that both of them can show signatures of simultaneous double escape. ii) Orientation of the molecule with respect to the field axis does not influence significantly the final momentum distribution for the initial energy range considered in the model. iii) Shorter and stronger laser pulses should enhance signature of simultaneous electron escape that will be visible in the momentum distributions as a more pronounced symmetrical escape of the electrons.

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[1] Super-Intense Laser-Atom Physics, Proceedings of the NATO Advanced Research Workshop, Han-sur-Lesse, Belgium, 1993, edited by B. Piraux, A. L’Huillier, and K. Rzązewski (Plenum Press, New York, 1993).
[2] Super-Intense Laser-Atom Physics, Proceedings of the NATO Advanced Research Workshop, Han-sur-Lesse, Belgium, 2000, edited by B. Piraux and K. Rzązewski (Kluwer Academic Publishers, Dordrecht, 2001).
[3] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Phys. Rev. Lett. 70, 1599 (1993).
[4] B. Yang, K. J. Schafer, B. Walker, K. C. Kulander, P. Agostini, L. F. DiMauro, Phys. Rev. Lett. 71, 3770 (1993).
[5] B. Eckhardt and K. Sacha, Phys. Scr. T90, 185 (2001).
[6] K. Sacha and B. Eckhardt, Phys. Rev. A 63, 043414 (2001).
[7] K. Sacha and B. Eckhardt, Phys. Rev. A 64, 053401 (2001).
[8] B. Eckhardt and K. Sacha, Europhys. Lett. 56, 651 (2001).
[9] K. Sacha and B. Eckhardt, J. Phys. B: At. Mol. Opt. Phys. 36, 3923 (2003).
[10] C. Cornaggia and P. Hering, J. Phys. B: At. Mol. Opt. Phys. 31, L503 (1998).
[11] C. Guo, M. Li, J. P. Nibarger, and G. N. Gibson, Phys. Rev. A 58, R4271 (1998).
[12] C. Guo, M. Li, J. P. Nibarger, and G. N. Gibson, Phys. Rev. A 61, 033413 (2000).
[13] E. Eremina, X. Liu, H. Rottke, W. Sandner, M. G. Schätzel, A. Dreischuh, G. G. Paulus, H. Walther, R. Moshammer and J. Ullrich, Phys. Rev. Lett. 92, 173001 (2004).
[14] J. S. Prauzner-Bechcicki, K. Sacha, B. Eckhardt, and J. Zakrzewski, e-print physics/0405137.
[15] P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).
[16] K. C. Kulander, K. J. Schafer, and J. L. Krause, in Super-Intense Laser-Atom Physics, Proceedings of the NATO Advanced Research Workshop, Han-sur-Lesse, Belgium, 1993, edited by B. Piraux, A. L’Huillier, and K. Rzążewski (Plenum Press, New York, 1993).
[17] K. Miyazaki, T. Shimizu and D. Normand, J. Phys. B: At. Mol. Opt. Phys. 37, 753 (2004).
[18] G. H. Wannier, Phys. Rev. 90, 817 (1953).
[19] R. Peterkop, J. Phys. B: At. Mol. Phys. 4, 513 (1971).
[20] A. R. P. Rau, Phys. Rep. 110, 369 (1984).
[21] J. M. Rost, Phys. Rep. 297, 271 (1998).
[22] J. M. Rost, Physica E 9, 467 (2001).