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On the absence of bound-state stabilization through short ultra-intense fields

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Abstract. We address the question of whether atomic bound states begin to stabilize in the short ultra-intense field limit. We provide a general theory of ionization probability and investigate its gauge invariance. For a wide range of potentials we find an upper and lower bound by non-perturbative methods, which clearly exclude the possibility that the ultra intense field might have a stabilizing effect on the atom. For short pulses we find almost complete ionization as the field strength increases.

1. Introduction

Fermi's golden rule, as one of the central elements in quantum mechanics, has served for many years for the understanding of photoionization rates of atoms in weak radiation fields. Its origin is, however, perturbative and therefore when applying very intense fields (with intensities which are greater or of the order of one atomic unit 3.5×10^{16} W cm^{-2}) one leaves its range of validity. With the advance of laser technology this high-intensity region has become accessible to real experiments in the form of laser pulses of 1 ps or less, at frequencies ranging from the infrared to the ultraviolet [1]. The predictions of atomic ionization rates are of practical importance for instance in the study of gas breakdown [2].

In order to treat the new regime, several alternative approximation methods have been proposed. On one hand [3–5] they are based on a perturbation around the Gordon–Volkov solution [6] of the Schrödinger equation. The question of convergence of these series and their precise range of validity has not yet been put on firm grounds. Despite these problems, these methods have been applied to find numerical solutions for the ionization probabilities. On the other hand there exist a vast number of numerical studies, which make use of numerical solutions of the Schrödinger equation, high-frequency approximations [7] or the Floquet approximation [8]. Most computations have been carried out in one dimension [9, 10], in the hope that the essentials of the full three-dimensional physics are already present in this simplified situation. There exist arguments which put them in question [11, 12], since in comparison with the full three-dimensional situation, they do not account for the full angular dependence and may provide misleading results. Recently there have

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been full three-dimensional computations [13–15, 12, 16, 17]. But the complete problem has not been solved yet and, as pointed out in [18], 'even the simplest one-electron atom in an intense laser field presents too great a challenge for truly *ab initio* numerical work, and a variety of compromises have been developed'. These compromises are partly located in the numerical methods themselves, but partly put constraints onto the physics, such as the introduction of mask functions or the approximation of the continuum by cutting of the high-energetic states, for instance.

Several authors claim to have found the very surprising and counter-intuitive result, that the bound state of the atom stabilizes as the field strength increases [14, 15, 12, 17]. Similar results have also been obtained by many authors for the one-dimensional situation [9]. In fact these findings are so surprising that 'a dramatic shift in viewpoint is required to explain the physics of atoms in very strong laser fields' [19]. We shall comment on these results below and for the moment refer the reader to the review article on these findings by Eberly and Kulander [19] and one by Geltman [20], who takes the opposite point of view that atomic bound states do not stabilize as the field strength is increased and who asserts that the 'conventional interpretation of the theory of the interaction of radiation and atoms is quite sound even in this regime'. The latter point of view is also supported in [21].

Evidence for atomic stabilization in superintense laser fields has also been obtained from the study of several classical dynamical systems [22].

Up to now no data exist for intensities of one atomic unit, i.e. in the high-intensity region for which the theoretical predictions are made, such that the controversy could be settled from the experimental side. So far some experiments exist for lower intensities 10^{13} W cm^{-2}, which provide evidence for some sort of stabilization [23].

The controversy is mainly based on numerical results and a detailed theoretical analysis of the problem which involves analytic expressions only does not exist so far. The main intention of our paper is to provide an alternative approach to the matter. We consider the Schrödinger equation for an atom in a linear polarized electric field,

$$\mathrm{i}\frac{\partial \psi}{\partial t} = \left(-\frac{\Delta}{2} + V + zE(t)\right)\psi,$$

where $E(t)$ stands for the intensity of the field and is supposed to have finite duration (for instance, 1 ps $= 4.17 \times 10^4$ au). We do not specify $E(t)$ in more detail: it can be, for instance, a pulse which contains a number of optical periods (the frequency of which is determined by the frequency of the laser) possibly with some turn-on and turn-off parts. Such kinds of pulses were used in the search for possible suppression of ionization. We note that 1 ps pulses have a duration comparable with a classical Kepler period for the highly excited Rydberg states ($T_K = 2\pi n^3$). Another example is half-cycle pulses with duration of about 500 fs, generated in the experiments of Jones *et al* [24]. However, the maximal intensity reached in these experiments was about 10^{-6} au, so that these pulses are ultimately far from the ultra-intense limit.

We suppose furthermore that the wavefunction $\psi(\vec{x}, t)$ is given by the bound state wavefunction of $-\Delta/2 + V$ before the pulse is turned on. One can easily estimate that relativistic effects might be appreciable as soon as $E(t)$ is so strong that the classical theory predicts electron velocities approaching the speed of light, or more precisely when in atomic units the electric field strength times the frequency is of the order of the fine-structure constant. According to the estimates in [18] this occurs for typical frequencies for laser intensities $E \approx 10^{18}$ W cm^{-2}.

Our results below show that atoms do not become resistant to ionization when exposed to short ultra-intense laser pulses. Our statements are of a qualitative nature, in the sense

that they provide upper and lower bounds and do not predict precise values of the ionization probabilities. The methods we use cover all possible pulses, i.e. also those which are very popular in the literature with smooth turn on and off. Our arguments cover all frequency regimes, including the high frequencies for which stabilization is supposed to occur. For pulses which are not switched on smoothly, our results typically hold for very short times of the order of one atomic unit. With a smooth switch on of the pulse one may extend the region of validity. We provide expressions for two upper bounds, (3.2) and (3.14) valid in the region when $(\int_0^\tau E(t)\,\mathrm{d}t)^2/2$ (the classical energy transfer of the pulse) is smaller than the binding energy and the other valid without restriction. The lower bound holds when $(\int_0^\tau E(t)\,\mathrm{d}t)^2/2 > -E$.

The paper is organized as follows. In section 2 we formulate a general theory of ionization probability and prove its gauge invariance. We also make contact with the various approximation methods based on perturbative expansions. In section 3 we briefly discuss these methods in the context of quantum mechanical one-particle Stark Hamiltonians and provide the proofs for the upper and lower bounds for the ionization probability for a wide range of one-particle potentials, which in particular include all potentials appearing in atomic and molecular physics. In section 4 we state our conclusions. In appendix A we provide an upper bound for the Coulomb potential and in appendix B we optimize this bound for the ground state of the hydrogen atom.

2. The general theory of ionization probability and its gauge invariance

In this section we will give a general discussion of the ionization probability and its gauge invariance. Gauge invariance is of course necessary for observable quantities and it is conventional wisdom for the case of the ionization probability. However, we were not able to locate an explicit reference with a proof and will therefore include a discussion on this issue. We will relate our arguments to familiar concepts in scattering theory and explicitly discuss its relevance in the context of the Stark Hamiltonian. In the last part of this section we will show the gauge covariance of time-dependent perturbation theory. In order to convey the general ideas we will avoid bulky mathematical notations in this section.

Let $H(t)(-\infty < t < \infty)$ be a general time-dependent selfadjoint Hamiltonian in some Hilbert space $\mathcal{H}$ and let $U(t,t')$ denote the resulting time evolution operator from t' to t, i.e. $U(t,t')$ satisfies

$$\begin{aligned} &\mathrm{i}\partial_t U(t,t') = H(t)U(t,t') \\ &U(t,t')U(t',t'') = U(t,t'') \\ &U(t,t) = \mathbf{1} \end{aligned} \tag{2.1}$$

for all t, t', t''. In the context of the Stark Hamiltonians, $U(t,t')$ exists for all t, t' and is unitary (see below).

Assume now that $H(t)$ approaches an operator H_+ for $t \to \infty$ and H_- for $t \to -\infty$, i.e.

$$\begin{aligned} H_+ &= \lim_{t\to\infty} H(t) \\ H_- &= \lim_{t\to-\infty} H(t) \end{aligned} \tag{2.2}$$

holds in a suitable sense. It is important to note that we do not assume H_+ to equal H_-. In fact, for the Stark Hamiltonian in certain gauges, these operators will in general differ (see

below). In analogy to the scattering matrix (see below) we define the abstract S-matrix to be the following weak limit, i.e. the limit for matrix elements (if it exists)

$$S = \lim_{\substack{t\to+\infty \\ t'\to-\infty}} \exp \mathrm{i}t H_+ U(t,t') \exp -\mathrm{i}t' H_- . \tag{2.3}$$

In particular S exists trivially and is unitary if $H(t)$ becomes stationary for all large $|t|$, i.e. if $H(t) = H_+$ for all $t \geqslant t_+$ and $H(t) = H_-$ for all $t \leqslant t_-$ for suitable finite t_-, t_+. In this case we will call $H(t)$ a *finitely pulsed Hamiltonian*. S then takes the form

$$S = \exp \mathrm{i}t H_+ U(t,t') \exp -\mathrm{i}t' H_- \tag{2.4}$$

for all $t \geqslant t_+$ and all $t' \leqslant t_-$. In particular S is then unitary.

Let P_+ be the orthogonal projection onto the subspace spanned by the bound states of H_+. P_- is defined analogously in terms of H_-. Then for any normalized state ψ in the range of P_- its ionization probability is defined to be

$$I(\psi) = \|(\mathbf{1} - P_+)S\psi\|^2. \tag{2.5}$$

Here $\|\psi\|$ denotes the Hilbert space norm, i.e. $\|\psi\|^2 = \langle\psi, \psi\rangle$. For the case when $H_+ = H_-$ this agrees with the definition used in [25–27]. For a finitely pulsed Hamiltonian we have

$$I(\psi) = \|(\mathbf{1} - P_+)U(t_+, t_-)\psi\|^2 \tag{2.6}$$

whenever ψ is a bound state of H_-.

Abstract gauge transformations are now introduced as follows. Let $A(t)$ $(-\infty < t < \infty)$ be a one-parameter family of unitary operators (suitably differentiable in t). If $\psi(t)$ is a solution of the Schrödinger equation

$$\mathrm{i}\partial_t \psi(t) = H(t)\psi(t)$$

then $\psi'(t) = A(t)\psi(t)$ is a solution of the equation

$$\begin{aligned} \mathrm{i}\partial_t \psi'(t) &= \mathrm{i}(\partial_t A(t))\psi(t) + A(t)\mathrm{i}\partial_t \psi(t) \\ &= \mathrm{i}(\partial_t A(t))A(t)^{-1}\psi'(t) + A(t)H(t)A(t)^{-1}\psi'(t) \\ &= H'(t)\psi'(t) \end{aligned} \tag{2.7}$$

with

$$H'(t) = \mathrm{i}(\partial_t A(t))A(t)^{-1} + A(t)H(t)A(t)^{-1} \tag{2.8}$$

being formally selfadjoint. If $U'(t,t')$ is the time evolution operator for $H'(t)$ then obviously

$$U'(t,t') = A(t)U(t,t')A(t')^{-1}. \tag{2.9}$$

We note that the set of all gauge transformations forms a non-commutative group under the obvious multiplication rule $(A_1A_2)(t) = A_1(t)A_2(t)$, with unit $\mathbf{1}(t) = \mathbf{1}$ and inverse $A^{-1}(t) = A(t)^{-1}$. The familiar interaction picture used in scattering theory is now a special case. Indeed, to be more specific assume $H(t)$ to be of the form $H(t) = H_0 + H_I(t)$, where H_0 is the 'free' Hamiltonian and $H_I(t)$ the (possibly time-dependent) interaction Hamiltonian. Set

$$A(t) = \exp \mathrm{i}t H_0. \tag{2.10}$$

Then

$$H'(t) = H'_I(t) \tag{2.11}$$

is the Hamiltonian in the interaction picture with

$$H'_I(t) = \exp \mathrm{i}t H_0 \cdot H_I(t) \cdot \exp -\mathrm{i}t H_0. \tag{2.12}$$

In this case the limit (if it exists)

$$S(H, H_0) = \lim_{\substack{t\to+\infty \\ t'\to-\infty}} U'(t, t') = \lim_{\substack{t\to+\infty \\ t'\to-\infty}} \exp \mathrm{i}t H_0 \cdot U(t, t') \cdot \exp -\mathrm{i}t' H_0 \quad (2.13)$$

is called the scattering matrix (S-matrix) for the pair (H, H_0). Let us elaborate briefly in what sense there is an analogy between the quantity (2.3) and the case of ordinary potential scattering, i.e. where H_0 is $-\Delta/2$ and $H_I(t) = V$ is a potential. In the latter case $H = H_0 + V$ is compared with H_0 in spatial regions far out, i.e. where V is small and where the incoming and outgoing wavepackets are located for large $|t|$. In the case we are interested in below, $H(t)$ is compared with H_+ for large positive times and with H_- for large negative times. To summarize: in one case we compare Hamiltonians for large spatial coordinates and in the other case for large time coordinates.

To apply this general concept of gauge transformations and gauge covariance to our discussion of ionization, assume now in addition that $A(t)$ approaches suitably unitary operators A_+ and A_- when $t \to +\infty$ and $t \to -\infty$, respectively.

Then $H'(t)$ (see (2.8)) approaches

$$H'_+ = A_+ H_+ A_+^{-1} \quad (2.14)$$

and

$$H'_- = A_- H_- A_-^{-1} \quad (2.15)$$

as $t \to +\infty$ and $t \to -\infty$ respectively.

By formal manipulations we therefore have

$$S' = A_+ S A_-^{-1}. \quad (2.16)$$

In particular if $H(t)$ is finitely pulsed and if in addition $A(t)$ is stationary for all large $|t|$, then $H'(t)$ is also finitely pulsed, S' exists, is unitary and (2.16) holds.

In general, by (2.14) and (2.15), if $P'_\pm$ are the orthogonal projections onto the space spanned by the bound states of $H'_\pm$, we have

$$P'_\pm = A_\pm P_\pm A_\pm^{-1}. \quad (2.17)$$

In particular $A_-\psi$ is in the range of P'_- if ψ is in the range of P_-. Inserting (2.16) and (2.17) gives the desired gauge invariance in the form

$$\begin{aligned} I'(A_-\psi) &= \|(\mathbf{1} - P'_+)S' A_-\psi\|^2 \\ &= \|A_+(\mathbf{1} - P_+)A_+^{-1} A_+ S A_-^{-1} A_-\psi\|^2 \\ &= \|(\mathbf{1} - P_+)S\psi\|^2 = I(\psi), \end{aligned} \quad (2.18)$$

since by assumption A_+ is unitary. In the example we will be interested in $A_- = \mathbf{1}$. In this case $H'_- = H_-$ and (2.18) takes the simpler form $I'(\psi) = I(\psi)$. From the proof we see that gauge invariance is an important regulating principle in the following sense. One has to choose the projection P_+ in (2.5) and not P_- in order to obtain gauge invariance.

We now apply these concepts to the theory of the time-dependent Stark Hamiltonian. In order to make notations more transparent we choose a linearly polarized electric field, which, however, does not limit our discussion since more general fields may be simply obtained by replacing $z \to \vec{x}$ and $E(t) \to \vec{E}(t)$ ($\vec{x} \in \mathbb{R}^3$, $\vec{E} \in \mathbb{R}^3$), such that particular other choices, such as for instance circular polarized light, may easily be derived from there. We do, however, assume a dipole approximation, such that the electric field becomes a function only of time

and thus is independent of space. Then using atomic units $\hbar = e = m_e = c\alpha = 1$ we consider on the Hilbert space $L^2(\mathbb{R}^3, \mathrm{d}^3x)$ the three time-dependent Hamiltonians

$$\begin{aligned} H_1(t) &= -\frac{\Delta}{2} + V + zE(t) \\ H_2(t) &= \tfrac{1}{2}(-\mathrm{i}\nabla - b(t)e_z)^2 + V \\ H_3(t) &= -\frac{\Delta}{2} + V(\vec{x} - c(t)e_z). \end{aligned} \tag{2.19}$$

Here V is an arbitrary potential, e_z is the unit vector in the z-direction, and $V(\vec{x} - \vec{y})$ is the shifted potential, i.e. the multiplication operator on wavefunctions given as $(V(\vec{x} - \vec{y})\psi)(\vec{x}) = V(\vec{x}-\vec{y})\psi(\vec{x})$. Also $E(t)$ is the electric field, assumed to vanish unless $0 \leqslant t \leqslant \tau$ (i.e. $t_- = 0$ and $t_+ = \tau > 0$ in the notation above). Apart from this condition the pulse $E(t)$ may be arbitrary. We only make the mathematical restriction, that $E(t)$ is piecewise continuous, which means that the pulse may have jumps and all commonly used enveloping shapes, for instance cosine squared, smooth adiabatic turn on and off, etc, are included. Then the following quantities $a(t), b(t)$ and $c(t)$ are well defined

$$b(t) = \int_0^t E(s)\,\mathrm{d}s \tag{2.20}$$

$$c(t) = \int_0^t b(s)\,\mathrm{d}s = tb(t) - \int_0^t sE(s)\,\mathrm{d}s \tag{2.21}$$

$$a(t) = \tfrac{1}{2}\int_0^t b(s)^2\,\mathrm{d}s. \tag{2.22}$$

Note that $b(\tau)e_z$ describes the classical momentum transfer of the pulse, such that $\frac{1}{2}b(\tau)^2$ is the classical energy transfer. Also $c(\tau)c_z$ is the classical displacement caused by the pulse. Then $H_2(t)$ is obtained from $H_1(t)$ by the gauge transformation

$$A_{2\leftarrow 1}(t) = \exp \mathrm{i}b(t)z.$$

$H_1(t)$ is obtained from $H_3(t)$ by the Kramers–Henneberger transformation [28–30].

$$A_{1\leftarrow 3}(t) = T(t) = \exp -\mathrm{i}a(t) \cdot \exp -\mathrm{i}b(t)z \cdot \exp \mathrm{i}c(t)\,p_z. \tag{2.23}$$

Therefore we call $H_3(t)$ the Hamiltonian in the Kramers–Henneberger gauge. We note that a corresponding transformation in quantum electrodynamics had already been introduced by Pauli and Fierz in 1938 [31] but with a different motivation. The Hamiltonian $H_1(t)$ is usually referred to as the Hamiltonian in the length or electric field gauge, whereas $H_2(t)$ is denoted as the Hamiltonian in the velocity, radiation or Coulomb gauge.

As a consequence $H_2(t)$ is obtained from $H_3(t)$ by the gauge transformation

$$A_{2\leftarrow 3}(t) = A_{2\leftarrow 1}(t)A_{1\leftarrow 3}(t) = \exp -\mathrm{i}a(t) \exp \mathrm{i}c(t)\,p_z. \tag{2.24}$$

The general gauge transformation $A_{j\leftarrow i}(t)$ for $H_i(t) \rightarrow H_j(t)$ is then obtained from the rules

$$\begin{aligned} A_{j\leftarrow i}(t) &= A_{i\leftarrow j}(t)^{-1} \\ A_{j\leftarrow k}(t) &= A_{j\leftarrow i}(t)A_{i\leftarrow k}(t). \end{aligned} \tag{2.25}$$

$H_1(t)$ and $H_2(t)$ are finitely pulsed. Note, however, that in general $H_3(t)$ is *not* finitely pulsed and hence does not always have a proper limit as $t \rightarrow +\infty$. This is due to the fact that $b(t)$ is constant for $t \geqslant \tau$ such that $c(t)$ grows linearly in $t \geqslant \tau$ whenever $b(\tau) \neq 0$, as is apparent from (2.21). Nevertheless the Kramers–Henneberger gauge is quite useful as we shall see below. These observations are related to the fact that $A_{2\leftarrow 1}(t)$ becomes

stationary for large t but in general not $A_{2\leftarrow 3}(t)$ and $A_{1\leftarrow 3}(t)$. Note that by assumption on $E(t)$, $A_{j\leftarrow i}(-\infty) = \mathbf{1}$ and thus

$$H_{1,+} = H_{1,-} = H_{2,-} = H_{3,-} = H_0 + V. \tag{2.26}$$

However, in general $H_{2,+} \neq H_{2,-}$.

The case $V \equiv 0$ is of special interest, since it corresponds to the situation in which the Schrödinger equation admits an exact solution, which is usually referred to as the Gordon–Volkov solution [6]. Call the resulting operators $H_{0,i}(t)$ $(i = 1, 2, 3)$, such that in particular $H_{0,3}(t) = -\Delta/2$. The kernels of the resulting time-evolution operators $U_{0,i}(t, t')$ can be calculated explicitly. Indeed, we start from the familiar relation for the free-particle evolution operator (see e.g. [36])

$$U_{0,3}(\vec{x}, t; \vec{x}', t') = \langle \vec{x}|U_{0,3}(t, t')|\vec{x}'\rangle = \left\langle \vec{x} \left| \exp \mathrm{i}(t - t')\frac{\Delta}{2} \right| \vec{x}' \right\rangle \tag{2.27}$$

$$= \frac{1}{(2\pi \mathrm{i}(t - t'))^{3/2}} \exp \mathrm{i}\frac{(\vec{x} - \vec{x}')^2}{2(t - t')}. \tag{2.28}$$

Obviously

$$\langle \vec{x}| \exp \mathrm{i}b(t)z|\vec{x}'\rangle = \exp \mathrm{i}b(t)z \cdot \delta^3(\vec{x} - \vec{x}')$$
$$\langle \vec{x}| \exp \mathrm{i}a(t)|\vec{x}'\rangle = \exp \mathrm{i}a(t) \cdot \delta^3(\vec{x} - \vec{x}')$$

and

$$\langle \vec{x}| \exp \mathrm{i}c(t)\, p_z|\vec{x}'\rangle = \langle \vec{x} - c(t)e_z, \vec{x}'\rangle = \delta^3(\vec{x} - \vec{x}' - c(t)e_z)$$

such that

$$\langle \vec{x}|T(t)|\vec{x}'\rangle = \overline{\langle \vec{x}'|T(t)^{-1}|\vec{x}\rangle}$$
$$= \exp -\mathrm{i}a(t) \cdot \exp -\mathrm{i}b(t)z \cdot \delta^3(\vec{x} - \vec{x}' - c(t)e_z). \tag{2.29}$$

By (2.9) we immediately obtain the following (again well known) relations (see e.g. [32, 33, 6, 13]).

$$U_{0,1}(\vec{x}, t; \vec{x}', t') = \frac{1}{(2\pi \mathrm{i}(t - t'))^{3/2}} \exp \mathrm{i}(a(t') - a(t))$$
$$\times \exp \mathrm{i}(b(t')z - b(t)z') \exp \mathrm{i}\frac{(\vec{x} - c(t)e_z - \vec{x}' + c(t')e_z)^2}{2(t - t')}$$

$$U_{0,2}(\vec{x}, t; \vec{x}', t') = \frac{1}{(2\pi i(t - t'))^{3/2}} \exp \mathrm{i}(a(t') - a(t)) \exp \mathrm{i}\frac{(\vec{x} - c(t)e_z - \vec{x}' + c(t')e_z)^2}{2(t - t')}.$$

The kernel of $U_{0,1}(t, t')$ is often called the Gordon–Volkov propagator (see e.g. [6, 32, 13]).

We finally give a discussion of time-dependent perturbation theory and its gauge covariance. Returning to the general set-up, let $H(t)$ be a 'perturbation' of $K(t)$. If $W(t, t')$ denotes the time evolution for $K(t)$, we have the generalized Du Hamel's formula (see e.g. [34]) in the form

$$U(t, t') = W(t, t') - \int_{t'}^{t} \frac{\mathrm{d}}{\mathrm{d}s}[U(t, s)W(s, t')]\,\mathrm{d}s$$
$$= W(t, t') - \mathrm{i}\int_{t'}^{t} U(t, s)[H(s) - K(s)]W(s, t')\,\mathrm{d}s. \tag{2.30}$$

We recall that in terms of the sometimes more familiar Green function

$$G_H(t, t') = -\mathrm{i}U(t, t')\theta(t - t'),$$

which satisfies

$$(\mathrm{i}\partial_t - H(t))G_H(t,t') = \delta(t-t')\mathbf{1},$$

relation (2.30) takes the form

$$G_H(t,t') = G_K(t,t') + \int_{-\infty}^{+\infty} G_H(t,s)[H(s)-K(s)]G_K(s,t')\,\mathrm{d}s. \tag{2.31}$$

Similarly one derives the relation

$$U(t,t') = W(t,t') - \mathrm{i}\int_{t'}^{t} W(t,s)[H(s)-K(s)]U(s,t')\,\mathrm{d}s \tag{2.30$'$}$$

and hence

$$G_H(t,t') = G_K(t,t') + \int_{-\infty}^{+\infty} G_K(t,s)[H(s)-K(s)]G_H(s,t')\,\mathrm{d}s. \tag{2.31$'$}$$

We recall that Du Hamel's formula in the form (2.31) and (2.31′) is the time-dependent version of the Lippmann–Schwinger equation (see e.g. [35, 36]) in the case where both $H(s)$ and $K(s)$ are actually time-independent. Indeed, the Lippmann–Schwinger equation may be obtained from these relations by taking Laplace transforms.

The equations (2.30), (2.30′), (2.31) and (2.31′) may be iterated by introducing the left-hand side into the right-hand side, resulting in a 'power series expansion' of $U(t,t')$ in powers of $H(s)-K(s)$ and involving W of the form

$$U(t,t') = \sum_{n=0}^{\infty} U_n(t,t') \tag{2.34}$$

with $U_0(t,t') = W(t,t')$ and with an analogous expansion for G_H. Let us consider what happens under gauge transformations. Du Hamel's formula is compatible with gauge transformations in the sense that the relation

$$U'(t,t') = W'(t,t') - \mathrm{i}\int_{t'}^{t} U'(t,s)[H'(s)-K'(s)]W'(s,t')\,\mathrm{d}s \tag{2.35}$$

either follows directly for the pair $H'(t), K'(t)$ or by applying the gauge transformation to (2.30) and using (2.8) and (2.9). This implies in particular that

$$U_n'(t,t') = A(t)U_n(t,t')A(t')^{-1} \tag{2.36}$$

for all n, which applies in particular to the choices $H(t) = H_i(t)$ and $K(t) = H_{0,i}(t)$ given by (2.19) and the gauge transformations which relate them. However, some of the approximation methods used for high intensities, on which we shall comment more below, use the fact that one can decompose the Stark Hamiltonian in two different ways, that is either treating the potential or the term related to the electric field as a perturbation. Hence two versions of (2.30) are obtained which may be combined iteratively. The series generated in this manner in general does not respect gauge invariance order by order. A discussion of this problem and a remedy for restoring gauge invariance by including some terms of next order, thus leading to cancellations, may be found in [32, 37].

In the case of the interaction picture (see above) and with the choice $K(t) = H_0$ such that $K'(t) = 0$, the iteration of (2.33) in powers of $H_I'(t) = H'(t) - K'(t)$ is just the famous Dyson series of the S-matrix $S(H, H_0)$ in the limit $t \to +\infty$, $t' \to -\infty$. In the time-independent context of the Lippmann–Schwinger equation (see above) this corresponds to iterating the Lippmann–Schwinger equation to obtain the Born series for the S-matrix (see e.g. [36]). In the context we are presently interested in, such series expansions for the

time-evolution operator of finitely pulsed Hamiltonians lead to a series expansion for the ionization probability when inserted in (2.6). In the next section we will discuss the various approaches used so far for the Hamiltonian $H_1(t)$ and its gauge transforms $H_2(t)$ and $H_3(t)$ given in (2.19).

3. Ionization of atoms in strong, short electric fields

Using the notions of the previous section we start with a review and comparison of methods and results obtained by previous authors. Then we relate this in a first step to a new, rigorous upper bound on the ionization probability, valid for all small τ and small classical momentum transfer $b(\tau)$ and small displacement $c(\tau)$ (see below for the upper bound). This result is also compared with another rigorous upper bound previously obtained by two of the authors (VK, RS) [26] as well as with a result obtained using a time–energy uncertainty relation given by Pfeifer [38]. Secondly we prove a lower bound below, valid for all small τ and all large $b(\tau)$, which in particular proves the absence of stabilization.

Taking $H(t) = H_0 + V + zE(t) = H_1(t)$ and $K(t) = H_0 + V$ in (2.30), the resulting perturbation series in the time-dependent interaction $H(t) - K(t) = zE(t)$ for $U_1(t, t')$, the time-evolution operator for $H_1(t)$, has been used by Lambropolous [39]. Certainly for high intensities $E(t)$ this is very problematic, since one requires several terms in the expansion to achieve a reasonable result.

A more promising approach has been advocated by Perelomov *et al* [4], who took $H(t) = H_0+V+zE(t) = H_1(t)$ and $K(t) = H_0+zE(t) = H_{0,1}(t)$. Then $H(t)-K(t) = V$, thus leading to a power-series expansion in V. Whenever $|E| > |V|$ this seems to be a very suggestive approximation and is based on the fact that the time-evolution operator $U_{0,1}(t, t')$ for $H_{0,1}(t)$ is the Gordon–Volkov solution, which is known exactly (see equation after (2.29)). By the discussion of section 2, in the Kramers–Henneberger gauge this corresponds to a power-series expansion in suitable translates of V.

A combination of these two methods has been proposed earlier in a seminal paper on the subject by Keldysh [3], who took the series for $U_1(t, t')$ with $zE(t)$ as a perturbation, but in the second iteration step inserted the time-evolution operator $U_{0,1}$ instead of the time-evolution operator for $H_0 + V$. In fact, it was demonstrated by Davidovich *et al* [32] that, to first order, the Keldysh approximation and the one of Perelomov *et al* [4] precisely coincide. When carrying out the same steps in the velocity gauge, i.e. for $H(t) = H_2(t)$, one obtains the so-called Faisal–Reiss approximation [5].

We want to point out that all such series expansions are somewhat problematic since a proper convergence of the series has not yet been established (the only known case is the Born series in scattering theory at high energies, see e.g. [36] and the references given there. For the one-dimensional situation convergence may be shown for integrable potentials [32]), and it is not straightforward to give precise quantum mechanical estimates of the first terms. Most statements seem to be based on crude semiclassical estimates [40] or in the belief that features which have been observed for relatively simple one-dimensional models, which are anyway put in question [11], carry over in general [13, 10].

For realistic pulses for instance with smooth adiabatic turn on and off, the first terms will only give reasonable results when the full power is reached but will be poor, if not completely invalid near the turn on and turn off point.

We now give a new rigorous upper bound on the ionization probability. The proof of this bound bypasses the problem of summing the whole perturbation series by staying strictly with the Du Hamel formula. In what follows the potential V will be supposed to satisfy the conditions given in [26] which are tailored to ensure the existence of the time-

evolution operators for $H_i(t)$ $(i = 1, 2, 3)$ given in (2.19). In particular such potentials V are Kato small (see e.g. [30, 34]), i.e. there are $a < 1$ and $b < \infty$ such that for all ψ in the domain $\mathcal{D}(H_0)$ of $H_0 = -\Delta/2$

$$\|V\psi\| \leqslant a\|-\Delta\psi\| + b\|\psi\|. \tag{3.1}$$

Also the domain $\mathcal{D}(H)$ of H coincides with $\mathcal{D}(H_0)$. We note that the potentials of atoms or molecules arising from Coulomb pair potentials belong to this wide class. Also all potentials (except the δ-potential) like smoothed or shielded Coulomb potentials used in numerical computations in this context are Kato small. Also Hamiltonians with Kato bounded potentials (3.1) are bounded below and if the (pair) potentials decay suitably at infinity, then there are no positive eigenvalues. Indeed, it has been shown [34, 41] for a large class of potentials including atomic and molecular ones that the eigenvalues are contained in $[\inf\sigma(H), 0]$ where $\sigma(H)$ denotes the spectrum of H. With these specifications on V in mind we are now in the position to state, prove and comment on the first main result of this section.

Upper bound 1. Let ψ be a normalized bound state of $H = H_0 + V$ with energy $E < 0$. Then for any pulse $E(t)$ with $\frac{1}{2}b(\tau)^2 < -E$, the ionization probability satisfies the upper bound

$$I(\psi)^{\frac{1}{2}} \leqslant \int_0^\tau \|(V(\vec{x} - c(t)e_z) - V(\vec{x}))\psi\|\,\mathrm{d}t + |c(\tau)|\|p_z\psi\| + \frac{|b(\tau)|}{-E - \frac{1}{2}b(\tau)^2}\|p_z\psi\|. \tag{3.2}$$

Note that the condition on $E(t)$ just says that the classical energy transfer of the pulse is less than the classical ionization energy.

The proof is based on a combination of arguments used in [25] and [26] and goes as follows. Since

$$\exp -\mathrm{i}tH\psi = \exp -\mathrm{i}tE\psi, \tag{3.3}$$

by using the Kramers–Henneberger transformation we have

$$\begin{aligned} I(\psi)^{\frac{1}{2}} &= \|(\mathbf{1} - P)U_1(\tau, 0)\psi\| = \|(\mathbf{1} - P)T(\tau)U_3(\tau, 0)\psi\| \\ &= \|(\mathbf{1} - P)\exp -\mathrm{i}b(\tau)z \cdot \exp \mathrm{i}c(\tau)p_z U_3(\tau, 0)\psi\| \\ &\leqslant \|(\mathbf{1} - P)\exp -\mathrm{i}b(\tau)z \cdot \exp \mathrm{i}c(\tau)p_z(U_3(\tau, 0) - \exp -\mathrm{i}\tau H)\psi\| \\ &\quad + \|(\mathbf{1} - P)\exp -\mathrm{i}b(\tau)z \cdot \exp \mathrm{i}c(\tau)p_z\psi\|. \end{aligned} \tag{3.4}$$

We start with an estimate of the first term on the r.h.s. of (3.4). Obviously it is bounded by

$$\|(U_3(\tau, 0) - \exp -\mathrm{i}\tau H)\psi\|. \tag{3.5}$$

We now invoke Du Hamel's formula to rewrite (3.5) as

$$\left\| \int_0^\tau U_3(\tau, t)[V(\vec{x} - c(t)e_z) - V(\vec{x})]\exp -\mathrm{i}(\tau - t)H \cdot \psi\,\mathrm{d}t \right\|. \tag{3.6}$$

Now we use the unitarity of $U_3(\tau, t)$ (besides the fact that we never iterate Du Hamel's formula, this is the crucial step in avoiding perturbation theory) and (3.3) to estimate (3.6) by

$$\int_0^\tau \|(V(\vec{x} - c(t)e_z) - V(\vec{x}))\psi\|\,\mathrm{d}t \tag{3.7}$$

which is the first term on the r.h.s. of (3.2). To estimate the second term in (3.4), we use the triangle inequality to obtain

$$\begin{aligned} &\|(\mathbf{1} - P)\exp -\mathrm{i}b(\tau)z| \cdot \exp \mathrm{i}c(\tau)p_z \cdot \psi\| \\ &\qquad \leqslant \|(\exp(\mathrm{i}c(\tau)p_z) - \mathbf{1} \cdot \psi\| + \|(\mathbf{1} - P)\exp -\mathrm{i}b(\tau)z| \cdot \psi\| \end{aligned} \tag{3.8}$$

Now we use the estimate

$$\|(\exp \mathrm{i}A - \mathbf{1})\psi\| \leqslant \|A\psi\|$$

valid for any selfadjoint operator to estimate the first term on the r.h.s. of (3.8) by

$$|c(\tau)|\|p_z\psi\| \tag{3.9}$$

which is the second term on the r.h.s. of (3.2). It remains to estimate the second term in (3.8). By assumption $(\mathbf{1} - P)H \geqslant 0$. Hence for any $\delta > 0$, $(\mathbf{1} - P)(H + \delta)^{-1}$ exists and is norm bounded by $1/\delta$. Therefore

$$\begin{aligned}\|(\mathbf{1} - P)\exp -\mathrm{i}b(\tau)z\psi\| &= \|(\mathbf{1} - P)(H + \delta)^{-1}(H + \delta)\exp -\mathrm{i}b(\tau)z\psi\| \\ &= \|(\mathbf{1} - P)(H + \delta)^{-1}\exp -\mathrm{i}b(\tau)z \exp \mathrm{i}b(\tau)z(H + \delta)\exp -\mathrm{i}b(\tau)z\psi\| \\ &\leqslant \frac{1}{\delta}\|\exp \mathrm{i}b(\tau)z(H + \delta)\exp -\mathrm{i}b(\tau)z\psi\|. \end{aligned} \tag{3.10}$$

We now use the fact that

$$\exp \mathrm{i}b(\tau)z\,H \exp -\mathrm{i}b(\tau)z = \tfrac{1}{2}(-\mathrm{i}\nabla - b(\tau)e_z)^2 + V = H - b(\tau)p_z + \tfrac{1}{2}b(\tau)^2. \tag{3.11}$$

Inserting this into (3.10) gives

$$\|(\mathbf{1} - P)\exp -\mathrm{i}b(\tau)z\psi\| \leqslant \frac{1}{\delta}\|(E - b(\tau)p_z + \tfrac{1}{2}b(\tau)^2 + \delta)\psi\| \tag{3.12}$$

Making the choice

$$\delta = -E - \tfrac{1}{2}b(\tau)^2 \tag{3.13}$$

which by assumption on $b(\tau)$ is > 0 and inserting into (3.12) gives the third term in (3.2) concluding the proof of the upper bound.

We now comment on this result.

Inspection of the proof of the main result in [26] shows that one has the alternative

Upper bound 2.

$$I(\psi)^{\frac{1}{2}} \leqslant \int_0^\tau \|(V(\vec{x} - c(t)e_z) - V(\vec{x}))\psi\|\,\mathrm{d}t + |c(\tau)|\|p_z\psi\| + |b(\tau)|\|z\psi\| \tag{3.14}$$

which differs from (3.2) only in the last term. Typically near threshold, i.e. for small $|E|$, both $\|z\psi\|$ and $1/(-E - \frac{1}{2}b(\tau)^2)$ become large, whereas

$$\|p_z\psi\| = \langle\psi, p_z^2\psi\rangle^{\frac{1}{2}} \leqslant (2\langle\psi, H_0\psi\rangle)^{\frac{1}{2}}$$

stays finite. Thus for V being the Coulomb potential

$$\langle\psi, H_0\psi\rangle = -E \tag{3.15}$$

by the virial theorem (see e.g. [42]). For s states ψ_{n00} can be improved slightly since $\|p_z\psi_{n00}\|^2 = \frac{2}{3}\langle\psi_{n00}, H_0\psi_{n00}\rangle = \frac{1}{3n^2}$. Similarly one has $\|z\psi\|^2 \leqslant \langle\psi, r^2\psi\rangle (= \frac{n^2}{2}[5n^2 + 1 - 3\ell(\ell + 1)]$ if $\psi = \psi_{n\ell m}$, see e.g. [43]). Again for s states $\|z\psi\|^2 = \frac{1}{3}\langle\psi, r^2\psi\rangle$, which is a slight improvement. Therefore (3.2) and (3.14) are essentially equivalent.

We now discuss the first two terms in (3.2) and (3.14). In general, for Kato bounded potentials $V(\vec{x})(-\Delta + \mathbf{1})^{-1}$ is a bounded operator. Also since $-\Delta$ is translation invariant, we have

$$\|V(\vec{x} - \vec{y})(-\Delta + \mathbf{1})^{-1}\| = \|V(\vec{x})(-\Delta + \mathbf{1})^{-1}\|. \tag{3.16}$$

In particular for the choice of the Coulomb potential, we prove in appendix A that

$$\left\|\frac{1}{r}(-\Delta + \mathbf{1})^{-1}\right\| \leqslant 6.35. \tag{3.17}$$

Thus for the general potentials V considered, the first term in (3.2) and (3.14) is bounded by

$$\begin{aligned}\tau\|V(\vec{x})(-\Delta+\mathbf{1})^{-1}\|\ \|(-\Delta+\mathbf{1})\psi\|+\tau\|V\psi\| \\ =\tau\|V\psi\|+\tau\|V(\vec{x})(-\Delta+\mathbf{1})^{-1}\|\|(2H_0+\mathbf{1})\psi\|\end{aligned} \tag{3.18}$$

which involves $E(t)$ only through its duration but not its strength. For the eigenfunctions $\psi_{n\ell m}$ of the hydrogen atom, one has (see e.g. [43])

$$\begin{aligned}\|(2H_0+\mathbf{1})\psi_{n\ell m}\|^2 &= \|((2H+\mathbf{1})-2V)\psi_{n\ell m}\|^2 \\ &= (2E_n+1)^2+2(2E_n+1)\left\langle\psi_{n\ell m},\frac{1}{r}\psi_{n\ell m}\right\rangle+4\left\langle\psi_{n\ell m},\frac{1}{r^2}\psi_{n\ell m}\right\rangle \\ &= 1-\frac{1}{n^4}+\frac{4}{n^3(\ell+\frac{1}{2})}.\end{aligned} \tag{3.19}$$

This quantity is $\leqslant 8$ and behaves like $1+\mathcal{O}(\frac{1}{n^3})$ for n large uniformly in $0\leqslant\ell\leqslant n-1$. Hence the r.h.s. of (3.18) for bound states of the hydrogen atom is bounded by 19.4τ uniformly in n and by 6.35τ for all large n.

For the ground state ψ_{100} of the hydrogen atom the first term in (3.2) and (3.14) has a much better estimate. As shown in appendix B

$$\|(V(\vec{x}-\vec{y})-V(\vec{x}))\psi_{100}\|\leqslant 2 \tag{3.20}$$

holds for all $\vec{y}\in\mathbb{R}^3$ such that the first term in (3.2) and (3.14) is now bounded by 2τ.

By a theorem of Pfeifer [38] for the survival probability $|\langle\psi,\psi_\tau\rangle|$ of a state with $\psi_\tau=U(\tau,0)\psi$ for any time-dependent Hamiltonian $H(t)$ one has for all small τ (see [38] for precise conditions)

$$|\langle\psi,\psi_\tau\rangle|\geqslant\cos\left(\int_0^\tau\Delta(t)\,\mathrm{d}t\right) \tag{3.21}$$

where in the present context with $H(t)=H_1(t)$ (see (2.19))

$$\begin{aligned}&\Delta(t)=|E(t)|a_\psi \\ &a_\psi=(\|z\psi\|^2-\langle\psi,z\psi\rangle^2)^{\frac{1}{2}}\leqslant\|z\psi\|.\end{aligned} \tag{3.22}$$

This gives for the ionization probability

$$\begin{aligned}I(\psi)\leqslant 1-|\langle\psi,\psi_\tau\rangle|^2\leqslant a_\psi^2\left(\int_0^\tau|E(t)|\,\mathrm{d}t\right)^2 \\ \leqslant\left(\int_0^\tau|E(t)|\,\mathrm{d}t\right)^2\|z\psi\|^2.\end{aligned} \tag{3.23}$$

This may be compared with the discussion above. Equation (3.23) is weaker than (3.2) and (3.14) in the sense that it does not show independence of the field strength when $b(\tau)$ and $c(\tau)$ are small or even zero. Otherwise it is basically equivalent to (3.2) and (3.14) or even stronger whenever the last two terms there dominate. We note that the rigorous bound (3.23) may be compared with first-order perturbation theory in $E(t)$ which gives

$$I^{(1)}_{\mathrm{pert}}=\left(\int_0^\tau E(t)\,\mathrm{d}t\right)^2\|Pz\psi\|^2\leqslant\left|\int_0^\tau E(t)\,\mathrm{d}t\right|^2\|z\psi\|^2. \tag{3.24}$$

We now turn to a comparison with other approximation methods based on perturbative expansions used in this context. Since all these approximations resolve around the same principle, i.e. an expansion involving the Gordon–Volkov time-evolution operator

$U_{0,1}(t,t')$, we will mainly concentrate on a recent work by Geltman [13], who presented an explicit, partly analytical, partly numerical analysis of the full three-dimensional hydrogen atom. Also a full discussion of other works may be found there. Geltman employs the approximation method of Perelomov *et al* [4] in order to compute the excitation and ionization rates for the 1s, 2s, 3s, 2p, 3p, 3d states of the hydrogen atom struck by a linearly polarized monochromatic laser pulse of the form $E(t) = E_0 \cos \omega t$. The value for the electric field strength in atomic units is chosen to be $E_0 = 5, 10, 20$ and the frequency $\omega = 1.5$. Geltman obtained the following general features.

(a) At integer cycles, that is for $\tau = 2\pi n/\omega$ the rate of ionization becomes independent of the electric field strength. In particular for the non-s states it goes to zero.

This is reflected qualitatively in our results in the following way. At integer cycles $b(\tau) = c(\tau) = 0$ such that the last two terms in (3.2) and (3.14) vanish. Also the first term is independent of the field strength, however, not zero and by the above discussion too large for the above choices of E_0 and ω.

(b) The maxima of $b(\tau)$ and $c(\tau)$ are located at half-integer cycles, i.e. $\tau = 2\pi(n+\frac{1}{2})/\omega$.

For the applied pulse the bounds (3.2) and (3.14) also reproduce this feature qualitatively but again we emphasize that these bounds hold in more generality for all Kato potentials and all states.

Stabilization for strong, short electric fields has been a highly controversial issue with disagreeing results between numerous authors on one hand, for a review see [20], as well as on the other hand, see [19] for a review on these.

The following result shows absence of stabilization for sufficiently strong, short pulses, namely when $b(\tau)$ becomes large and τ small the ionization probability is close to 1.

Lower bound. Let ψ be a normalized bound state of $H = H_0 + V$ with energy $E < 0$. Then for any pulse $E(t)$ with $\frac{1}{2}b(\tau)^2 > -E$ the ionization probability satisfies a lower bound of the form

$$I(\psi) \geqslant 1 - \left\{ \int_0^\tau \|(V(\vec{x} - c(t)e_z) - V(\vec{x}))\psi\| \, \mathrm{d}t + \frac{1}{E + \frac{1}{2}b(\tau)^2} \|(V(\vec{x} - c(\tau)e_z) - V(\vec{x}))\psi\| + \frac{|b(\tau)|}{E + \frac{1}{2}b(\tau)^2} \|p_z \psi\| \right\}^2. \quad (3.25)$$

Note that now the condition on $E(t)$ is that the classical energy transfer of the pulse is larger than the classical ionization energy. Recall that by our previous discussion the norms appearing in the first two terms in the bracket may be estimated independently of the field strength, such that (3.25) gives a bound which involves $E(t)$ only through τ and $b(\tau)$.

We turn to a proof. In order to obtain a lower bound on

$$I(\psi) = \|(\mathbf{1} - P)U_1(\tau, 0)\psi\|^2 = 1 - \|PU_1(\tau, 0)\psi\|^2$$

it suffices to obtain an upper bound on $\|PU_1(\tau, 0)\psi\|$. First we write

$$\begin{aligned} \|PU_1(\tau, 0)\psi\| &= \|PT(\tau)U_3(\tau, 0)\psi\| \\ &= \|P \exp -\mathrm{i}b(\tau)z \exp \mathrm{i}c(\tau)p_z U_3(\tau, 0)\psi\| \\ &\leqslant \|P \exp -\mathrm{i}b(\tau)z \exp \mathrm{i}c(\tau)p_z (U_3(\tau, 0) - \exp -\mathrm{i}\tau H)\psi\| \\ &\quad + \|P \exp -\mathrm{i}b(\tau)z \exp \mathrm{i}c(\tau)p_z \psi\|. \end{aligned} \quad (3.26)$$

The first term on the r.h.s. is estimated by (3.5) yielding the first term in the bracket in (3.25).

The second term in (3.26) is treated as follows. By assumption $PH \leqslant 0$. Let $\delta > 0$ be arbitrary. Then $P(H-\delta)^{-1}$ is a well defined operator with operator norm $\leqslant 1/\delta$. Hence

$$\begin{aligned}\|P\exp -\mathrm{i}b(\tau)z\exp \mathrm{i}c(\tau)p_z\psi\| &= \|P(H-\delta)^{-1}(H-\delta)\exp -\mathrm{i}b(\tau)z\cdot\exp \mathrm{i}c(\tau)p_z\cdot\psi\| \\ &\leqslant \frac{1}{\delta}\|(H-\delta)\exp -\mathrm{i}b(\tau)z\cdot\exp \mathrm{i}c(\tau)p_z\cdot\psi\|. \end{aligned} \tag{3.27}$$

In analogy to (3.11) we now use the relation

$$\begin{aligned}&\exp -\mathrm{i}c(\tau)p_z\cdot\exp \mathrm{i}b(\tau)z\cdot H\cdot\exp -\mathrm{i}b(\tau)z\cdot\exp \mathrm{i}c(\tau)p_z \\ &\qquad = \tfrac{1}{2}(-\mathrm{i}\nabla - b(\tau)e_z)^2 + V(\vec{x}-c(\tau)e_z) \\ &\qquad = H - b(\tau)p_z + \tfrac{1}{2}b(\tau)^2 + V(\vec{x}-c(\tau)e_z) - V(\vec{x}). \end{aligned} \tag{3.28}$$

Inserting this into (3.27) we obtain

$$\begin{aligned}\|P\exp -\mathrm{i}b(\tau)z\cdot\exp \mathrm{i}c(\tau)p_z\cdot\psi\| &\leqslant \frac{1}{\delta}\|(V(\vec{x}-c(\tau)e_z)-V(\vec{x})\psi\| \\ &\quad +\frac{1}{\delta}\|(E-b(\tau)p_z+\tfrac{1}{2}b(\tau)^2-\delta)\psi\|. \end{aligned} \tag{3.29}$$

We now make the choice

$$\delta = E + \tfrac{1}{2}b(\tau)^2 \tag{3.30}$$

which by assumption on $b(\tau)$ is > 0 and when inserted into (3.29) immediately yields the remaining two terms in the bracket of (3.25), thus concluding the proof of the lower bound.

We note that these two theorems are compatible with the result in [25] on the Stark kick, i.e. $E(t) = F_0\delta(t)$. There it was shown that for fixed ionization probability of any bound state $\psi_{n\ell m}$ of the hydrogen atom F_0 scales like $1/n$, as predicted by Reinhold *et al* [44].

We now return to a comparison between our results and those obtained by employing approximation methods based on perturbative expansions and we will include the lower bound in the discussion. We stress once more the point that the lower bound definitely excludes the possibility of stabilization of the bound states for increasing electric field strength, when the applied pulse is short in duration, since (3.7), despite the dependence on $c(t)$, may be estimated by a constant, say C, independent of the electric field. Hence

$$\lim_{|E(t)|\to\infty} I(\psi) \geqslant 1 - \tau^2 C. \tag{3.31}$$

This shows clearly that the electric field has no stabilizing effect and we are therefore in disagreement with [12, 15, 14]. The lower bound also reproduces the result obtained through an expansion around the Gordon–Volkov solution, namely for monochromatic linearly polarized laser pulses at integer cycles, i.e. $b(\tau) = c(\tau) = 0$, the ionization probability becomes independent of the electric field strength. We may qualitatively relate the term proportional to τ^2 to a term also observed in perturbative expansion methods and which is interpreted as the spreading of the wave.

To illustrate our results further, we consider now the concrete example of the hydrogen atom. For the ψ_{100} state we obtain as our best estimate

$$I(\psi_{100}) \leqslant \left(2\tau + |b(\tau)| + \frac{1}{\sqrt{3}}|c(\tau)|\right)^2 \tag{3.32}$$

$$I(\psi_{100}) \geqslant 1 - \left(2\tau + \frac{4}{b(\tau)^2-1} + \frac{2}{\sqrt{3}}\frac{|b(\tau)|}{b(\tau)^2-1}\right)^2. \tag{3.33}$$

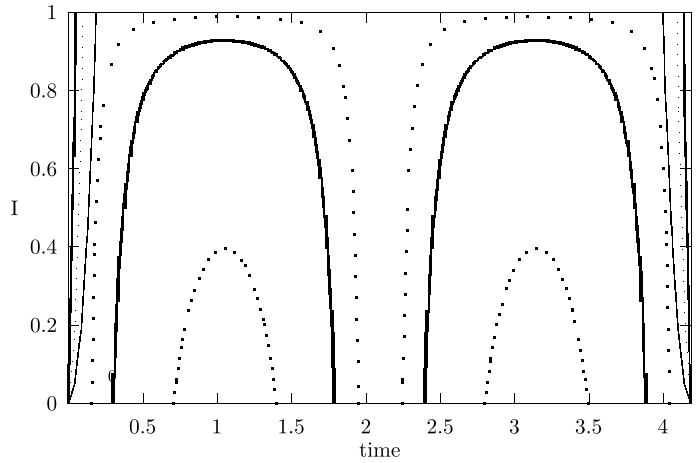

Figure 1. Upper and lower bounds for the ionization probability for the Ψ_{100} state of the hydrogen atom in the first cycle of an applied field $E_z = E_0 \cos(1.5t)$ for $E_0 = 5, 10, 20$, when neglecting the 'spreading of the wave'.

Taking the pulse to be of the form $E_z(t) = E_0 \cos \omega t$ for $0 \leqslant t \leqslant \tau$ and zero otherwise, we have

$$|b(\tau)| = \frac{E_0}{\omega} |\sin \omega\tau| \qquad \text{and} \qquad |c(\tau)| = \frac{2E_0}{\omega^2} \sin^2\left(\frac{\omega\tau}{2}\right). \tag{3.34}$$

Figure 1 shows a plot of one cycle, when neglecting the term which is independent of the electric field strength, i.e. the term 2τ, in the upper (3.32) and lower (3.33) bound. At the far ends one observes the curves for the upper bounds, which approach the vertical for increasing E_0. That is starting from the outside and going inwards the full curve $\equiv E_0 = 20$, dotted curve $\equiv E_0 = 10$ and the next full curve $\equiv E_0 = 5$. Next we have the lower bounds for $E_0 = 20 \equiv$ dotted curve, $E_0 = 10 \equiv$ full curve and $E_0 = 5 \equiv$ dotted curve. We have used the same values as in [13] and compare with figure 3 therein. Figure 1 clearly reproduces the features of Geltman's results, indicating the dips at the half cycles and producing an increasing ionization probability for increasing field strength. However, we do not observe any crossing for different field intensities. When including the 2τ-term, this pattern will be moved above the trivial bound 1. This may be avoided when achieving a better estimate for the factor in front of τ, for instance when integrating explicitly (3.7) for a given pulse [45]. Figure 2 shows the upper bound for four cycles and reproduces the well known oscillatory behaviour superimposed by a spreading of the wavepacket of the Gordon–Volkov solution, the so-called over-the-barrier ionization.

4. Conclusions

In conclusion we can say that, according to our arguments, atoms do not become resistant to ionization when exposed to short ultra-intense laser pulses. We therefore disagree with the opposite point of view, which is sustained through numerous numerical studies partly based on explicit solutions of the Schrödinger equation and partly based on perturbative methods. In particular we have commented above on the problems of the latter methods. It is not the intention of this paper to discuss the problems of numerical methods, but we would like to remark that those studies are in general very complex and subject to many

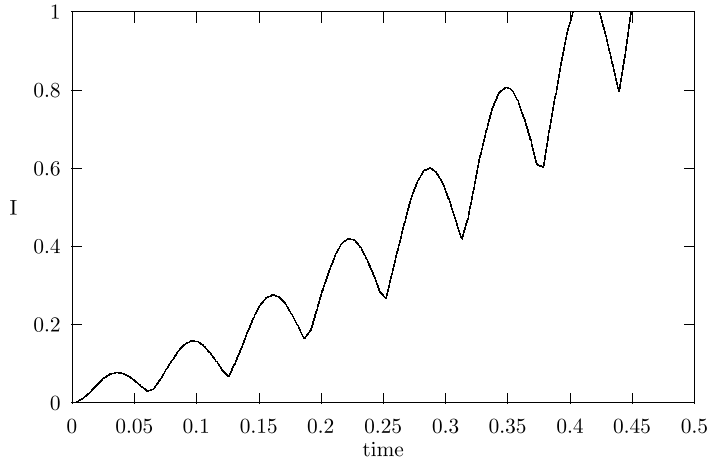

Figure 2. Upper bound for the ionization probability for the ψ_{100} state of the hydrogen atom in the first four cycles of an applied field $E_z = 10\cos(50t)$.

possible errors which are difficult to check for second parties. We think that the virtue of our arguments is that they are analytic and transparent to the reader. An extension of our results to multiparticle systems (thus including atoms and molecules with several electrons and not necessarily electrically neutral) may be found in [27]. Needless to say, since our results are of a qualitative nature, in the sense that they merely provide bounds and that, since there are no explicit solutions for the Schrödinger equation available, for precise predictions of ionization rates one needs more numerical data.

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Appendix A

The aim of this appendix is to prove the following bound for the Coulomb potential $V(\vec{x}) = -\frac{1}{r}(r = |\vec{x}|)$ on $L^2(\mathbb{R}^3, \mathrm{d}^3x) = L^2$.

Lemma A. The following operator norm bound holds

$$\left\| \frac{1}{r}(-\Delta + \mathbf{1})^{-1} \right\| \leqslant 6.35. \tag{A.1}$$

The proof optimizes well known *a priori* bounds which we take from [34].

Let L^∞ be the space of all Lebesgue measurable functions φ on $\mathbb{R}^3$ such that $|\varphi(\vec{x})| \leqslant M < \infty$ almost everywhere. The smallest such M is denoted by $\|\varphi\|_\infty$. Then one has the *a priori* estimate ([34] p 56)

$$\|\varphi\|_\infty \leqslant a(\rho)\| - \Delta\varphi\|_2 + b(\rho)\|\varphi\|_2 \tag{A.2}$$

where $\|\ \|_2$ denotes the L^2 norm, i.e. $\|\varphi\|_2 = \int |\varphi(\vec{x})^2|\, \mathrm{d}^3x$. Here

$$\begin{aligned} a(\rho) &= c\rho^{-1} \\ b(\rho) &= c\rho^3 \end{aligned} \tag{A.3}$$

with

$$c = \left(\int_{-\infty}^{+\infty} \frac{1}{(1+\lambda^2)^2}\, \mathrm{d}\lambda \right)^{\frac{1}{2}} = \sqrt{\frac{\pi}{2}} \tag{A.4}$$

and $\rho > 0$ may be chosen arbitrarily.

Let $R > 0$ be arbitrary and write

$$\frac{1}{r} = V_1^R + V_2^R$$

with

$$\begin{aligned} V_1^R(r) &= \frac{\theta(r < R)}{r} \\ V_2^R(r) &= \frac{\theta(r \geqslant R)}{r}. \end{aligned}$$

Then one has the *a priori* bound (see [34] p 165)

$$\left\| \frac{1}{r}\varphi \right\|_2 \leqslant a(\rho)\|V_1^R\|_2\| - \Delta\varphi\|_2 + (b(\rho) + \|V_2^R\|_\infty)\|\varphi\|_2. \tag{A.5}$$

Since the estimates $\|\varphi\|_2 \leqslant \|(-\Delta + \mathbf{1})\varphi\|_2$, $\| - \Delta\varphi\|_2 \leqslant \|(-\Delta + \mathbf{1})\varphi\|_2$ are trivially valid, this gives

$$\left\| \frac{1}{r}(-\Delta + \mathbf{1})^{-1} \right\| \leqslant a(\rho)\|V_1^R\|_2 + b(\rho) + \|V_2^R\|_\infty. \tag{A.6}$$

Obviously

$$\begin{aligned} \|V_1^R\|_2 &= (4\pi R)^{\frac{1}{2}}, \\ \|V_2^R\|_\infty &= \frac{1}{R}. \end{aligned} \tag{A.7}$$

Inserting (A.3), (A.4) and (A.7) into (A.6) gives

$$\left\| \frac{1}{r}(-\Delta + \mathbf{1})^{-1} \right\| \leqslant \pi\sqrt{2}\rho^{-1}R^{\frac{1}{2}} + \sqrt{\frac{\pi}{2}}\rho^3 + \frac{1}{R}. \tag{A.8}$$

for all $\rho > 0$, $R > 0$. The claim now follows by optimizing w.r.t. ρ and R. A short calculation finally gives

$$\left\| \frac{1}{r}(-\Delta + \mathbf{1})^{-1} \right\| \leqslant 11\frac{\pi^{\frac{7}{11}}}{2^{\frac{6}{11}}3^{\frac{9}{11}}} \tag{A.9}$$

which is (A.1).

Appendix B

In this section we will study the quantity $\langle\psi|V(\vec{x}-\vec{y})^k|\psi\rangle$ for $k=1$ and 2 where $V(\vec{x})=-\frac{1}{|\vec{x}|}(\vec{x}\in\mathbb{R}^3)$ is the Coulomb potential and where ψ is the normalized ground-state wavefunction ψ_{100} for the hydrogen atom, which is rotationally invariant. Therefore this quantity depends on $|\vec{y}|$ only.

Lemma B. Both $-\langle\psi|V(\vec{x}-\vec{y})|\psi\rangle$ and $\langle\psi|V(\vec{x}-\vec{y})^2|\psi\rangle$ are decreasing functions of $|\vec{y}|$ for $\psi=\psi_{100}$.

Intuitively this result is clear: $\psi(\vec{x})$ has its maximum at $\vec{x}=0$ and $-V(\vec{x}-\vec{y})\geqslant 0$ has its singularity at $\vec{x}=\vec{y}$ so their overlap is maximal when $\vec{y}=0$.

Before we give a proof, we first establish an important consequence. Indeed we claim that for $\psi=\psi_{100}$

$$\|(V(\vec{x}-\vec{y})-V(\vec{x}))\psi\|\leqslant 2 \tag{B.1}$$

for all $\vec{y}\in\mathbb{R}^3$. To see this we write

$$\begin{aligned}\|(V(\vec{x}-\vec{y})-V(\vec{x}))\psi\|^2 &= \langle\psi, V(\vec{x}-\vec{y})^2\psi\rangle - 2\langle\psi, V(\vec{x}-\vec{y})V(\vec{x})\psi\rangle + \langle\psi, V(\vec{x})^2\psi\rangle\\ &\leqslant \langle\psi, V(\vec{x}-\vec{y})^2\psi\rangle + \langle\psi, V(\vec{x})^2\psi\rangle\end{aligned} \tag{B.2}$$

since $V(\vec{x}-\vec{y})V(\vec{x})\geqslant 0$ as an operator. By lemma B, the r.h.s. of (B.2) takes its maximum at $\vec{y}=0$ proving the claim since $\langle\psi|\frac{1}{|\vec{x}|^2}|\psi\rangle=2$ (see e.g. [43]).

To prove the lemma, it suffices to consider $\vec{y}$ to be of the form $\vec{y}=ce_z$ with $c>0$. Now we use the well known formula

$$\frac{1}{|\vec{x}-ce_z|}=\frac{1}{r_>}\sum_{\ell=0}^{\infty}\left(\frac{r_<}{r_>}\right)^{\ell}P_\ell(\cos\vartheta) \tag{B.3}$$

where $r_>=\max(r,c)$, $r_<=\min(r,c)$ and where $(r=|\vec{x}|,\vartheta,\varphi)$ are the polar coordinates of $\vec{x}$. By the orthogonality relations of the Legendre polynomials and since ψ is the ground state we obtain

$$\begin{aligned}-\langle\psi|V(\vec{x}-ce_z)|\psi\rangle &= \langle\psi|\frac{1}{r_>}|\psi\rangle\\ &= 4\int_0^\infty r^2\mathrm{e}^{-2r}\frac{1}{r_>}\,\mathrm{d}r\\ &= \frac{4}{c}\int_0^c r^2\mathrm{e}^{-2r}\,\mathrm{d}r+4\int_c^\infty r\mathrm{e}^{-2r}\,\mathrm{d}r.\end{aligned} \tag{B.4}$$

This function is differentiable in c for $c>0$ and its derivative is easily seen to be $\leqslant 0$, proving the first claim. Next we have (again by the orthogonality of the Legendre polynomials)

$$\begin{aligned}\langle\psi|V(\vec{x}-ce_z)^2|\psi\rangle &= \left\langle\psi\left|\frac{1}{(r_>)^2}\sum_{\ell=0}^{\infty}\left(\frac{r_<}{r_>}\right)^{2\ell}\frac{1}{2\ell+1}\right|\psi\right\rangle\\ &= \frac{4}{c^2}\int_0^c r^2\mathrm{e}^{-2r}\sum_{\ell=0}^{\infty}\left(\frac{r}{c}\right)^{2\ell}\frac{1}{2\ell+1}\,\mathrm{d}r+4\int_c^\infty\mathrm{e}^{-2r}\sum_{\ell=0}^{\infty}\left(\frac{c}{r}\right)^{2\ell}\frac{1}{2\ell+1}\,\mathrm{d}r.\end{aligned} \tag{B.5}$$

Now for $0<x<1$ we have

$$\sum_{\ell=0}^{\infty}x^{2\ell}\frac{1}{2\ell+1}=\frac{1}{x}\sum_{\ell=0}^{\infty}\frac{x^{2\ell+1}}{2\ell+1}=\frac{1}{2x}(\ln(1+x)-\ln(1-x)). \tag{B.6}$$

Inserting (B.6) into (B.5) gives

$$\langle\psi|V(\vec{x}-ce_z)^2|\psi\rangle = \frac{2}{c}\int_0^c r\mathrm{e}^{-2r}\left(\ln\left(1+\frac{r}{c}\right)-\ln\left(1-\frac{r}{c}\right)\right)\mathrm{d}r + \frac{2}{c}\int_c^\infty r\mathrm{e}^{-2r}\left(\ln\left(1+\frac{c}{r}\right)-\ln\left(1-\frac{c}{r}\right)\right)\mathrm{d}r. \tag{B.7}$$

Now the r.h.s. is not differentiable in c. To remedy this we regularize and consider the quantity $(0<\varepsilon<1)$

$$0 \leqslant F(c,\varepsilon) = \frac{G(c,\varepsilon)}{c} = \frac{2}{c}\int_0^c r\mathrm{e}^{-2r}\left[\ln\left(1+\frac{r}{c}\right)-\ln\left(1-\frac{r}{c}(1-\varepsilon)\right)\right]\mathrm{d}r + \frac{2}{c}\int_c^\infty r\mathrm{e}^{-2r}\left[\ln\left(1+\frac{c}{r}\right)-\ln\left(1-\frac{c}{r}(1-\varepsilon)\right)\right]\mathrm{d}r. \tag{B.8}$$

Since $\lim_{\varepsilon\to 0} F(c,\varepsilon) = \langle\psi|V(\vec{x}-ce_z)^2|\psi\rangle$ it suffices to show that

$$\frac{\mathrm{d}}{\mathrm{d}c}F(c,\varepsilon) = \frac{1}{c^2}\left(c\frac{\mathrm{d}G}{\mathrm{d}c}(c,\varepsilon)-G(c,\varepsilon)\right) \leqslant 0 \tag{B.9}$$

for all $0<\varepsilon<1$, since then (B.7) is also monotonically decreasing in $c>0$. Now

$$\frac{\mathrm{d}}{\mathrm{d}c}G(c,\varepsilon) = -2\int_0^c r\mathrm{e}^{-2r}\left[\frac{r}{c^2}\frac{1}{1+r/c}+\frac{r(1-\varepsilon)}{c^2}\frac{1}{1-r/c(1-\varepsilon)}\right]\mathrm{d}r + 2\int_c^\infty r\mathrm{e}^{-2r}\left[\frac{1}{r}\frac{1}{1+c/r}+\frac{(1-\varepsilon)}{r}\frac{1}{1-(c/r)(1-\varepsilon)}\right]\mathrm{d}r. \tag{B.10}$$

The first integral on the r.h.s. of (B.10) is negative. By (B.8), (B.9) and (B.10) it therefore suffices to show that for any $r>0$

$$\frac{c}{r}\frac{1}{1+c/r}-\ln\left(1+\frac{c}{r}\right) \leqslant 0. \tag{B.11}$$

and

$$\frac{c(1-\varepsilon)}{r}\frac{1}{1-(c/r)(1-\varepsilon)}+\ln\left(1-\frac{c}{r}(1-\varepsilon)\right) \leqslant 0. \tag{B.12}$$

Now

$$\ln\left(1+\frac{c}{r}\right) = \frac{1}{r}\int_0^c \frac{1}{1+c'/r}\,\mathrm{d}c' \geqslant \frac{1}{r}\frac{1}{1+c/r}c$$

which is (B.11). (B.12) is proved in the same fashion. This concludes the proof of lemma B.

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