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Ionization of atoms and molecules by short, strong laser pulses

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Abstract. This article provides an extension to multiparticle systems (like atoms and molecules with many electrons) of nonperturbative results obtained previously by the authors in collaboration with Fring on the ionization of atomic bound states under the influence of short, ultra-intense laser pulses. We give upper and lower bounds which in particular exclude stabilization.

1. Introduction

With the advance of modern laser technology photoionization of atoms and molecules under the influence of short, ultra-intense fields has become the object of an intensive theoretical discussion. Mostly, however, perturbative arguments have been used, often supported by a computer analysis (see e.g. [1, 2] and references therein).

Within the theoretical context of a (nonrelativistic) quantum mechanical one-particle a.c. Stark Hamiltonian in the dipole approximation (including the hydrogen atom as a particular case) and in collaboration with Enss and Fring the authors have proposed a nonperturbative approach [3–5]. In particular [5] provides rigorous upper and lower bounds on the ionization of bound states.

The upper bound is valid for (classical) electric fields of short duration and whose classical energy transfer is smaller than the classical ionization energy. The lower bound also holds for short pulses but now the classical energy transfer has to be larger than the classical ionization energy.

The aim of this article is to extend these one-particle results to (nonrelativistic) quantum mechanical multiparticle systems with Coulomb interaction. The lower bound is relevant in the context of the so-called stabilization found by the majority of the atomic-physics community (see e.g. [1, 6] and references therein). This means that the probability of ionization by a pulse of laser radiation, which for low intensities increases with increasing intensity, reaches some sort of maximum at high intensities and commences to decrease. This picture is counter-intuitive and doubts about the existence of this phenomenon have been raised by some authors [2, 7–11], who do not find evidence for it in their numerical calculations. Since we view this article as a companion to [5], we refer the reader to this

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article and to [12] for a detailed discussion of the relevance of our bounds to the issue of stabilization.

The strategy of our discussion will be to separate out the centre of mass motion, which will be nontrivial if the system is not neutral.

We will then give a definition of ionization probability for a bound state in the centre of mass system. Strictly speaking one should actually speak of ionization-fragmentation probability. For brevity, however, we will use the notion ionization probability, since our definition does not yet allow for a distinction between one-electron and multi-electron ionization nor for the fragmentation into two or more molecules nor all effects combined. Such a discussion would require a multichannel analysis which is beyond the scope of this work. We note that a multi-channel analysis is also necessary in (nonrelativistic) multiparticle scattering processes and it requires sophisticated methods (see e.g. [13, 14]). Our definition of the total ionization probability, however, allows for a discussion of gauge invariance and we refer the reader to [5] for a general discussion and with an application to the Stark Hamiltonian. The proof of the upper and lower bounds follows the strategy used in [4] and [5]. The new ingredient is to extend the Kramers–Henneberger transformation [15–17] to the multiparticle system and to show that it decomposes into a part for the centre of mass motion and a part for the centre of mass frame. The remainder of the arguments may then essentially be taken over from [4] and [5]. We also note that earlier the Kramers-Henneberger relation was utilized in [18] for an analysis of three-particle quantum systems in a periodic electric field.

The electric field is assumed to be only time-dependent and thus independent of space, which means that we use a dipole approximation. The only other restriction on its shape is that it vanishes unless $0 \leqslant t \leqslant \tau$ and that it is piecewise continuous, which means that we allow for jumps. In particular no smooth switch on and off is required. For simplicity we will work with linearly polarized electric fields. Our arguments can be easily adopted for a case of circular or elliptical polarized field. Throughout we use units where $\hbar = e = 1$.

2. Ionization bounds for atoms and molecules

Let the molecule (atom or ion as particular cases) consist of N nuclei with charges $Q_i (1 \leqslant i \leqslant N, Q_i > 0$ integer) and masses $M_i > 0$, and N' electrons with charge -1 and mass m. In particular for an electrically neutral system $Q = \sum_{i=1}^{N} Q_i - N' = 0$. Our discussion below, however, will also cover the case $Q \neq 0$.

Let $\mathbf{x} = (\vec{x}_1, \ldots \vec{x}_N) \in \mathbb{R}^{3N}$ be the coordinates of the nuclei and $\mathbf{x}' = (\vec{x}'_1, \ldots \vec{x}'_{N'}) \in \mathbb{R}^{3N'}$ those of the electrons.

For a linearly polarized electric field $\mathcal{E}(t)$ in the z-direction the a.c. Stark Hamiltonian is given by

$$H(t) = H_0 + V + z \cdot \mathcal{E}(t)$$

where

$$z = \sum_{i=1}^{N} z_i Q_i - \sum_{j=1}^{N'} z'_j.$$

Here z_i and z'_j are the z-components of $\vec{x}_i$ and $\vec{x}'_j$, respectively. Also H_0 is the free Hamiltonian

$$H_0 = -\sum_{i=1}^{N} \frac{1}{2M_i} \Delta_i - \frac{1}{2m} \sum_{j=1}^{N'} \Delta'_j = \sum_{i=1}^{N} \frac{\vec{p}_i^{\,2}}{2M_i} + \sum_{j=1}^{N'} \frac{\vec{p}_j^{\,\prime 2}}{2m}$$

where $\Delta_i(\Delta'_j)$ is the Laplacian with respect to the variable $\vec{x}_i(\vec{x}'_j)$. The potential V is supposed to be Coulombic, i.e. of the form

$$V(\mathbf{x},\mathbf{x}') = \sum_{1\leqslant i<i'\leqslant N} \frac{Q_i Q_{i'}}{|\vec{x}_i - \vec{x}_{i'}|} - \sum_{\substack{1\leqslant i\leqslant N\\ 1\leqslant j\leqslant N'}} \frac{Q_i}{|\vec{x}_i - \vec{x}'_j|} + \sum_{1\leqslant j<j'\leqslant N'} \frac{1}{|\vec{x}'_j - \vec{x}'_{j'}|}.$$

Instead of the two-body Coulomb potential other forms of two-body potentials may also be considered (see e.g. [4]). We set

$$H = H_0 + V$$

such that $H(t) = H + z\cdot\mathcal{E}(t)$. With $M = \sum_{i=1}^{N} M_i + mN'$ being the total mass of the system, the coordinate of the centre of mass is given by

$$\vec{X} = \frac{1}{M}\left(\sum_{i=1}^{N} M_i\vec{x}_i + m\sum_{j=1}^{N'}\vec{x}'_j\right) \in \mathbb{R}^3.$$

Then

$$H_{0,cm} = H_0 + \frac{1}{2M}\Delta_{\vec{X}} = H_0 - H'_0$$

is the kinetic energy in the centre of mass frame, where $\Delta_{\vec{X}}$ is the Laplace operator for the centre of mass motion. We set

$$H_{cm} = H + \frac{1}{2M}\Delta_{\vec{X}} = H - H'_0 = H_{0,cm} + V$$

and

$$H_{cm}(t) = H(t) + \frac{1}{2M}\Delta_{\vec{X}} - Q\cdot Z\cdot\mathcal{E}(t) = H(t) - H'(t)$$

with Z being the z-component of $\vec{X}$.

These operators are supposed to act in the Hilbert space $\mathcal{H}\subset L^2(\mathbb{R}^{3(N+N')})$ of all square integrable functions $\psi(\mathbf{x},\mathbf{x}')$ which are antisymmetric in the electron variables $(\vec{x}'_1,\ldots\vec{x}'_{N'})$. Let $\mathcal{H}_{cm}\subset L^2(\mathbb{R}^{3(N+N'-1)})$ be the Hilbert space of square integrable functions in relative coordinates, antisymmetric with respect to permutations of the electrons $(j\leftrightarrow j')$. Our discussion below could also apply to a situation where some of the nuclei have to be treated as identical particles (boson or fermions). We will call $\mathcal{H}_{cm}$ (the Hilbert space of) the centre of mass system.

In what follows, we will often identify operators B_{cm} acting in $\mathcal{H}_{cm}$ with operators acting in $\mathcal{H} = \mathcal{H}_{cm}\otimes L^2(\mathbb{R}^3)$, where $L^2(\mathbb{R}^3)$ is the Hilbert space for the centre of mass motion (see e.g. [19]). Since V depends on the relative coordinates only, it defines a multiplication operator on $\mathcal{H}_{cm}$. Therefore we have

$$\begin{aligned} H_0 &= H_{0,cm}\otimes\mathbf{1} + \mathbf{1}\otimes H'_0\\ H &= H_{cm}\otimes\mathbf{1} + \mathbf{1}\otimes H'_0\\ H(t) &= H_{cm}(t)\otimes\mathbf{1} + \mathbf{1}\otimes H'(t). \end{aligned} \tag{1}$$

It is well known (see e.g. [19]) that H and H_{cm} define self-adjoint operators. This is also true for $H(t)$, $H_{cm}(t)$ and $H'(t)$ (see e.g. [20, 21]). By definition, a bound state of the system is an eigenstate of H_{cm} with eigenvalue $E<0$. Let P be the orthogonal projection on the subspace of $\mathcal{H}_{cm}$ spanned by the bound state eigenfunctions.

If S denotes the scattering operator for the pair $(H(\cdot), H)$ (see below), then we will see that S decomposes as $S = S_{cm}\otimes S'$, where S_{cm} is the scattering operator in $\mathcal{H}_{cm}$ for the

pair $(H_{cm}(\cdot), H)$ and S' is the scattering operator for the pair $(H'(\cdot), H'_0)$ in $L^2(\mathbb{R}^3)$. The ionization (probability) for a normalized bound state $\psi \in \mathcal{H}_{cm}$ is defined as

$$I(\psi) = \|(\mathbf{1} - P)S_{cm}\psi\|^2 = 1 - \|PS_{cm}\psi\|^2. \tag{2}$$

As mentioned in the introduction, this is a total ionization probability in the sense that it does not distinguish between the different channels with continuous spectrum. To state the results of this article, we need some further notation.

For given pulse $\mathcal{E}(t)$, we introduce the quantities

$$b(t) = \int_0^t \mathcal{E}(s)\mathrm{d}s$$
$$c(t) = \int_0^t b(s)\mathrm{d}s = tb(t) - \int_0^t s\mathcal{E}(s)\mathrm{d}s$$
$$a(t) = \frac{1}{2}\int_0^t b(s)^2\mathrm{d}s.$$

Also we introduce the time dependent vectors

$$\mathbf{x}(t) = (\vec{x}_1(t), ..., \vec{x}_N(t))$$
$$\mathbf{x}'(t) = (\vec{x}'_1(t), ..., \vec{x}'_{N'}(t))$$

with

$$\vec{x}_i(t) = \vec{x}_i - \frac{Q_i}{M_i}c(t)e_z \qquad 1 \leqslant i \leqslant N$$
$$\vec{x}'_j(t) = \vec{x}'_j + \frac{c(t)}{m}e_z \qquad 1 \leqslant j \leqslant N'$$

where e_z is the unit vector in the z-direction. Let $V(t)$ be the multiplication operator on $\mathcal{H}$ given as

$$V(t)(\mathbf{x}, \mathbf{x}') = V(\mathbf{x}(t), \mathbf{x}'(t)).$$

Again $V(t)$ only depends on the relative coordinates and, therefore, also defines a multiplication operator in $\mathcal{H}_{cm}$.

Finally $p_{cm,z}$ denotes the z-component of a certain momentum operator in $\mathcal{H}_{cm}$ (defined by relations (10), (12) and (13) below).

By μ we denote the mass parameter defined by

$$\mu^{-1} = \sum_{i=1}^{N}\frac{Q_i^2}{M_i} + \frac{N'}{m} - \frac{Q^2}{M}.$$

It is easy to see that $\mu > 0$.

Let $E_0 < 0$ denote the infimum of the continuous spectrum $\sigma_{ess}(H_{cm})$ (i.e. E_0 is the first ionization or fragmentation threshold). Also let $E_1 < 0$ denote the supremum of the discrete spectrum $\sigma_d(H_{cm})$ (which in particular cases can also be an ionization or fragmentation threshold). We note that due to symmetry some of the eigenvalues of H_{cm} can be embedded in the continuous spectrum. Therefore E_1 can be larger than E_0. For example, in case of the helium atom $E_0 = -2$ a.u., whereas $E_1 = -0.5$ a.u.

We are now in a position to state our main result.

Upper bound. Let $\psi \in \mathcal{H}_{cm}$ be a normalized bound state of H_{cm} with energy $E < 0$. Then the ionization satisfies an upper bound of the form

$$I(\psi)^{\frac{1}{2}} \leqslant \int_0^\tau \|(V(t) - V)\psi\|\mathrm{d}t + |c(\tau)|\ \|p_{cm,z}\psi\| + |b(\tau)|\ \|(z - QZ)\psi\|. \tag{3}$$

Alternatively for any pulse $\mathcal{E}(t)$ with $1/2\mu \cdot b(\tau)^2 < E_0 - E$ one has

$$I(\psi)^{\frac{1}{2}} \leqslant \int_0^\tau \|(V(t) - V)\psi\| \mathrm{d}t + |c(\tau)| \, \|p_{cm,z}\psi\| + \frac{b(\tau)}{E_0 - E - \frac{1}{2\mu} b(\tau)^2} \, \|p_{cm,z}\psi\|. \tag{4}$$

Lower bound. Let $\psi \in \mathcal{H}_{cm}$ be a normalized bound state of H_{cm} with energy $E < 0$. Then for any pulse $\mathcal{E}(t)$ with $1/2\mu \cdot b(\tau)^2 > E_1 - E$ the ionization satisfies a lower bound of the form

$$(1 - I(\psi))^{\frac{1}{2}} \leqslant \int_0^\tau \|(V(t) - V)\psi\| \mathrm{d}t + \frac{1}{E - E_1 + \frac{1}{2\mu} b(\tau)^2} \|(V(\tau) - V)\psi\| + \frac{b(\tau)}{E - E_1 + \frac{1}{2\mu} b(\tau)^2} \|p_{cm,z}\psi\|. \tag{5}$$

In particular (5) proves the absence of stabilization.

Since the bounds (4), (5) have the same structure as those obtained in [4, 5], the discussion and interpretation of (3)–(5) in relation to the issue of stabilization may be taken over from there. Also, [12] will contain a more detailed discussion of the influence of pulse shape. We only add a few remarks. First $V(t)$ is again Kato bounded with respect to $H_{0,cm}$. Therefore, $\|(V(t) - V)\psi\|$ may be estimated independently of $\mathcal{E}(t)$ and the first term in (3)–(5) satisfies a bound of the form

$$\int_0^\tau \|(V(t) - V)\psi\| \mathrm{d}t \leqslant C\tau \tag{6}$$

where the constant C depends on the energy E of the bound state, the charges Q_i, the masses M_i and m. A precise form of C is given in the Appendix. In particular for the ionization bounds, obtained by replacing the left-hand side of (6) by the right-hand side, to be nontrivial (i. e. $\leqslant 1$ and $\geqslant 0$ respectively) one has to have a pulse duration satisfying $\tau < 1/C$. The second term in (5) may be treated analogously. We will see below that $(z - QZ)$ defines an operator in $\mathcal{H}_{cm}$. Now $\|(z - QZ)\psi\|$ is bounded due to the O'Connor–Combes–Thomas theorem (see e.g. [22]) and

$$\|p_{cm,z}\psi\| \leqslant c\langle\psi, H_{0,cm}\psi\rangle^{\frac{1}{2}}$$

holds for a suitable constant c depending on m and the M_i and Q_i. By the techniques used in the Appendix, this leads to a bound of the form $\|p_{cm,z}\psi\| \leqslant C'$, where C' is a new constant depending again on E, Q_i, M_i and m.

Many of the pulses discussed in the literature, for example linearly polarized monochromatic light (possibly with a trapezoidal or sine-squared enveloping function) or chirps, typically have $b(\tau) \neq 0$, such that the lower bound indeed becomes relevant (for a more detailed discussion see [12]).

We turn to the proof of these bounds, which is an adaptation of arguments used in [3–5]. Let $U(t, t')$ be the unitary time evolution for the Hamiltonian $H(t)$, i.e. $U(t, t')$ satisfies

$$\begin{aligned} i\partial_t U(t, t') &= H(t)U(t, t') \\ U(t, t')U(t', t'') &= U(t, t'') \\ U(t, t) &= \mathbf{1} \end{aligned}$$

for all t, t', t''. The existence of these unitary operators can be established by arguments similar to those used in [4], which extend results in [20, 21]. Since by assumption $\mathcal{E}(t)$ vanishes unless $0 \leqslant t \leqslant \tau$, we obviously have

$$U(t, t') = \exp -i(t - \tau)H \cdot U(\tau, 0) \cdot \exp it'H$$

for $t > \tau$ and $t' < 0$.

In analogy with scattering theory (see e.g. [5] for a more detailed discussion), one may define the scattering matrix for the pair $(H(\cdot), H)$ as the weak limit

$$S = w - \lim_{\substack{t\to+\infty \\ t'\to-\infty}} \exp\ itH \cdot U(t,t') \cdot \exp -it'H \tag{7}$$

which, by (21) in the present situation, takes the form

$$S = \exp\ i\tau H \cdot U(\tau, 0).$$

In particular S is unitary.

Let $U_{cm}(t,t')$ and $U'(t,t')$ be the analogous time evolution operators for the operators $H_{cm}(t)$ and $H'(t)$ (see (9)), respectively. $U'(t,t')$ can be given explicitly (see e.g. [5]) and is commonly referred to as the Gordon–Volkov solution [23]. By the last relation in (1) we obviously have

$$U(t,t') = U_{cm}(t,t') \otimes U'(t,t'). \tag{8}$$

In analogy to (7) we may define unitary operators S_{cm} and S' resulting from the pairs $(H_{cm}(t), H_{cm})$ and $(H'(t), H_0')$, respectively. Thus we have

$$\begin{aligned} S_{cm} &= \exp\ i\tau H_{cm} \cdot U_{cm}(\tau, 0) \\ S' &= \exp\ i\tau H_0' \cdot U'(\tau, 0). \end{aligned} \tag{9}$$

In particular the scattering matrix S' for the centre of mass motion can be given in closed form and is nontrivial in general unless $Q = 0$. By (1), (8) and (9) we have

$$S = S_{cm} \otimes S'.$$

Now we invoke the unitary Kramers–Henneberger transformation. Suggested by its form in the one-particle theory (see e.g. (11) below), consider the unitary operators on $\mathcal{H}$ given as

$$T(t) = \exp -ia(t)\Omega \cdot \exp -ib(t)z \cdot \exp\ ic(t)p_z.$$

Here Ω is a number given by

$$\Omega = \sum_{i=1}^{N} \frac{Q_i^2}{M_i} + \frac{N'}{m}$$

and p_z can be expressed through the canonically conjugated momenta $p_{i,z}$ and $p'_{j,z}$ of z_i and z'_j as

$$p_z = \sum_{i=1}^{N} \frac{Q_i}{M_i} p_{i,z} - \frac{1}{m} \sum_{j=1}^{N'} p'_{j,z}. \tag{10}$$

Analogously we define the Kramers–Henneberger transformation for the centre of mass motion as

$$T'(t) = \exp -ia(t)\Omega' \cdot \exp -ib(t)QZ \cdot \exp\ ic(t)\frac{Q}{M}P_z \tag{11}$$

where $\Omega' = Q^2/M$. Also P_z is the z-component of the momentum operator $\vec{P}$ canonically conjugate to $\vec{X}$. Thus $\vec{P}$ is nothing but the total momentum operator, i.e.

$$\vec{P} = \sum_{i=1}^{N} \vec{p}_i + \sum_{j=1}^{N'} \vec{p}'_j \tag{12}$$

such that we may write $H_0' = -\frac{1}{2M}\Delta_{\vec{X}} = \frac{\vec{P}^2}{2M}$. Now let $\Omega_{cm} = \Omega - \Omega'$ and

$$p_{cm,z} = p_z - \frac{Q}{M}P_z. \tag{13}$$

As suggested by the notation, $p_{cm,z}$ defines an operator in $\mathcal{H}_{cm}$. The easiest way to see this is to notice that $p_{cm,z}$ commutes with $\vec{X}$ and with $\vec{P}$, which is trivial. By similar arguments $z - QZ$ defines an operator in $\mathcal{H}_{cm}$. Therefore

$$T_{cm}(t) = \exp -ia(t)\Omega' \cdot \exp -ib(t)(z - QZ) \cdot \exp\ ic(t)p_{cm,z}$$

is a unitary operator in $\mathcal{H}_{cm}$ and we have the desired tensor product decomposition

$$T(t) = T_{cm}(t) \otimes T'(t). \tag{14}$$

The relevance of these Kramers–Henneberger transformations is given by the following property. Let $U_1(t,t')$, $U_{1,cm}(t,t')$ and $U_0'(t,t')$ be the time evolution operators for the Hamiltonians

$$\begin{aligned} H_1(t) &= H_0 + V(t) \\ H_{1,cm}(t) &= H_{0,cm} + V(t) = H_1(t) + \frac{1}{2M}\Delta_{\vec{X}} \end{aligned}$$

and H_0', respectively. Then one has

$$\begin{aligned} U(t,t') &= T(t)U_1(t,t')T(t')^{-1} \\ U_{cm}(t,t') &= T_{cm}(t)U_{1,cm}(t,t')T_{cm}(t')^{-1} \\ U'(t,t') &= T'(t)U_0'(t,t')T'(t')^{-1}. \end{aligned} \tag{15}$$

Note that the relations (15) are compatible with the relations (8), (14) and

$$U_1(t,t') = U_{1,cm}(t,t') \otimes U_0'(t,t')$$

as they should be.

Using (9) this gives in particular

$$I(\psi) = \|(\mathbf{1} - P)T_{cm}(\tau)U_{1,cm}(\tau,0)\|^2.$$

The bounds (3), (4) and (5) now follow the line of arguments given in [4] and [5] with the following modifications. In place of the relations (3.11) and (3.28) in [5] we now use the identities

$$\begin{aligned} &\exp\ ib(\tau)z \cdot H \cdot \exp -ib(\tau)z \\ &\quad = \sum_{i=1}^{N} \frac{1}{2M_i}(\vec{p}_i - Q_i b(\tau)e_z)^2 + \sum_{j=1}^{N'} \frac{1}{2m}(\vec{p}_j' + b(\tau)e_z)^2 + V \\ &\quad = H - \left(\sum_{i=1}^{N} \frac{1}{M_i}Q_i \cdot p_{i,z} - \frac{1}{m}\sum_{j=1}^{N'} p_{j,z}'\right) \cdot b(\tau) \\ &\quad + \left(\sum_{i=1}^{N} \frac{Q_i^2}{2M_i} + \frac{N'}{2m}\right)b(\tau)^2 \end{aligned}$$

and

$$\begin{aligned} &\exp\ ib(\tau)QZ \cdot \frac{\vec{P}^2}{2M} \cdot \exp -ib(\tau)QZ \\ &\quad = \frac{1}{2M}(\vec{P} - Qb(\tau)e_z)^2 \\ &\quad = \frac{\vec{P}^2}{2M} - \frac{Q}{M}P_z \cdot b(\tau) + \frac{Q^2}{2M}b(\tau)^2 \end{aligned}$$

such that

$$\exp\ ib(\tau)(z - QZ) \cdot H_{cm} \cdot \exp -ib(\tau)(z - QZ) = H_{cm} - p_{cm,z} \cdot b(\tau) + \frac{1}{2\mu} b(\tau)^2.$$

In particular these relations explain the origin of the mass parameter μ.

Similarly one derives

$$\begin{aligned}\exp -ic(\tau)p_{cm,z} \cdot \exp\ ib(\tau)(z - QZ) \cdot H_{cm} \cdot \exp -ib(\tau)(z - QZ) \cdot \exp\ ic(\tau)p_{cm,z} \\ = H_{0,cm} - p_{cm,z} \cdot b(\tau) + \frac{1}{2\mu} b(\tau)^2 + V(\tau) \\ = H_{cm} - p_{cm,z} \cdot b(\tau) + \frac{1}{2\mu} b(\tau)^2 + V(\tau) - V. \qquad (16)\end{aligned}$$

With the help of these relations, the arguments in [4] and [5] may now be taken almost verbatim to finish the proofs of (3), (4) and (5). As an example we now give the proof of the lower bound (5). In order to obtain a lower bound on

$$I(\psi) = \|(\mathbf{1} - P)T_{cm}(\tau)U_{1,cm}(\tau, 0)\psi\|^2 = 1 - \|PT_{cm}(\tau)U_{1,cm}(\tau, 0)\psi\|^2$$

it suffices to obtain an upper bound for $\|PT_{cm}(\tau)U_{1,cm}(\tau, 0)\psi\|$. First we write

$$\begin{aligned}\|PT_{cm}(\tau)U_{1,cm}(\tau, 0)\psi\| \\ = \|P \exp\ -ib(\tau)(z - QZ) \cdot \exp\ ic(\tau)p_{cm,z} \cdot U_{1,cm}(\tau, 0) \cdot \psi\| \\ \leqslant \|P \exp\ -ib(\tau)(z - QZ) \cdot \exp\ ic(\tau)p_{cm,z} \\ \times \left(U_{1,cm}(\tau, 0) - \exp\ -i\tau H_{cm}\right) \cdot \psi\| \\ + \|P \exp\ -ib(\tau)(z - QZ) \cdot \exp\ ic(\tau)p_{cm,z} \cdot \psi\|. \qquad (17)\end{aligned}$$

The first term on the right-hand side is bounded by

$$\left\|\left(U_{1,cm}(\tau, 0) - \exp\ -i\tau H_{cm}\right)\psi\right\|. \qquad (18)$$

We now invoke Du Hamel's formula to rewrite (18) as

$$\left\| \int_0^\tau U_{1,cm}(\tau, t)[V(t) - V] \exp\ -i(\tau - t)H_{cm} \cdot \psi\ \mathrm{d}t \right\|. \qquad (19)$$

Now we use the unitarity of $U_{1,cm}(\tau, t)$ and the fact that ψ is an eigenstate of H_{cm} to estimate (19) by

$$\int_0^\tau \|(V(t) - V)\psi\| \mathrm{d}t$$

which is the first term on the right-hand side of (5). The second term in (17) is treated as follows. Let $\delta > E_1$ be arbitrary. The operator $P(H_{cm} - \delta)^{-1} \leqslant 0$ is well defined with operator norm $\leqslant (\delta - E_1)^{-1}$. Hence

$$\begin{aligned}\|P \exp\ -ib(\tau)z \cdot \exp\ ic(\tau)p_{cm,z} \cdot \psi\| \\ = \|P(H_{cm} - \delta)^{-1}(H_{cm} - \delta) \exp\ -ib(\tau)z \cdot \exp\ ic(\tau)p_{cm,z} \cdot \psi\| \\ \leqslant \frac{1}{\delta - E_1} \|(H_{cm} - \delta) \exp -ib(\tau)z \cdot \exp ic(\tau)p_{cm,z} \cdot \psi\|.\end{aligned}$$

Now using the relation (16) we obtain

$$\begin{aligned}\|P \exp\ -ib(\tau)z \cdot \exp\ ic(\tau)p_{cm,z} \cdot \psi\| \\ \leqslant \frac{1}{\delta - E_1} \|(V(\tau) - V)\psi\| + \frac{1}{\delta - E_1} \|(E - b(\tau)p_{cm,z} \\ + \frac{1}{2\mu} b(\tau)^2 - \delta)\psi\|. \qquad (20)\end{aligned}$$

We now make the choice

$$\delta = E + \frac{1}{2\mu} b(\tau)^2$$

which by assumption on $b(\tau)$ is larger than E_1 and when inserted into (20) immediately yields the remaining two terms, thus completing the proof of the lower bound.

We finally note that we may replace P in (2) by the projection $P_{\widetilde{E}}$ onto the space spanned by all states in $\mathcal{H}_{cm}$ with energy smaller or equal to $\widetilde{E}$ ($\widetilde{E} > 0$). Then

$$I_{\widetilde{E}}(\psi) = \|(\mathbf{1} - P_{\widetilde{E}}) S_{cm} \psi\|^2$$

measures the ionization probability into the space of states with energy larger than $\widetilde{E}$. By arguments similar to those used above one obtains an upper bound for $I_{\widetilde{E}}(\psi)$ by replacing E in (4) by $E - \widetilde{E}$. This bound is valid under the restriction $b(\tau)^2/2\mu < E_0 + \widetilde{E} - E$. Similary there is a lower bound for $I_{\widetilde{E}}(\psi)$ obtained from (5) again by replacing E by $E - \widetilde{E}$. This bound now is valid under the restriction $b(\tau)^2/2\mu > E_1 + \widetilde{E} - E$. In particular for ultra-short, ultra-intense fields $\mathcal{E}(t)$ with $\tau \to 0$ and $|b(\tau)| \to \infty$ the ionization probability $I_{\widetilde{E}}(\psi)$ tends to 1 for any $\widetilde{E}$. This shows that the absence of stabilization is not a threshold effect.

In conclusion we can say that, according to our arguments, multiparticle systems (like atoms, molecules and ions) do not stabilize when exposed to short ultra-intense laser pulses with large classical momentum transfer. For a more detailed discussion we again refer to [5]. In fact our bounds are structurally similar to the one-electron case. The difference to the one-electron case is that whereas in the one-electron case there is only one continuous channel (i.e. ionization), there are several continuous channels (one-electron ionization, multi-electron ionization, fragmentation and combinations thereof) in the multiparticle case (several electrons and/or atoms). Our proof of absence of stabilization in this context means the statement that the total transition probability to all these channels tends to one.

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Appendix

The aim of this appendix is to provide a bound on $\int_0^\tau \|(V(t) - V)\psi\| \mathrm{d}t$ for any normalized bound state ψ. First we need the following fact for the Coulomb potential $1/|x|$ on the one-particle Hilbert space $L^2(\mathbb{R}^3)$, which may easily be proven using theorems IX.28 and X.15 in [24]. For any $0 < \epsilon < 1$ and all $\phi \in L^2(\mathbb{R}^3)$ in the domain of the Laplacean $-\Delta$, one has

$$\left\| \frac{1}{|x|} \phi \right\| \leqslant \epsilon \| - \Delta \phi\| + B(\epsilon) \|\phi\|$$

with

$$B(\epsilon) = \frac{\pi^2 2^{1/4}}{\epsilon^{3/2}} + 1. \tag{A.1}$$

We make use of this in the following way. For notational convenience we set $Q_i = -1$, $M_i = m$, and $x_i = x'_{i-N}$ for $N+1 \leqslant i \leqslant N+N'$ such that V takes the form

$$V = \sum_{1 \leqslant i < i' \leqslant N+N'} \frac{Q_i Q_{i'}}{|\vec{x}_i - \vec{x}_{i'}|} = \sum_{1 \leqslant i < i' \leqslant N+N'} V_{ii'}.$$

Also we introduce the reduced masses $\mu_{ii'}$ $(i \neq i')$ by

$$\mu_{ii'}^{-1} = \frac{1}{M_i} + \frac{1}{M'_i}.$$

This gives, for $\psi \in \mathcal{H}_{cm}$,

$$\|V_{ii'}\psi\| \leqslant 2\epsilon |Q_i Q_{i'}| \mu_{ii'} \left\| -\frac{\Delta_{ii'}}{2\mu_{ii'}} \psi \right\| + B(\epsilon) |Q_i Q_{i'}| \, \|\psi\|$$

where $\Delta_{ii'}$ is the Laplacean with respect to the relative coordinate $x_i - x_{i'}$ $(1 \leqslant i < i' \leqslant N+N')$.

Since $-\Delta_{ii'}/2\mu_{ii'} \leqslant H_{0,cm}$ and since these two operators commute, by the spectral theorem we have

$$\|V_{ij}\psi\| \leqslant 2\epsilon |Q_i Q_{i'}| \mu_{ii'} \|H_{0,cm}\psi\| + B(\epsilon) |Q_i Q_{i'}| \, \|\psi\|.$$

This gives

$$\|V\psi\| \leqslant 2\epsilon \left(\sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \mu_{ii'} \right) \|H_{0,cm}\psi\| + B(\epsilon) \sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \, \|\psi\|. \tag{A.2}$$

Since $H_{0,cm}$ commutes with the unitary transformation given by any shift $x_i - x_{i'} \mapsto x_i - x_{i'} + a$ $(a \in \mathbb{R}^3)$, we also have

$$\|V(t)\psi\| \leqslant 2\epsilon \left(\sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \mu_{ii'} \right) \|H_{0,cm}\psi\| + B(\epsilon) \sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \, \|\psi\|. \tag{A.3}$$

Therefore (A.2) and (A.3) combined with the trivial estimate

$$\|H_{0,cm}\psi\| \leqslant \|(H_{0,cm} + V)\psi\| + \|V\psi\|$$

give

$$\begin{aligned} \|(V(t) - V)\psi\| &\leqslant \|V(t)\psi\| + \|V\psi\| \\ &\leqslant 4\epsilon \left(\sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \mu_{ii'} \right) \|(H_{0,cm} + V)\psi\| \\ &\quad + 4\epsilon \left(\sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \mu_{ii'} \right) \|V\psi\| \\ &\quad + 2B(\epsilon) \sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \, \|\psi\|. \end{aligned} \tag{A.4}$$

In the same way (A.2) gives

$$\begin{aligned} \|V\psi\| &\leqslant 2\epsilon \left(\sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \mu_{ii'} \right) \|(H_{0,cm} + V)\psi\| \\ &\quad + 2\epsilon \left(\sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \mu_{ii'} \right) \|V\psi\| \\ &\quad + B(\epsilon) \sum_{1 \leqslant i < i' \leqslant N+N'} |Q_i Q_{i'}| \, \|\psi\|. \end{aligned} \tag{A.5}$$

Now fix ϵ such that

$$2\epsilon \sum_{1\leqslant i<i'\leqslant N+N'} |Q_i Q_{i'}|\mu_{ii'} = 1/2. \tag{A.6}$$

Then (A.5) leads to

$$\|V\psi\| \leqslant \|(H_{0,cm}+V)\psi\| + 2B(\epsilon) \sum_{1\leqslant i<i'\leqslant N+N'} |Q_i Q_{i'}|\ \|\psi\|. \tag{A.7}$$

With the choice (A.6) for ϵ and hence for $B(\epsilon)$ (see (A.1)), the insertion of (A.7) into (A.4) gives, for any normalized bound state ψ of $H_{cm} = H_{0,cm} + V$ with energy E, the estimate

$$\int_0^\tau \|(V(t)-V)\psi\|\mathrm{d}t \leqslant 4\tau\Bigg[|E| + \Bigg(\sum_{1\leqslant i<i'\leqslant N+N'} |Q_i Q_{i'}|\Bigg) \cdot\Bigg(\frac{2^{1/4}\pi^2}{\left(\sum_{1\leqslant i<i'\leqslant N+N'} |Q_i Q_{i'}|\mu_{ii'}\right)^{3/2}} + 1\Bigg)\Bigg].$$

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