A general model and toolkit for the ionization of three or more electrons in strongly driven atoms using an effective Coulomb potential for the interaction between bound electrons

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We formulate a three-dimensional semi-classical model to address triple and double ionization in three-electron atoms driven by intense infrared laser pulses. During time propagation, our model fully accounts for the Coulomb singularities, the magnetic field of the laser pulse and for the motion of the nucleus at the same time as for the motion of the three electrons. The framework we develop is general and can account for multi-electron ionization in strongly-driven atoms with more than three electrons. To avoid unphysical autoionization arising in classical models of three or more electrons, we replace the Coulomb potential between pairs of bound electrons with effective Coulomb potentials. The Coulomb forces between electrons that are not both bound are fully accounted for. We develop a set of criteria to determine when electrons become bound during time propagation. We compare ionization spectra obtained with the model developed here and with the Heisenberg model that includes a potential term restricting an electron from closely approaching the core. Such spectra include the sum of the electron momenta along the direction of the laser field as well as the correlated electron momenta. We also compare these results with experimental ones.

I. INTRODUCTION

Multi-electron ionization in atoms and molecules driven by intense infrared laser fields is of fundamental interest since it is mediated by electronic correlation. The theoretical study of correlated multi-electron dynamics in strongly-driven atoms and molecules poses a significant challenge. Indeed, three-dimensional (3D) ab-initio quantum-mechanical methods are mostly limited to double ionization in two-electron atoms [1, 2]. Various quantum mechanical [3, 4] and semi-classical techniques [5–7] that include the Coulomb singularity have been developed to address double ionization. However, for three-electron escape, due to the larger degree of complexity involved, only few theoretical studies exist that have a number of approximations. These studies include classical models with reduced-dimensionality [8] and with soft core Coulomb potentials [9–13], reduced-dimensionality quantum mechanical treatments [14–16] and semi-classical models with Heisenberg potentials [17]. On the experimental front, several studies have addressed multi-electron ionization in strongly-driven Ar and Ne [18–24]. For weak fields, striking angular patterns of three-electron escape and the underlying collision mechanisms were identified with 3D semi-classical models and ab-initio quantum mechanical techniques [25–28].

The main challenge facing quantum mechanical studies of triple ionization in strongly-driven systems is the significant amount of computational resources. This explains the development of reduced dimensionality quantum mechanical models [14–16]. On the other hand, the main difficulty encountered by 3D semi-classical studies of multi-electron ionization that include the Coulomb singularity is unphysical autoionization. Namely, one of the bound electrons can undergo a close encounter with the core and acquire a very negative energy leading to the escape of another bound electron. This is avoided in quantum mechanical treatments of multi-electron systems due to the lower energy bound of an electron. Adding a Heisenberg potential is an approach adopted to exclude unphysical autoionization in 3D semi-classical treatments [29]. This potential amounts to adding a potential barrier that mimics the Heisenberg uncertainty principle and prevents each electron from a close encounter with the core. The addition to the Hamiltonian of an extra momentum and position dependent term results in the momentum of a particle being no longer directly related to the rate of change of its position, \( p \neq m \dot{r} \) [30, 31]. In what follows, we refer to this model as H-model. An advantage of this model is that electronic interactions are accounted for with Coulomb forces at all times during propagation. However, due to the Heisenberg potential, each electron accesses a reduced phase space resulting in a less accurate description of the interaction of each electron with the core. Indeed, in what follows we show that the H-model gives rise to “softer” re-collisions upon the return of an electron to the core.

Here, we take another approach to addressing unphysical autoionization in 3D semi-classical models that include the Coulomb singularity. We develop a 3D semi-classical model that describes the interaction between a pair of bound electrons via an effective Coulomb potential [32]. The interaction between all other pairs of electrons are described with Coulomb forces. This model advances our previous work of triple ionization in strongly-driven \( \mathrm{H}_2\mathrm{He}^+ \), where we switched off the Coulomb force between bound electrons [33]. In the current work, we develop an efficient set of criteria to determine on the fly, i.e. during time-propagation whether an electron is bound or “quasi-free”. Hence, we determine on the fly whether
the interaction between two electrons is described by the Coulomb or the effective Coulomb potential. We refer to this model as ECBB—effective Coulomb potential for bound-bound electrons. We show that the ECBB-model accurately describes three- and two-electron ionization spectra in strongly-driven three-electron atoms.

Our motivation for developing the ECBB-model is the accurate description at all times of the Coulomb interaction of each electron with the core, unlike the H-model. The importance of this interaction has been demonstrated in the finger-like structure in the correlated electron momenta in double ionization of strongly-driven Helium. This structure was predicted theoretically [2], observed experimentally [34, 35] and explained theoretically within a classical framework [7, 36]. On the other hand, the H-model accurately accounts for the interaction between all pairs of electrons with Coulomb forces. The ECBB-model does so for pairs of electrons where at least one electron is “quasi-free”. In the ECBB-model the interaction between two bound electrons is described by effective Coulomb potentials and is thus less accurate. However, bound electrons have a restricted dynamics compared to “quasi-free” electrons. Hence, one can argue that it is more important to accurately describe the interaction of each electron with the core rather than the interaction between bound electrons.

Here, we formulate the ECBB-model and employ the H-model fully accounting for both the motion of the core and all three electrons and for the magnetic field component of the Lorentz force. That is, we formulate both models in the non-dipole approximation. This is unlike previous theoretical studies of strongly-driven atoms. Our formalism is general and can be applied to treat multi-electron ionization in more than three-electron strongly-driven atoms. Here, we employ both models in the context of strongly-driven Ar. We note that our 3D semi-classical model for two-electron atoms has previously yielded very good agreement with experimental observables for double ionization in strongly-driven Ar driven by few-cycles laser pulses [37]. In this latter model of double ionization, we did not need to address unphysical autoionization. Moreover, we discuss in detail the differences of the ECBB- and H-model concerning triple and double ionization observables. Such spectra include the probability distribution of the sum of the electron momenta components along the direction of the laser field and the correlated electron momenta. Also, we compare our results for the sum of the momenta with experimental ones [21, 22]. Finally, we obtain the probability distributions of the angle of escape between two electrons and between an electron and the core.

II. METHOD

In what follows, we describe in detail the formulation of the ECBB-model and the H-model that address multi-electron escape in strongly-driven atoms. The two methods resolve in a different way unphysical autoionization in 3D semi-classical models that fully account for the Coulomb singularity. In both methods, we propagate in time all three electrons and the core. Moreover, we formulate both methods in the non-dipole approximation fully accounting for the magnetic field component of the laser field. The Hamiltonian of a four-body system in the non-dipole approximation is given by

$$H = \sum_{i=1}^{N} \left[ \frac{\tilde{p}_i - Q_i A(r_i, t)}{2m_i} \right]^2 + \sum_{i=1}^{N} \sum_{j=i+1}^{N} \frac{Q_i Q_j}{|r_i - r_j|},$$

where $Q_i$ is the charge, $m_i$ is the mass, $r_i$ is the position vector and $\tilde{p}_i$ is the canonical momentum vector of particle $i$. The mechanical momentum $p_i$ is given by

$$p_i = \tilde{p}_i - Q_i A(r_i, t),$$

where $A(r_i, t)$ is the vector potential and $E(r_i, t) = -\partial A(r_i, t)/\partial t$ is the electric field. Modifying Eq. (1), in the following sections, we formulate the Hamiltonian for the ECBB- and the H-model. For three-electron Ar, the charge of the core is equal to $Q_1 = 3$ a.u. while the mass of the core is equal to $m_1 = 72820.8$ a.u.

A. Global regularisation

In both methods, we perform a global regularisation to avoid any numerical issues arising from the Coulomb singularities. For strongly-driven $\text{H}_2$, we previously used this regularisation scheme to study double and “frustrated” double ionization within the dipole approximation [38] as well as non-dipole effects in non-sequential double ionization [39]. In this scheme, we define the relative position between two particles $i$ and $j$ as

$$q_{ij} = r_i - r_j$$

and

$$\rho_{ij} = \frac{1}{N} \left( \tilde{p}_i - \tilde{p}_j - \frac{m_j - m_i}{M} \langle \rho \rangle \right),$$

where

$$\langle \rho \rangle = \sum_{i=1}^{N} \tilde{p}_i \text{ and } M = \sum_{i=1}^{N} m_i.$$  

The inverse transformation is given by

$$r_i = \frac{1}{M} \sum_{j=i+1}^{N} m_j q_{ij} - \frac{1}{M} \sum_{j=1}^{i-1} m_j q_{ij} + \langle q \rangle.$$
\[ \tilde{p}_i = \sum_{j=i+1}^{N} \rho_{ij} - \sum_{j=1}^{i-1} \rho_{ji} + \frac{m_i}{M} \langle \rho \rangle, \]  \hspace{1cm} (7)

where

\[ \langle q \rangle = \frac{1}{M} \sum_{i=1}^{N} m_i r_i. \]  \hspace{1cm} (8)

Next, we define a fictitious particle \( k \) for each pair of particles \( i, j \) as follows

\[ k(i, j) = (i - 1)N - \frac{i(i + 1)}{2} + j, \]  \hspace{1cm} (9)

with \( j > i \) and the total number of fictitious particles being equal to \( K = \frac{N(N-1)}{2} \). In addition, we define the parameters \( \alpha_{ik} \) and \( \beta_{ik} \), as \( \alpha_{ik} = 1, \beta_{ik} = \frac{m_j}{M} \) and \( \alpha_{ik} = -1, \beta_{ik} = -\frac{m_i}{M} \) when \( k = k(i, j) \), otherwise \( \alpha_{ik} = \beta_{ik} = 0. \) Given the above, Eqs. (6) and (7) take the following simplified form

\[ \tilde{p}_i = \sum_{k=1}^{K} \alpha_{ik} \rho_k + \frac{m_i}{M} \langle \rho \rangle, \]  \hspace{1cm} (10)

and

\[ r_i = \sum_{k=1}^{K} \beta_{ik} q_k + \langle q \rangle. \]  \hspace{1cm} (11)

### B. Heisenberg potential method

#### 1. Description of the model

The Heisenberg potential, originally proposed by Kirschbaum and Wilets in Ref. [29], is given by

\[ V_{H,i} = \frac{\xi^2}{4\alpha\mu r_{i,1}^2} \exp \left\{ \alpha \left[ 1 - \left( \frac{r_{i,1}p_{i,1}}{\xi} \right)^4 \right] \right\}, \]  \hspace{1cm} (12)

where \( r_{i,1} = r_1 - r_i \) is the relative position of each one of the three electrons \( i=2,3,4 \) with respect to the core \( i=1 \), \( p_{i,1} \) is the corresponding relative momentum

\[ p_{i,1} = \frac{m_i p_i - m_1 p_1}{m_1 + m_i}, \]  \hspace{1cm} (13)

and \( \mu = \frac{m_1 m_i}{m_1 + m_i} \) is the reduced mass of the electron-core system. This potential restricts the relative position and momentum of electron \( i \) according to

\[ r_{i,1}p_{i,1} \geq \xi. \]  \hspace{1cm} (14)

Hence, the Heisenberg potential acts as a repulsive potential when the electron is close to the core.

#### 2. Hamilton’s equations of motion

Including the Heisenberg potential for each one of the electron-core pairs, the Hamiltonian is given by

\[ H = \sum_{i=1}^{N} \left[ \frac{\tilde{p}_i - Q_i \mathbf{A}(r_i, t)}{2m_i} \right]^2 + \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} Q_i Q_j |r_i - r_j| + \sum_{i=2}^{N} V_{H,i}. \]  \hspace{1cm} (15)

Substituting Eq. (3) and Eq. (10) in Eq. (15), we obtain the Hamiltonian in regularized coordinates as follows

\[ H = \sum_{k,k'=1}^{K} T_{kk'} \rho_k \rho_{k'} + \langle \rho \rangle^2 \frac{2}{2M} + \sum_{k=1}^{K} U_k \]  \hspace{1cm} (16)

\[ + \sum_{i=2}^{N} Q_i^2 A^2 (r_i, t) - \sum_{i=1}^{N} \frac{Q_i^2}{m_i} \tilde{p}_i \cdot \mathbf{A} (r_i, t) \]  \hspace{1cm} (16)

\[ + \sum_{i=1}^{N-1} \xi^2 \frac{\alpha m_i}{4\alpha \mu r_{i,1}^2} \exp \left\{ \alpha \left[ 1 - \left( \frac{r_{i,1}p_{i,1}}{\xi} \right)^4 \right] \right\}, \]  \hspace{1cm} (16)

where \( \tilde{p}, \mathbf{r} \) are expressed in terms of \( \rho \) and \( q \) via Eqs. (10) and (11). In Eq. (16), \( U_k \) is equal to \( Q_i Q_j \). Using Eq. (16), we obtain Hamilton’s equations of motion
\[
\frac{d\mathbf{q}_k}{dt} = 2 \sum_{k'=1}^{K} T_{kk'} \rho_{k'} - \sum_{i=1}^{N} \frac{Q_i}{m_i} \alpha_{ik} \mathbf{A}(\mathbf{r}_i, t) - \sum_{i=1}^{N-1} \frac{(q_{i+1,i+1})^2}{\mu \xi^2} \frac{m_{i+1} \alpha_{ik} - m_i \alpha_{i+1,k}}{m_i + m_{i+1}} \exp \left\{ \alpha \left[ 1 - \left( \frac{q_{i+1,i+1}}{\xi} \right)^4 \right] \right\} \mathbf{p}_{i+1,i+1}
\]

\[
\frac{d(\mathbf{q})}{dt} = \frac{1}{M}(\mathbf{p}) - \sum_{i=1}^{N} \frac{Q_i}{M} \mathbf{A}(\mathbf{r}_i, t)
\]

\[
\frac{d\rho_k}{dt} = \frac{U_k \mathbf{q}_k}{q_k^2} + \sum_{i=1}^{N} \frac{Q_i}{m_i} \left[ \frac{\hat{p}_i - Q_i \mathbf{A}(\mathbf{r}_i, t)}{\partial \mathbf{q}_k} \right] \cdot \frac{\partial \mathbf{A}(\mathbf{r}_i, t)}{\partial \mathbf{q}_k} + \sum_{i=1}^{N-1} \frac{(q_{i+1,i+1})^2}{\mu (m_i + m_{i+1}) \xi^2} \exp \left\{ \alpha \left[ 1 - \left( \frac{q_{i+1,i+1}}{\xi} \right)^4 \right] \right\} \mathbf{p}_{i+1,i+1} \cdot \left[ m_{i+1} Q_i \frac{\partial \mathbf{A}(\mathbf{r}_i, t)}{\partial \mathbf{q}_i} - m_i Q_{i+1} \frac{\partial \mathbf{A}(\mathbf{r}_{i+1}, t)}{\partial \mathbf{q}_{i+1}} \right]
\]

\[
\frac{d(\rho)}{dt} = \sum_{i=1}^{N} \frac{Q_i}{m_i} \left[ \frac{\hat{p}_i - Q_i \mathbf{A}(\mathbf{r}_i, t)}{\partial \mathbf{q}} \right] \cdot \frac{\partial \mathbf{A}(\mathbf{r}_i, t)}{\partial \mathbf{q}} - \sum_{i=1}^{N-1} \frac{(q_{i+1,i+1})^2}{\mu (m_i + m_{i+1}) \xi^2} \exp \left\{ \alpha \left[ 1 - \left( \frac{q_{i+1,i+1}}{\xi} \right)^4 \right] \right\} \mathbf{p}_{i+1,i+1} \cdot \left[ m_{i+1} Q_i \frac{\partial \mathbf{A}(\mathbf{r}_i, t)}{\partial \mathbf{q}} - m_i Q_{i+1} \frac{\partial \mathbf{A}(\mathbf{r}_{i+1}, t)}{\partial \mathbf{q}} \right].
\]

(17)

3. Propagation technique

To integrate the equations of motion in Eq. (17), we use a leapfrog technique \cite{40, 41} jointly with the Bulirsch-Stoer method \cite{42, 43}. We have previously developed this leapfrog technique to study non-dipole effects in non-sequential double ionization in strongly driven H\textsubscript{2} \cite{39}. This leapfrog technique allows to solve Hamilton’s equations when the derivative of the position and the momentum depends on the quantities themselves. It is an extension of the leapfrog technique we employed for strongly driven two-electron molecules in the dipole approximation \cite{38}. In the latter case the derivative of the position and the momenta do not depend on themselves.

C. Effective Coulomb potential method

1. Derivation of the effective Coulomb potential

In what follows, we formulate a method that avoids unphysical autoionization between two bound electrons. To do so, we describe the interaction between two bound electrons with an effective Coulomb potential. However, we describe the interaction between a “quasi-free” and a bound electron as well as between two quasi-free electrons with the full Coulomb potential. In the next subsection, we define the time when an electron transitions from bound to quasi-free and from quasi-free to bound.

The effective Coulomb potential that electron \( i \) experiences due to the charge \( \zeta_j \) of electron \( j \), denoted by \( V_{\text{eff}}(\zeta_j, |\mathbf{r}_1 - \mathbf{r}_i|) \), is derived as follows \cite{32}. We approximate the wavefunction of a bound electron \( j \) with a hydrogenic wavefunction

\[
\psi(\zeta_j, |\mathbf{r}_1 - \mathbf{r}_i|) = \left( \frac{\zeta_j}{\pi} \right)^{1/2} e^{-\zeta_j |\mathbf{r}_1 - \mathbf{r}_i|},
\]

(18)

where the parameter \( \zeta_j \) is later defined in Eq. (21). The electric charge contained within a sphere of radius \( r \) from the core is given by

\[
Q(\zeta_j, r) = -\int \int \int |\psi(\zeta_j, \mathbf{r})|^2 dV,
\]

(19)

where \( dV \) is the volume element in spherical coordinates.

Using Gauss’s law, we find that the effective Coulomb potential that an electron \( i \) experiences at a distance \( |\mathbf{r}_1 - \mathbf{r}_i| \) from the core due to the charge distribution of electron \( j \) is equal to (see Appendix A)

\[
V_{\text{eff}}(\zeta_j, |\mathbf{r}_1 - \mathbf{r}_i|) = 1 - \frac{1 + \zeta_j |\mathbf{r}_1 - \mathbf{r}_i| e^{-2\zeta_j |\mathbf{r}_1 - \mathbf{r}_i|}}{|\mathbf{r}_1 - \mathbf{r}_i|}.
\]

(20)

\( V_{\text{eff}}(\zeta_j, |\mathbf{r}_1 - \mathbf{r}_i|) \) is a repulsive potential which has limiting values of \( \zeta_j \) when \( |\mathbf{r}_1 - \mathbf{r}_i| = 0 \) and 0 when \( |\mathbf{r}_1 - \mathbf{r}_i| \to \infty \). If the effective charge \( \zeta_j(t) \) is zero then the effective potentials \( V_{\text{eff}}(\zeta_j, |\mathbf{r}_1 - \mathbf{r}_i|) \) is zero. The effective charge \( \zeta_j(t) \), at any time during the propagation of the four-body system, is proportional to the energy \( E_j(t) \) of electron \( j \), assuming electron \( j \) is bound with an energy greater than a lower limit. We set this lower limit to be equal to the ground state energy \( E_{1s} \) of a hydrogenic atom with core charge equal to \( Q_1 \), i.e. \( E_{1s} = \frac{Q_1^2}{2} \). Moreover, when the energy of electron \( j \), \( E_j(t) \), is greater than zero, we set \( \zeta_j(t) \) equal to zero, while if the energy is less than the lower limit \( E_{1s} \) we set \( \zeta_j(t) \) equal to \( Q_1 \). Hence, we define
\( \zeta_j(t) \) as follows

\[
\zeta_j(t) = \begin{cases} 
  Q_1 & \mathcal{E}_j(t) \leq \mathcal{E}_{1s} \\
  (Q_1/\mathcal{E}_{1s}) \mathcal{E}_j(t) & \mathcal{E}_{1s} < \mathcal{E}_j(t) < 0 \\
  0 & \mathcal{E}_j(t) \geq 0,
\end{cases}
\]  

(21)

where the energy \( \mathcal{E}_j(t) \) of electron \( j \) is given by

\[
\mathcal{E}_j(t) = \frac{(\hat{p}_j - Q_j \mathbf{A}(r_j,t))^2}{2m_j} + \frac{Q_j Q_1}{|r_1 - r_j|} - Q_j q_j \cdot \mathbf{E}(r_j,t)
\]

(22)

\[
+ \sum_{i \neq j} c_{i,j}(t) V_{\text{eff}}(\zeta_i, |r_1 - r_j|) 
\]

The functions \( c_{i,j}(t) \) determine whether the full Coulomb interaction or the effective \( V_{\text{eff}}(\zeta_i, |r_1 - r_j|) \) and \( V_{\text{eff}}(\zeta_j, |r_1 - r_j|) \) potential interactions are on or off for any pair of electrons \( i \) and \( j \) during the time propagation. Specifically, the limiting values of \( c_{i,j}(t) \) are zero and one. The value zero corresponds to the full Coulomb potential being turned on while the effective Coulomb potentials are off. This occurs for a pair of electrons \( i \) and \( j \) where either \( i \) or \( j \) is quasi-free. The value one corresponds to the effective Coulomb potentials \( V_{\text{eff}}(\zeta_i, |r_1 - r_j|) \) and \( V_{\text{eff}}(\zeta_j, |r_1 - r_j|) \) being turned on while the full Coulomb potential is off. This occurs for bound electrons \( i \) and \( j \). For simplicity, we choose \( c_{i,j}(t) \) to change linearly with time between the limiting values zero and one. Hence, \( c_{i,j}(t) \) is defined as follows

\[
c_{i,j}(t) = \begin{cases} 
  0 & c(t) \leq 0 \\
  c(t) & 0 < c(t) < 1 \\
  1 & c(t) \geq 1,
\end{cases}
\]

(23)

where \( c(t) = \beta(t - t_{s}^{ij}) + c_0 \), and \( c_0 \) is the value of \( c_{i,j}(t) \) just before a switch at time \( t_{s}^{ij} \). A switch at time \( t_{s}^{ij} \) occurs if the interaction between electrons \( i, j \) changes from full Coulomb to effective Coulomb potential or vice versa. At the start of the propagation at time \( t_0 \), \( t_{s}^{ij} \) is equal to \( t_0 \) and \( c_0 \) is one for pairs of electrons that are bound and zero otherwise. To allow for a smooth switch on or switch off of the effective Coulomb potential we choose \( \beta \) equal to \( \pm 0.1 \); plus corresponds to a switch on and minus to a switch off of the effective Coulomb potential.

2. Derivation of the time derivative of the effective charges

Including the effective Coulomb potentials, the Hamiltonian of the four-body system is given by

\[
H = \sum_{i=1}^{N} \frac{\hat{p}_i - Q_i \mathbf{A}(r_i,t)^2}{2m_i} + \sum_{i=2}^{N} \frac{Q_i Q_j}{|r_i - r_j|} 
\]

(24)

\[
+ \sum_{i=2}^{N-1} \sum_{j=i+1}^{N} \frac{(1 - c_{i,j}(t)) Q_i Q_j}{|r_i - r_j|} 
\]

\[
+ \sum_{i=2}^{N-1} \sum_{j=i+1}^{N} c_{i,j}(t) \left[V_{\text{eff}}(\zeta_i, |r_1 - r_i|) + V_{\text{eff}}(\zeta_j, |r_1 - r_j|) \right] 
\]

The dipole term \( -Q_j q_j \cdot \mathbf{E}(r_j,t) \) of Eq. (22) involving the electric field does not appear in the Hamiltonian (24). There is no contradiction. Indeed, the gauge-invariant energy of a particle does not always coincide with the gauge-dependent Hamiltonian, as discussed in Ref. [44, 45]. We note that the Hamiltonian in Eq. (24) depends not only on positions, momenta and time but also on the effective charges. Since the effective charge \( \zeta_j \) is proportional to the energy \( \mathcal{E}_j(t) \), see Eq. (21), it follows that we must obtain the derivative with time of \( \mathcal{E}_j(t) \). We note that this is necessary at any time during propagation if at least two electrons are bound. To do so, we apply the chain rule in Eq. (22) and obtain

\[
\dot{\mathcal{E}}_j(t) = \frac{\partial \mathcal{E}_j(t)}{\partial r_j} \cdot \dot{r}_j + \frac{\partial \mathcal{E}_j(t)}{\partial \hat{p}_j} \cdot \dot{\hat{p}}_j + \frac{\partial \mathcal{E}_j(t)}{\partial r_1} \cdot \dot{r}_1 + \sum_{i=2}^{N} \frac{\partial \mathcal{E}_j(t)}{\partial \zeta_i} \dot{\zeta}_i + \frac{\partial \mathcal{E}_j(t)}{\partial \zeta_1} \dot{\zeta}_1 + \frac{\partial \mathcal{E}_j(t)}{\partial t} = \frac{\partial \mathcal{E}_j(t) - H}{\partial r_j} \cdot \dot{r}_j + \frac{\partial \mathcal{E}_j(t)}{\partial r_1} \cdot \dot{r}_1 + \sum_{i=2}^{N} \frac{\partial \mathcal{E}_j(t)}{\partial \zeta_i} \dot{\zeta}_i + \frac{\partial \mathcal{E}_j(t)}{\partial t} 
\]

(25)
\[ + \sum_{i,j=1, i \neq j}^{N} \left[ c_{i,j}(t) \frac{\partial V_{\text{eff}}(\zeta_i, |r_i - r_j|)}{\partial \zeta_i} \dot{\zeta}_i + c_{i,j}(t) V_{\text{eff}}(\zeta_i, |r_i - r_j|) \right] \dot{r}_j = -Q_j \ddot{E}(r_j, t), \]

where we use \( \dot{r}_j = \frac{\partial E_j(t)}{\partial \rho} \) and \( \ddot{r}_j = \frac{\partial H}{\partial r_j} \). The above expression can be finally written as

\[ \dot{E}_j(t) = f_j + \sum_{i=2}^{N} c_{i,j}(t) \frac{\partial V_{\text{eff}}(\zeta_i, |r_i - r_j|)}{\partial \zeta_i} \dot{\zeta}_i, \quad (26) \]

where \( f_j(r, p, t, \psi) \) are all the terms in Eq. (25) that do not depend on \( \dot{\zeta}_i(t) \). The time derivative of \( \zeta_i(t) \) is given by

\[ \dot{\zeta}_i = \begin{cases} 0 & \text{if } \zeta_i(t) \leq \zeta_{1a} \\ -\frac{Q_{i}}{m_{i}} \dot{E}_i(t) & \text{if } \zeta_i(t) < \zeta_{1a} \leq 0 \\ 0 & \text{if } \zeta_i(t) \geq 0, \end{cases} \quad (27) \]

We obtain an equation similar to Eq. (26) for each electron. Hence, at any time during propagation, we solve a system of equations to obtain the derivative in time of the energies of each electron. As a result, we express each \( \dot{E} \) as a function of \( (r, p, t, \psi) \) with no dependence on the derivatives of the energies.

3. Hamilton’s equations of motion

Substituting Eqs. (3) and (10) in Eq. (24), we find the Hamiltonian in regularized coordinates to be given by

\[ H = \sum_{k,k'=1}^{K} T_{kk'} \rho_k \dot{p}_{k'} + \frac{(\rho)^2}{2M} + \sum_{k=1}^{K} \left[ 1 - c_k(t) \right] U_k \frac{q_k}{q_{\text{eff}}} \]

\[ + \sum_{i=1}^{N} \frac{q_i^2}{2m_i} A_i^2(r_i, t) - \sum_{i=1}^{N} \frac{Q_{i}}{m_{i}} \dot{\rho}_i \cdot A_i(r_i, t) \]

\[ + \sum_{k=1}^{K} c_{k}(t) V_k, \quad (28) \]

where

\[ V_k = V_{\text{eff}}(\zeta_i, |r_1 - r_i|) + V_{\text{eff}}(\zeta_i, |r_1 - r_i|), \quad (29) \]

and \( \dot{\rho}, \dot{r}, \) are expressed in terms of \( \rho \) and \( q \) via Eqs. (10) and (11). Moreover, for \( k=1,2,3 \) \( q_k \) corresponds to the relative distance between each one of the three electrons and the core. Since, the Coulomb force between each of the three electrons and the core is always on, we set \( c_k(t) = 0 \), for \( k=1,2,3 \). Using Eq. (28), we find that Hamilton’s equations of motion are given by

\[ \frac{dq_k}{dt} = 2 \sum_{k'=1}^{K} T_{kk'} \rho_{k'} - \sum_{i=1}^{N} \frac{Q_{i}}{m_{i}} \alpha_{ik} \vec{A}_i(r_i, t) \]

\[ \frac{d(\rho)}{dt} = \frac{1}{M}(\rho) - \sum_{i=1}^{N} \frac{Q_{i}}{m_{i}} \vec{A}_i(r_i, t) \]

\[ \frac{dp_k}{dt} = \left[ 1 - c_k(t) \right] U_k \frac{q_k}{q_{\text{eff}}} - \sum_{k'=1}^{K} c_{k'}(t) \frac{\partial V_{k'}}{\partial q_k} \]

\[ + \sum_{i=1}^{N} \frac{Q_{i}}{m_{i}} [\dot{\rho}_i - Q_{i} \vec{A}_i(r_i, t)] \cdot \frac{\partial \vec{A}_i(r_i, t)}{\partial q_k}, \quad (30) \]

\[ \frac{d(\rho)}{dt} = \sum_{i=1}^{N} \frac{Q_{i}}{m_{i}} [\dot{\rho}_i - Q_{i} \vec{A}_i(r_i, t)] \cdot \frac{\partial \vec{A}_i(r_i, t)}{\partial (q)}, \]

where

\[ \frac{\partial V'_{k}(i,j)}{\partial q_k} = \frac{\partial V'_{k}}{\partial q_k} \delta_{k,k_{(1,i)}} + \frac{\partial V'_{k}}{\partial q_k} \delta_{k,k_{(1,j)}}, \quad (31) \]

where \( \dot{\rho}, \dot{r} \) are expressed in terms of \( \rho \) and \( q \) via Eqs. (10) and (11). From Eqs. (30) and (31) it follows that the term \( \sum_{k'=1}^{K} c_{k'}(t) \frac{\partial V_{k'}}{\partial q_k} \) is non zero for \( k = 1,2,3 \) and has the following form

\[ \sum_{k'=1}^{K} c_{k'}(t) \frac{\partial V_{k'}}{\partial q_1} = c_{i}(t) \frac{\partial V_{\text{eff}}(\zeta_3, |r_1 - r_2|)}{\partial q_1} + c_{5}(t) \frac{\partial V_{\text{eff}}(\zeta_3, |r_1 - r_2|)}{\partial q_1} \]

\[ = c_{4}(t) \frac{1 + [1 + 2 \zeta_{q_1}(1 + \zeta_{q_1})] e^{-2 \zeta_{q_1}}}{q_1^2} q_1 + c_{5}(t) \frac{1 + [1 + 2 \zeta_{q_1}(1 + \zeta_{q_1})] e^{-2 \zeta_{q_1}}}{q_1^2} q_1 \]

\[ \sum_{k'=1}^{K} c_{k'}(t) \frac{\partial V_{k'}}{\partial q_2} = c_{1}(t) \frac{\partial V_{\text{eff}}(\zeta_2, |r_1 - r_3|)}{\partial q_2} + c_{6}(t) \frac{\partial V_{\text{eff}}(\zeta_2, |r_1 - r_3|)}{\partial q_2} \]

\[ = c_{4}(t) \frac{1 + [1 + 2 \zeta_{q_2}(1 + \zeta_{q_2})] e^{-2 \zeta_{q_2}}}{q_2^2} q_2 + c_{6}(t) \frac{1 + [1 + 2 \zeta_{q_2}(1 + \zeta_{q_2})] e^{-2 \zeta_{q_2}}}{q_2^2} q_2 \]

\[ \sum_{k'=1}^{K} c_{k'}(t) \frac{\partial V_{k'}}{\partial q_3} = c_{2}(t) \frac{\partial V_{\text{eff}}(\zeta_1, |r_2 - r_3|)}{\partial q_3} + c_{7}(t) \frac{\partial V_{\text{eff}}(\zeta_1, |r_2 - r_3|)}{\partial q_3} \]

\[ = c_{4}(t) \frac{1 + [1 + 2 \zeta_{q_2}(1 + \zeta_{q_2})] e^{-2 \zeta_{q_2}}}{q_2^2} q_2 + c_{7}(t) \frac{1 + [1 + 2 \zeta_{q_2}(1 + \zeta_{q_2})] e^{-2 \zeta_{q_2}}}{q_2^2} q_2 \]
In addition to Eq. (30), we have three more equations for $\mathcal{E}(q, p, t, \epsilon)$.

### 4. Propagation technique

In our formulation, we fully account for the Coulomb singularities. Hence, an electron can approach infinitely close to the nucleus during time propagation. To ensure the accurate numerical treatment of the N-body problem in the laser field, we perform a global regularisation. This regularisation was introduced in the context of the gravitational N-body problem [46]. Here, we integrate the equations of motion using a leapfrog technique [40, 41] jointly with the Bulirsch-Stoer method [42, 43]. This leapfrog technique allows integration of Hamilton’s equation when the derivatives of the positions and the momenta depend on the quantities themselves. We previously employed this technique in our studies of non-dipole effects in non-sequential double ionization of strongly driven H$_2$ [39]. The difference between the leapfrog technique employed in this work and the one previously employed in [39] is that the former is more involved. Indeed, in the current leapfrog technique we also need to propagate in time the energies $\mathcal{E}(t)$, see Eq. (25). The steps involved in this leapfrog technique are as follows.

First, we perform a time transformation $t \rightarrow s$, where

$$ds = \Omega(q)dt,$$

with $\Omega(q)$ an arbitrary positive function of $q$. We select the function

$$\Omega(q) = \sum_{k=1}^{K} \frac{1}{|q_k|},$$

which forces the time step to decrease when two particles undergo a close encounter and to increase when all particles are far away from each other. The equations of motion now take the following form

\[
\begin{align*}
q' &= \dot{q}(q, p, t) / \Omega(q) \\
p' &= \dot{p}(q, p, t, \mathcal{E}) / \Omega(q) \\
t' &= 1 / \Omega(q) \\
\mathcal{E}' &= \dot{\mathcal{E}}(q, p, t, \mathcal{E}) / \Omega(q),
\end{align*}
\]

with prime denoting the derivative with respect to the new variable $s$. The integration is based on the leapfrog technique described in [39] that introduces four auxiliary variables, two vectors $W^q$, $W^p$ and two scalars $W^t$, $W^\mathcal{E}$. As a result, an extended system is obtained where the derivatives of the position, the momenta and the energies no longer depend on the quantities themselves. The extended equations are given by

$$\begin{align*}
q' &= \dot{q}(W^q, p, W^t) / \Omega(W^q) \\
p' &= \dot{p}(W^q, p, W^t, \mathcal{E}) / \Omega(W^q) \\
t' &= 1 / \Omega(W^q) \\
\mathcal{E}' &= \dot{\mathcal{E}}(W^q, p, W^t, \mathcal{E}) / \Omega(W^q),
\end{align*}$$

and

$$\begin{align*}
W^q' &= \dot{q}(q, W^p, t) / \Omega(q) \\
p' &= \dot{p}(q, W^p, t, W^\mathcal{E}) / \Omega(q) \\
t' &= 1 / \Omega(q) \\
\mathcal{E}' &= \dot{\mathcal{E}}(q, W^p, t, W^\mathcal{E}) / \Omega(q).
\end{align*}$$

We propagate for a time step, by propagating for half a step each quadruplet of variables $(q, W^p, t, W^\mathcal{E})$ and $(W^q, p, W^t, \mathcal{E})$ in an alternating way, see the leapfrog algorithm described in Appendix B. Moreover, to achieve better accuracy, we incorporate the leapfrog method in the Bulirsch-Stoer extrapolation scheme [42, 43]. In this scheme, a propagation over a step $H$ is split into $n$ sub steps of size $h = H / n$. We use the leapfrog method to propagate over each sub step. In Fig. 1, we offer a schematic illustration of the propagation during a time sub step of size $h$. The detailed algorithm is described in the Appendix B. This process is repeated with increasing number of sub steps, i.e. $n \rightarrow \infty$, until an extrapolation with a satisfactory error is achieved.
5. Definition of quasi-free and bound electron

In the ECBB-model the interaction between a pair of electrons where at least one is quasi-free is described with Coulomb forces. The interaction between bound electrons is described with effective Coulomb potentials. Hence, we need to define during time propagation, i.e. on the fly, if an electron is quasi-free or bound. At the start of propagation, the electron that tunnel-ionises (electron 2) is considered quasi-free and the other two (electrons 3 and 4) are bound. We denote the core as particle 1.

At times \( t > t_0 \), a quasi-free electron \( i \) transitions to bound if the following conditions are satisfied: (i) the potential of electron \( i \) with the core, \( V_{i,c} \), is larger than a threshold value, i.e. \( V_{i,c} > V_{\text{min}} \) at \( t_1 \), and \( V_{i,c} \) is continuously increasing, i.e. \( \frac{dV_{i,c}(t)}{dt} > \frac{dV_{i,c}(t_{n-5})}{dt} \) for five times \( t_n \) which are five time steps apart with the first one being at time \( t_1 \), see Fig. 2(a); (ii) the position of electron \( i \) along the electric field, i.e. \( z \) axis here, has at least two extrema of the same kind, i.e. two maxima or two minima, in a time interval less than half a period of the laser field. We start checking if condition (ii) is satisfied at time \( t_2 \) when electron \( i \) has the closest approach to the core, i.e. \( V_{i,c} \) is maximum. We stop checking whether condition (ii) is satisfied at time \( t_3 \) when \( V_{i,c} \) is smaller than the threshold value \( V_{\text{min}} \) and \( V_{i,c} \) is continuously decreasing, i.e. \( \frac{dV_{i,c}(t_{n})}{dt} < \frac{dV_{i,c}(t_{n-5})}{dt} \) for five times \( t_n \) which are five time steps apart with the last one being at time \( t_3 \), see Fig. 2(a). In the current study, we set \( V_{\text{min}} \) equal to \( 3/15 \) which is equal to 0.2 a.u. We find that our results remain almost the same for a range of values of \( V_{\text{min}} \).

Also, at the end of the laser pulse, we check whether a quasi-free electron has positive or negative compensated energy \([39]\). If the latter occurs, we consider the electron to be bound. Accounting for the effective Coulomb potential, the compensated energy of electron \( i \) is given by

\[
\varepsilon_i^{\text{comp}}(t) = \frac{\mathbf{p}_i^2}{2m_i} + \frac{Q_i Q_j}{|r_i - r_j|} + \sum_{j=2}^{N} c_{ij}(t) V_{\text{eff}}(\zeta_j, |r_i - r_j|).
\]

\[(35)\]

A bound electron transitions to quasi-free at time \( t > t_0 \) if either one of the following two conditions is satisfied: (i) at time \( t \) the compensated energy of electron \( i \) converges to a positive value; (ii) at times \( t = t_3 \), \( V_{i,c} \) is smaller than the threshold value \( V_{\text{min}} \) and \( V_{i,c} \) is continuously decreasing, i.e. \( \frac{dV_{i,c}(t_{n})}{dt} < \frac{dV_{i,c}(t_{n-5})}{dt} \) for five \( t_n \) which are five time steps apart, the last one being at \( t_3 \).

We illustrate the above criteria in Fig. 2(b). We denote the times \( t_1, t_2 \) and \( t_3 \) with red, grey and blue vertical dashed lines, respectively. In the left column, we plot the position \( r_z \) and the potential \( V_{i,c} \) of a quasi-free electron as it transitions to bound. In the right column, we plot the position \( r_z \), the potential \( V_{i,c} \) and the compensated energy of a bound electron as it transitions to quasi-free.

The black dashed line denotes the time when the compensated energy converges and the electron transitions from bound to quasi-free. For this specific trajectory, the compensated energy converges prior to \( t_3 \) and hence electron \( i \) transitions from bound to quasi-free at \( t < t_3 \).

FIG. 2. Schematic illustration of the criteria to determine when a quasi-free electron becomes bound (left column) and when a bound electron becomes quasi-free (right column).

We note that the criteria for the convergence of the compensated energy and the number of extrema in the position of the electron along the laser field have been used to determine whether an electron is quasi-free or bound in our previous work on strongly-driven three electron triatomic molecules \([33]\). However, the criteria presented above are considerably refined compared to the
ones in Ref. [33], allowing for the full Coulomb forces to be turned on for a longer time interval. Moreover, in the ECBB-model we account for the interaction between bound electrons with effective Coulomb potentials, while in Ref. [33] this interaction was set equal to zero.

D. Initial conditions

1. Tunnel-ionizing electron

In both methods, electron 2 tunnel-ionizes at time $t_0$ through the field-lowered Coulomb-barrier with a rate that is described by the quantum mechanical Ammosov-Delone-Krainov (ADK) formula [48, 49]. To obtain the ADK rate, we use the value of the energy needed to ionize one electron from Ar, i.e. we use $I_{P1} = 0.579$ a.u. We find $t_0$, using importance sampling [50] in the time interval $[-2\tau, 2\tau]$ where the electric field is non-zero; $\tau$ is the full width at half maximum of the pulse duration in intensity. The importance sampling distribution is given by the ADK ionization rate. The exit point of electron 2 is along the direction of the laser field and is computed using parabolic coordinates [51]. The momentum of electron 2 is taken to be equal to zero along the laser field. The transverse momentum is given by a Gaussian distribution which represents the Gaussian-shaped filter with an intensity-dependent width arising from standard tunneling theory [49, 52, 53].

2. Position and momentum distributions of the bound electrons in the H-model

In the Heisenberg potential, see Eq. (12), for a given $\alpha$, we find the value of $\xi$ that ensures that the minimum of the one-electron Hamiltonian

$$H_i = \frac{p_i^2}{2m_i} + \frac{Q_i Q_{i-1}}{|r_i - r_{i-1}|} + \frac{\xi^2}{4\alpha \mu r_{i-1}^2} \exp \left\{ \alpha \left[ 1 - \frac{r_{i-1}|p_i|}{\xi} \right]^4 \right\}$$

(36)
corresponds to the third ionization potential of Ar ($I_{P3} = 1.497$ a.u.) [54-56]. To minimize Eq. (36) with respect to the relative distance $r_{i-1}$, we start from the lower limit of the constraint

$$r_{i-1}|p_i| = \xi \Rightarrow p_i = \xi/r_{i-1}$$

(37)

Since the mass of the core $m_1 \gg m_i$ it follows that $p_i \approx p_1$. Hence, Eq. (37) can be written as $p_i = \xi/r_{i-1}$ and substituting in Eq. (36) we obtain

$$H_i = \frac{\xi^2}{2m_1 r_{i-1}^2} + \frac{Q_i Q_{i-1}}{r_{i-1}} + \frac{\xi^2}{4\alpha \mu r_{i-1}^2}.$$

(38)

The minimum of Eq. (38) with respect to $r_{i-1}$, occurs at

$$r_{i-1}^{\text{min}} = -\frac{2\alpha \mu + m_i}{2\alpha \mu m_i Q_i Q_{i-1}} \xi^2,$$

(39)

and the energy is given by

$$H_i^{\text{min}} = -\frac{\alpha \mu m_i (Q_i Q_{i-1})^2}{(m_1 + 2\alpha \mu) \xi^2}.$$  

(40)

Setting this energy equal to $I_{P3}$, we find $\xi = 1.55$ a.u for $\alpha = 2$ and $\xi = 1.63$ a.u. for $\alpha = 4$. Hence, for $\alpha = 2$ the electrons access a larger phase space during the time propagation.

To find the initial position and momentum vectors of the two initially bound electrons at time $t_0$, we apply a trial and error method similar to the one proposed by Cohen [31]. First, we randomly sample the magnitude of the position and the momentum vector for each electron in the intervals $[0, r_{\text{max}}]$, $[0, p_{\text{max}}]$. We find that it is sufficient to consider $r_{\text{max}} = 3$ a.u. and $p_{\text{max}} = 3$ a.u. The $\theta, \phi$ polar and azimuthal angles of the position and the momentum of electrons 3 and 4 are obtained as uniform random numbers of $\cos \theta$ in the interval $[-1,1]$ and $\phi$ in the interval $[0,2\pi]$. Using the position and momenta of electrons 3 and 4, we determine the total energy of the two electrons in the absence of the electric field

$$H_{3,4} = \sum_{i=3}^{4} \frac{p_i^2}{2} + \frac{Q_i Q_{i-1}}{|r_i - r_{i-1}|} + \sum_{i=3}^{4} V_{H,i} + \frac{Q_i Q_{i-1}}{|r_i - r_{i-1}|}.$$  

(41)

If the energy $H_{3,4}$ is within 1% of the binding energy $I_{P3} + I_{P4}$, we accept the initial conditions of electron 3 and 4, otherwise we reject them. For Ar, the energy to ionize a second electron is $I_{P2} = 1.015$ a.u. Using the above procedure, we plot in Fig. (3), the probability distribution of the initial position and momentum of electrons 3 and 4 as well as of the Heisenberg potential.

![FIG. 3. Probability distribution of r (a) and p (b) for each of the electrons 3, 4 as well as the Heisenberg potential $V_H$ (c) at time $t_0$, for $\alpha = 2$.](image)

3. Position and momentum distributions of the bound electrons in the ECBB-model

In the ECBB-model, we obtain the initial position and momentum of electron 4 at time $t_0$ using a microcanoni-
classical distribution with an energy
\[ E_4(t_0) = \frac{p_i^2}{2m_i} + \frac{Q_i Q_j}{|r_1 - r_i|} + V_{\text{eff}}(\zeta_3, |r_1 - r_j|), \] (42)
and similarly for electron 3. We take the energy
\[ E_3(t_0) = E_4(t_0) = -Ip_2 \] and using Eq. (21) we find that
\[ \zeta_3(0) = \zeta_4(0) = -(Q_3/E_{34}) E_4(t_0). \] The reason we set the initial energy of each electron equal to \(-Ip_2\) is that \(E_4(t_0)\) and \(E_3(t_0)\) include the interaction with the other electron via \(V_{\text{eff}}\). Hence, \(E_3(t_0)\) and \(E_4(t_0)\) correspond to the energy needed to remove an electron from \(Ar^+\). Using the above defined microcanonical distribution, we obtain the initial position and momentum of each bound electron [57]. In Fig. (4), we plot the probability distribution for the initial position and momentum of electrons 3 and 4 as well as of the \(V_{\text{eff}}\).

FIG. 4. Probability distribution of \(r\) (a) and \(p\) (b) for each of the electrons 3, 4 as well as the effective Coulomb potential \(V_{\text{eff}}\) (c) at time \(t_0\).

### III. RESULTS

In what follows, we compare observables for triple ionization (TI) and double ionization (DI) obtained with the ECBB-model and the H-model. If available, we also compare these observables with experimental results [21, 22]. In our formulation both the ECBB-model and the H-model fully account for non-dipole effects and treat the motion of the electrons and the core on an equal footing.

Here, we employ a vector potential of the form
\[ A(y, t) = -\frac{E_0}{\omega} \exp \left[ -2 \ln(2) \left( \frac{ct - y}{c\tau} \right)^2 \right] \sin(\omega t - ky)z, \] (43)
where \(k = \omega/c\) is the wave number of the laser field and \(\tau\) is the full width at half maximum of the pulse duration in intensity. The direction of both the vector potential and the electric field is along the \(z\) axis. We take the propagation direction of the laser field to be along the \(y\) axis and hence the magnetic field points along the \(x\) axis. We study \(Ar\) driven by a laser pulse with intensities ranging from \(2 \times 10^{14}\) \(\text{W/cm}^2\) to \(5 \times 10^{14}\) \(\text{W/cm}^2\) and durations of \(\tau = 20\) fs, 25 fs and 30 fs at 800 nm.

The time propagation of strongly-driven \(Ar\) starts at time \(t_0\) and stops at an asymptotically large time \(t_f\). For each trajectory, if the energies of three (two) electrons are positive, we label the trajectory as a triple (double) ionization event. The DI and TI probabilities are
\[ P_{\text{DI}} = \frac{N_{\text{DI}}}{N}, \quad P_{\text{TI}} = \frac{N_{\text{TI}}}{N}, \] (44)
where \(N_{\text{DI}}, N_{\text{TI}}\) and \(N\) are the numbers of doubly-ionized, triply-ionized and all events, respectively. Here, we mainly focus on non-sequential double ionization (NSDI) and on non-sequential triple ionization (NSTI) events. NSDI and NSTI involve an electron accelerating in the laser field and coming back to the core to transfer energy to bound electrons via a recollision. This energy transfer can lead to the escape of two electrons (NSDI) or three electrons (NSTI). Electronic correlation, a fundamental interaction, underlies this field-assisted recollision [58].

To identify a recollision in either one of the two models, we monitor the Coulomb potential between all pairs of a quasi-free and a bound electron. We identify the maxima in the inter-electronic Coulomb potential energy as function of time. We label the times when the inter-electronic distance is minimum as recollision times \(t_{\text{rec}}\).

Also, we define the ionization time of electron \(i\), \(t_{\text{ion}}^i\), to be the time when the compensated energy becomes positive and remains positive thereafter [47]. We used the same definition for \(t_{\text{ion}}^i\) in all our previous studies, see for instance [7, 59]. The compensated energy is given by
\[ \varepsilon_i^{\text{comp}}(t) = \frac{p_i^2}{2m_i} + \frac{Q_i Q_j}{|r_1 - r_i|} + \sum_{j=2}^{N} c_{i,j}(t) V_{\text{eff}}(\zeta_j, |r_1 - r_j|), \]
\[ \varepsilon_i^{\text{comp}}(t) = \frac{p_i^2}{2m_i} + \frac{Q_i Q_j}{|r_1 - r_i|} + V_{\text{H,i}}, \]
for the ECBB- and the H-model respectively. Moreover, a TI or DI event is labelled as direct if the energy transferred from a recolliding electron to bound electrons suffices for the simultaneous ionization, shortly after recollision, of three or two electrons. In Appendix C, we outline the algorithm used to label an event as direct. Here, we label the remaining events as delayed TI and DI events.

### A. DI and TI ionization probabilities

We find that the DI and TI probabilities are consistently larger for the H-model for both \(\alpha = 2, 4\) compared to the ECBB-model. This is consistent with the different initial conditions the bound electrons have in the two models. The initial momenta of the bound electrons are higher in the H-model versus the ECBB-model, compare Fig. 3(b) with Fig. 4(b). Also, the repulsive Heisenberg potential reduces the attraction of each electron from the core resulting in higher ionization probabilities. Regarding the DI probability, for the H-model, we find that at intensities \(2 \times 10^{14}\) \(\text{W/cm}^2\), \(4 \times 10^{14}\) \(\text{W/cm}^2\), and \(5 \times 10^{14}\) \(\text{W/cm}^2\), we observe
W/cm² and 20 fs pulse duration the DI probability is consistently higher for α = 4 compared to α = 2. However, while at the two smallest intensities the difference in the DI probability is small for the two values of α, at 5 × 10¹⁴ W/cm² the DI probability is almost 71 % higher for α = 4 compared to α = 2. Hence, the DI probability depends significantly on the value of α, a disadvantage of the H-model. In what follows, we consider α = 2 for the H-model, unless otherwise stated, since this value allows the electrons to access a larger phase space. As we increase the intensity from 2 × 10¹⁴ W/cm² to 5 × 10¹⁴ W/cm², we find that the ratio of the DI probabilities between the two models, P_D[ECBB]/P_D[H], decreases from 1.1 to 0.4. However, the ratio of the TI probabilities P_T[ECBB]/P_T[H] increases from 0.03 to 0.2. Hence, for the intensities considered here, the DI and TI probabilities are smaller for the ECBB-model.

B. Distribution of the sum of the electron momenta

In Fig. 5, we plot the TI and DI probability distribution of the sum of the pₓ momenta of the ionizing electrons at intensities 2 × 10¹⁴ W/cm², 4 × 10¹⁴ W/cm² and 20 fs pulse duration and 5 × 10¹⁴ W/cm² and 25 fs pulse duration. The highest intensity pulse allows for a direct comparison with experimental results [19]. In Figs. 5(a1) and 5(a2), for the ECBB-model, we plot the TI and DI probability distributions of the sum of the pₓ momenta of the ionizing electrons. For DI (Fig. 5(a2)), we find that the probability distribution is centered around zero and the width decreases with increasing intensity. This is in accord with our previous findings for two-electron Ar driven by a 4 fs pulse for intensities from 2 × 10¹⁴ W/cm² to 5 × 10¹⁴ W/cm² [37]. For TI (Fig. 5(a1)), we find similar doubly-peaked distributions for 4 × 10¹⁴ W/cm² and 5 × 10¹⁴ W/cm². The TI probability at 2 × 10¹⁴ W/cm² is very low and we do not consider this intensity in Fig. 5(a1).

In Figs. 5(b1) and 5(b2), for the H-model for α = 2, 4, we plot the TI and DI probability distributions of the sum of the pₓ momenta of the ionizing electrons. For DI (Fig. 5(b2)), we find that the probability distribution is centered around zero and the width decreases with increasing intensity for both α. For TI (Fig. 5(b1)), we find that at 2 × 10¹⁴ W/cm² the distribution is doubly-peaked, while at higher intensities the distribution is centered around zero. Finally, we find that the TI distributions are similar for the two values of α, while the DI distributions are more centered around zero for α = 4. This is consistent with each electron being less attracted from the core for larger values of α resulting in smaller final momenta. Hence, the probability distributions depend on the value of α.

Comparing the TI (Fig. 5(c1)) and DI (Fig. 5(c2)) probability distributions of the ECBB- and H-model for α = 2, we find that all distributions are more centered around zero for the H-model. Moreover, the TI distributions at higher intensities are doubly-peaked for the ECBB-model and centered around zero for the H-model.

![FIG. 5. Probability distributions of the sum of the electron momentum components parallel to the polarization of the laser field for TI (left column) and DI (right column) at intensities 2 × 10¹⁴ W/cm² (20 fs), 4 × 10¹⁴ W/cm² (20 fs) and 5 × 10¹⁴ W/cm² (25 fs). The ECBB-model results are presented in the top row, the H-model in the middle row and comparison of the two models in the third row. All probability distributions are normalized to one.](attachment:image_url)
consistent with our previous results of double ionization of two-electron Ar driven by short pulses [37]. We find the percentage of direct events to be significantly larger for the ECBB- compared to the H-model. The contribution of direct events to DI is roughly 50 % for the ECBB-model, while it decreases from 16 % to 5 % with increasing intensity for the H-model. The contribution of direct events to TI is roughly 20 % for the ECBB-model while it is roughly 5 % for the H-model at the two highest intensities. For the delayed pathway, for DI, the distributions are centered around zero for both models (Fig. 6(b2)), while for TI the distributions are less centered around zero for the ECBB-model (Fig. 6(b1)).

FIG. 6. Probability distributions of the sum of electron momenta components parallel to the polarization of the laser field for TI (left column) and DI (right column) at intensities $2 \times 10^{14}$ W/cm$^2$ (20 fs), $4 \times 10^{14}$ W/cm$^2$ (20 fs) and $5 \times 10^{14}$ W/cm$^2$ (25 fs). The direct pathway distributions are plotted on the top row and the delayed pathway distributions are plotted on the bottom row. All probability distributions are normalized to one.

Next, we compare with experimental results the findings of the ECBB- and H-model for the DI distribution of the sum of the p$_z$ electron momenta of Ar at $4 \times 10^{14}$ W/cm$^2$ (30 fs) (Fig. 7(c)) and $5 \times 10^{14}$ W/cm$^2$ (25 fs) (Fig. 7(b)) [21, 22] as well as the TI distribution at $5 \times 10^{14}$ W/cm$^2$ (25 fs) [22] (Fig. 7(a)). The experimental DI distributions have a slight double-peaked structure and agree more with the results of the ECBB-model. Indeed, the H-model produces DI distributions that are highly centered around zero, which is significantly less the case for the ECBB-model. The experimental TI distributions have a slight double-peaked structure which is only reproduced by the ECBB-model. For the H-model, weaker recollisions are consistent with the DI and TI distributions of the sum of the p$_z$ electron momenta being more centered around zero, see Fig. 5.

C. Strength of the recollision in DI and TI events

For each DI and TI event we register all the maxima of the Coulomb inter-electronic potential energy as a function of time and identify the largest maximum $V_{\text{max}}$. That is, we identify the most important recollision for each event. We plot the distribution of $V_{\text{max}}$ for TI (Fig. 8(a)) and DI (Fig. 8(b)) events. We find that recollisions are significantly stronger for the ECBB-model, with the most probable value of $V_{\text{max}}$ being roughly 1 a.u. for DI and TI at all intensities. In contrast, for the H-model, at the higher intensities, the most probable value of $V_{\text{max}}$ is close to 0 a.u. both for DI and TI. For the H-model, weaker recollisions are consistent with the DI and TI distributions of the sum of the p$_z$ electron momenta being more centered around zero, see Fig. 5.
FIG. 8. Probability distributions of the largest value of the Coulomb inter-electronic potential energy for TI (a) and DI (b) at intensities $2 \times 10^{14}$ W/cm$^2$ (20 fs), $4 \times 10^{14}$ W/cm$^2$ (20 fs) and $5 \times 10^{14}$ W/cm$^2$ (25 fs). All probability distributions are normalized to one.

D. Correlated momenta

In Fig. 9, for DI, we plot the correlated electron momenta at intensities $2 \times 10^{14}$ W/cm$^2$ (20 fs), $4 \times 10^{14}$ W/cm$^2$ (20 fs) and $5 \times 10^{14}$ W/cm$^2$ (25 fs) obtained with the ECBB-model ((a1)-(a3)) and the H-model ((b1)-(b3)). At the three intensities, we find that correlated electron escape prevails mostly for the ECBB-model which produces roughly 10% more correlated events than the H-model. Also, at intensities $4 \times 10^{14}$ W/cm$^2$ and $5 \times 10^{14}$ W/cm$^2$, the electrons escape with considerably higher momenta in the ECBB-model, compare Fig. 9 (a2) with Fig. 9 (b2) and Fig. 9 (a3) with Fig. 9 (b3). The above are consistent with the ECBB-model resulting in more direct events (Sec. III B) and stronger recollisions (Fig. 8) than the H-model.

In Fig. 10, for TI, we plot the correlated electron momenta at intensities $4 \times 10^{14}$ W/cm$^2$ (20 fs) and $5 \times 10^{14}$ W/cm$^2$ (25 fs) obtained with the ECBB-model ((a1)-(a2)) and the H-model ((b1)-(b2)). We find that correlated three-electron escape is clearly prevalent at both intensities for the ECBB-model which produces roughly 10% more correlated events than the H-model. Also, at intensities $4 \times 10^{14}$ W/cm$^2$ and $5 \times 10^{14}$ W/cm$^2$, the electrons escape with considerably smaller momenta compared to the H-model, compare Fig. 10 (a1) with Fig. 10 (b1) and Fig. 10 (a2) with Fig. 10 (b2). As for DI, this is consistent with the ECBB-model yielding more direct events and stronger recollisions versus the H-model.

E. Angular distributions

In Fig. 11, we plot the TI (left column) and DI (right column) probability distributions of the angles of the ionizing electrons and the core at intensity $4 \times 10^{14}$ W/cm$^2$ at 20 fs. We obtain similar results for the other intensities considered in this work (not shown). We find that the angle between any pair of escaping electrons $\theta_{ee}$ (black color) is mostly peaked at small angles indicating a correlated electron escape. For both the ECBB-model and the H-model, we find that the angle of inter-electronic escape is smaller for TI versus DI. This is consistent with
the electron momenta being more correlated for TI versus DI, compare Fig. 9 with Fig. 10. We also find that a small angle of inter-electronic escape is significantly more favoured by the ECBB-model, which results in more direct events and stronger recollisions.

We also find that the angle of any escaping electron with the z axis, \( \theta_{e-z} \), (dark gray color) peaks at small and large values for TI and DI for both models. That is, the ionizing electrons escape mostly along \( (0^\circ) \) or opposite \( (180^\circ) \) the direction of the electric field. However, the peaks of the distributions of \( \theta_{e-z} \) are sharper, i.e. the distributions are less wide, for the H-model. This is in accord with our finding that the H-model gives rise to a significantly higher number of TI and DI events where no-recollision takes place compared to the ECBB-model. As a result, in the H-model, the electrons ionize mostly due to the field for a larger number of events, with the electrons escaping more along or opposite the direction of the field. Moreover, we find that the distributions of the angle of the core with the z axis, \( \theta_{c-z} \), (blue color) are wide for both TI and DI for both models. However, the distribution is wider for DI versus TI. This is consistent with the core having a higher charge equal to 3 for TI versus 2 for DI. As a result, the electric field exerts a larger force on the core in TI leading the core to escape more along or opposite the direction of the electric field. Finally, we find that the distribution of the angle \( \theta_{c-e} \) (light grey color) between an ionizing electron and the core peaks mostly at large angles, that is, the electron and the core escape in opposite directions. This is consistent with the electric field exerting opposite forces to particles of opposite charges. We find that the angle of escape between an electron and the core is larger for TI compared to DI for both models. This is consistent with the larger core charge for TI resulting in the core escaping more along or opposite the field direction.

IV. CONCLUSIONS

We formulate a 3D semi-classical model to address three-electron dynamics in a strongly driven atom where the electron and core dynamics are treated at the same time. Our formulation includes the magnetic field of the laser field as well as the Coulomb singularities. We address unphysical autoionization present in semi-classical models where the Coulomb singularities are accounted for and more than one electron is bound. We do so by substituting the Coulomb repulsion between bound electrons with effective potentials where an effective charge is associated with every bound electron. The interaction between pairs of electrons that are not both bound is accounted for with the full Coulomb potential and all other forces are fully accounted for. This model, developed in this work and referred to as the ECBB-model, identifies on the fly during time propagation if an electron is bound or not. We compare the ionization spectra obtained with the ECBB-model with the ones obtained with a model previously developed—referred to here as the H-model. In the latter model, a potential is added for each electron that mimics the Heisenberg uncertainty principle and restricts the accessible phase space of each electron preventing autoionization. The advantage of the ECBB-model is that it accurately treats the interaction of each electron with the core and all other interactions while it treats less accurately the interaction between bound electrons. The advantage of the H-model is that it accurately treats the interaction between all electrons while it treats less accurately the interaction of each electron with the core.

Using these 3D semi-classical models, we address triple and double ionization in a strongly-driven atom, namely Ar. We compare the ionization spectra obtained with the two models as well as with experiment for various pulse durations and intensities. We find that both double and triple ionization probabilities are greater for the H-model compared to the ECBB-model. We conjecture that this difference in the probabilities is due to the Heisenberg potential resulting in larger initial momenta of the bound electrons as well as in a significant less attraction of each electron from the core. We find that in the H-model for a significant number of events the electrons ionize without a recollision, i.e. ionize due to the laser field and the recollisions are significantly weaker compared to the ECBB-model. These findings are consistent with our results for the distribution of the sum of the momenta of the ionizing electrons along the direction of the laser field. For all the intensities and pulse durations considered here, we find that these distributions are broader for the ECBB-model. For triple ionization, we find that the distributions of the
sum of the electron momenta have a double peak for the ECBB-model while they are centered around zero for the H-model. We find this to be due to the ECBB-model producing more direct ionization events than the H-model. This is also evident in the correlated electron momenta where the distributions obtained with the ECBB-model are consistently more correlated compared to the ones obtained with the H-model. Moreover, we identify another disadvantage of the H-model, namely, the distributions of the momenta and ionization probabilities depend on the parameter $\alpha$ in the Heisenberg potential. Comparing with experimental distributions of the sum of the momenta we find that the distributions obtained with the ECBB-model have a better agreement with experiment mainly for double ionization. Finally, our formulation of the ECBB-model and of the H-model that account for electron and core motion and for non-dipole effects is general and can be generalized to strongly-driven atoms with more than three electrons.

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Appendix A: Derivation of the effective potential

The electric field produced by a charge $Q(\zeta_j, r)$ that is contained within a spherical shell of radius $r$ from the core is obtained by Gauss’s law as follows:

$$
E(\zeta_j, r) = \frac{Q(\zeta_j, r)}{r^2} \hat{r}.
$$

(A1)

The work $W$ done on a particle $i$ due to the electric field $E(\zeta_j, r)$ is equal to minus the change in potential energy $\Delta V_{\text{eff}}$:

$$
W = -\Delta V_{\text{eff}} = - [V_{\text{eff}}(\zeta_j, r) - V_{\text{eff}}(\zeta_j, \infty)] - V_{\text{eff}}(\zeta_j, r).
$$

(A2)

where we have used that $V_{\text{eff}}(\zeta_j, \infty) = 0$. The work $W$ is also given by

$$
W = \int_{\infty}^{r} F \cdot dr' = -\int_{\infty}^{r} E \cdot dr'
$$

$$
= -\int_{\infty}^{r} E(\zeta_j, r')(i' \cdot r') dr'
$$

$$
= \int_{\infty}^{r} \left[ \frac{1}{r'^2} - \frac{e^{-2\zeta_j r'}(1 + 2\zeta_j r')}{r'^2} - 2\zeta_j^2 e^{-2\zeta_j r'} \right] dr'
$$

$$
= \left[ -\frac{1}{r'} + \frac{e^{-2\zeta_j r'}}{r'} + \zeta_j e^{-2\zeta_j r'} \right]|_{\infty}^{r}
$$

$$
= 1 - (1 + \zeta_j r)e^{-2\zeta_j r}.
$$

(A3)

where we have used that particle $i$ is an electron and hence $F = -E$ as well as that particle $j$ is an electron and $Q(\zeta_j, r)$ is given by Eq. (19). Using Eqs. (A2) and (A3), we find

$$
V_{\text{eff}}(\zeta_j, r) = \frac{1 - (1 + \zeta_j r)e^{-2\zeta_j r}}{r},
$$

(A4)

which is the potential energy that an electron $i$ has at a distance $r$ from the core due to a bound electron $j$.

Appendix B: Leapfrog Algorithm

In what follows, we describe the leapfrog algorithm. First, we initialize the auxiliary variables $W_0^q = q_0, W_0^\rho = \rho_0, W_0^t = t_0$ and $W_0^\xi = \xi_0$. Then, we propagate for a time step equal to $h$, by propagating for half a step each quadruplet of variables $(q, W^\rho, t, W^\xi)$ and $(W^q, \rho, W^t, \xi)$ in an alternating way as follows:

$$
q_{1/2} = q_0 + \frac{h}{2} \frac{q^q_0}{\Omega(q_0^q)}
$$

$$
W_0^\rho + \frac{h}{2} \frac{\rho^q_0}{\Omega(q_0^q)}
$$

$$
t_{1/2} = t_0 + \frac{h}{2} \frac{1}{\Omega(q_0^q)}
$$

$$
W_0^\xi + \frac{h}{2} \frac{\xi^q_0}{\Omega(q_0^q)}
$$

$$
W_1^q = W_0^q + h \frac{q_{1/2}}{\Omega(q_{1/2})}
$$

$$
W_1^\rho = W_0^\rho + h \frac{\rho_{1/2}}{\Omega(q_{1/2})}
$$

$$
W_1^t = W_0^t + h \frac{t_{1/2}}{\Omega(q_{1/2})}
$$

$$
W_1^\xi = W_0^\xi + h \frac{\xi_{1/2}}{\Omega(q_{1/2})}.
$$
\[ \mathcal{E}_1 = \mathcal{E}_0 + h \frac{\dot{\mathcal{E}}(q_{1/2}, W^\rho_{1/2}, t_{1/2}, W^\varepsilon_{1/2})}{\Omega(q_{1/2})} \]

\[ q_{1/2} = q_0 + h \frac{\dot{q}(W^q_0, \rho_0, W^\rho_0)}{2 \Omega(W^q_0)} \]

\[ W^\rho_{1/2} = W^\rho_0 + h \frac{\dot{\rho}(W^q_0, \rho_0, W^\rho_0, \mathcal{E}_0)}{2 \Omega(W^q_0)} \]

\[ t_{1/2} = t_0 + h \frac{1}{2 \Omega(W^q_0)} \]

\[ W^\varepsilon_{1/2} = W^\varepsilon_0 + h \frac{\dot{\mathcal{E}}(W^q_0, \rho_0, W^\rho_0, \mathcal{E}_0)}{2 \Omega(W^q_0)} \]

\[ \mathcal{E}_0 = \mathcal{E}_0 + h \frac{\dot{\mathcal{E}}(q_{1/2}, W^\rho_{1/2}, t_{1/2}, W^\varepsilon_{1/2})}{\Omega(q_{1/2})} \]

The subscripts 0,1/2,1 denote the value of each variable at the start, the middle and the end of the time step \( h \).

Next, we describe the algorithm that incorporates the leapfrog method in the Bulirsch-Stoer extrapolation scheme over a step \( H \), which is split into \( n \) sub steps of size \( h = H/n \):

\[ q_{m-1/2} = q_{m-3/2} + h \frac{\dot{q}(W^q_{m-1}, \rho_{m-1}, W^\rho_{m-1})}{2 \Omega(W^q_{m-1})} \]

\[ W^\rho_{m-1/2} = W^\rho_{m-3/2} + h \frac{\dot{\rho}(W^q_{m-1}, \rho_{m-1}, W^\rho_{m-1}, \mathcal{E}_{m-1})}{2 \Omega(W^q_{m-1})} \]

\[ t_{m-1/2} = t_{m-3/2} + h \frac{1}{2 \Omega(W^q_{m-1})} \]

\[ W^\varepsilon_{m-1/2} = W^\varepsilon_{m-3/2} + h \frac{\dot{\mathcal{E}}(W^q_{m-1}, \rho_{m-1}, W^\rho_{m-1}, \mathcal{E}_{m-1})}{2 \Omega(W^q_{m-1})} \]

\[ W^q_m = W^q_{m-1} + h \frac{\dot{q}(q_{m-1/2}, W^\rho_{m-1/2}, t_{m-1/2}, W^\varepsilon_{m-1/2})}{\Omega(q_{m-1/2})} \]

\[ W^\rho_m = W^\rho_{m-1} + h \frac{\dot{\rho}(q_{m-1/2}, W^\rho_{m-1/2}, t_{m-1/2}, W^\varepsilon_{m-1/2})}{\Omega(q_{m-1/2})} \]

\[ W^\varepsilon_m = W^\varepsilon_{m-1} + h \frac{\dot{\mathcal{E}}(q_{m-1/2}, W^\rho_{m-1/2}, t_{m-1/2}, W^\varepsilon_{m-1/2})}{\Omega(q_{m-1/2})} \]

\[ \mathcal{E}_m = \mathcal{E}_{m-1} + h \frac{\dot{\mathcal{E}}(q_{m-1/2}, W^\rho_{m-1/2}, t_{m-1/2}, W^\varepsilon_{m-1/2})}{\Omega(q_{m-1/2})} \]

where \( m=2,...,n-1 \).

**Appendix C: Identifying the direct pathway of TI and DI events**

We obtain the TI and DI events with a code that incorporates the formulation of the ECBB-model described in Sec. II C and a code that incorporates the formulation of the H-model described in Sec. II B. Once we obtain these events, we perform a detailed analysis with a different set of codes. In both analysis codes, i.e. one for each model, we use the framework we developed in Sec. II C 5 to determine on the fly during propagation if an electron is quasi-free or bound. We register a TI or DI event as direct if a recollision is associated with the simultaneous ionization of three or two electrons. We take the following steps to identify direct events:

1. We find the ionization time of each electron, \( t^i_{\text{ion}} \), with \( i = 1,2,3 \) for TI and \( i = 1,2 \) for DI.

2. We register the maxima in the inter-electronic potential energies as a function of time between electron pairs \( i,j \) and \( i,k \) and \( j,k \) during the time intervals when in these pairs one electron is quasi-free and the other is bound. Next, for each electron \( i \), we identify the maximum for each one of the \( i,j \) and
i, k potential energies that is closest to the time $t^{i_{\text{ion}}}_{\text{ion}}$. We denote these times as $t^{i,j}_{\text{rec}}$ and $t^{i,k}_{\text{rec}}$. We obtain at most six such times for TI events and four for DI events.

3. For each time $t^{i,j}_{\text{rec}}$ we identify the time $t_{2}$ (see Sec. II C 5) of closest approach to the core of the quasi-free electron (either electron i or j) that is closest to $t^{i,j}_{\text{rec}}$ and denote it as $t^{i,j}_{2}$. We obtain at most six such times for TI events and four for DI events.

We label a TI event as direct if four of the times $t^{i,j}_{2}$ are the same, accounting for one electron being quasi-free and the other two bound. That is, if electron i is quasi-free during the recollision closest to the ionisation time $t^{ion}_{i}$ then the times $t^{i,j}_{2}$, $t^{i,k}_{2}$, $t^{i,j}_{2}$ and $t^{i,k}_{2}$ should be the same. The times $t^{i,j}_{2}$ and $t^{i,k}_{2}$ are associated with the recollision times $t^{i,j}_{\text{rec}}$ and $t^{i,k}_{\text{rec}}$ for the bound electron j and k respectively. For the quasi-free electron we obtain two recollision times $t^{i,j}_{\text{rec}}$ and $t^{i,k}_{\text{rec}}$ associated with the ionization time $t^{i_{\text{ion}}}_{\text{ion}}$. We choose the one that has the largest difference from $t^{i_{\text{ion}}}_{\text{ion}}$, guaranteeing a stricter criterion for direct TI events. Next, we check whether $t^{i,j}_{\text{rec}} - t^{i_{\text{ion}}}_{\text{ion}} < t\text{diff}$ or $(t^{i_{\text{ion}}}_{\text{ion}} < t^{i,j}_{\text{rec}}$ & $t^{i_{\text{ion}}}_{\text{ion}} < t^{i,k}_{\text{rec}}$) and $t^{i,j}_{\text{rec}} - t^{i_{\text{ion}}}_{\text{ion}} < t\text{diff}$ and $(t^{i,k}_{\text{rec}} - t^{i_{\text{ion}}}_{\text{ion}} < t\text{diff})$. If the latter conditions are satisfied then we label the event as direct TI. The condition $t^{i_{\text{ion}}}_{\text{ion}} < t^{i,j}_{\text{rec}}$ & $t^{i_{\text{ion}}}_{\text{ion}} < t^{i,k}_{\text{rec}}$ has also been used in our previous studies [37, 61] to account for a quasi-free electron ionising significantly earlier before recollision. This happens mostly at high intensities. A similar process is followed to identify a DI event.

The interval $t\text{diff}$ is defined as the time duration where the inter-electronic potential energy undergoes a sharp change due to recollision. For the laser field intensities considered in this work, we find $t\text{diff}$ to be roughly equal to 1/8 laser cycle (T) for the EBBB-model and 1/6 T for the H-model. The difference in $t\text{diff}$ between the two models is due to the stronger electron-core interaction for the EBBB-model resulting in sharper changes to the electron-electron interaction. The choice of $t\text{diff}$ does not significantly change the percentage contribution of direct TI and DI events [37].

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