Suppression of Ionization in Superintense Fields without Dichotomy

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We examine the dynamics of atomic ionization in superintense fields directly in the Kramers-Henneberger frame. We show that atomic stabilization is intimately dependent not only on intensity and frequency, but also on pulse turn-on characteristics which determine the distribution of Kramers-Henneberger states in the superposition which is excited.

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A number of theoretical investigations have shown that the lifetime of an atom can be surprisingly long in currently available superintense fields [1] with intensities $I > I_{\rm at} = 3.51\times10^{16}$ W cm^{-2}; it can *increase* with increasing intensity [2]. One parameter characterizing the transition from multiphoton ionization (which increases with increasing intensity) to stabilization is the classical excursion amplitude α_0 of a free electron of mass m oscillating in an electromagnetic field of amplitude E_0 and frequency ω, $\alpha_0 = eE_0/m\omega^2$, and $\alpha_0 > 1$ (in atomic units) marks the onset of stabilization and the emergence of a (sometimes) dichotomous wave packet [2]. However, satisfying this criterion alone does not necessarily bring about stabilization. We shall show that stabilization is linked to the application of smoothly turned-on pulses of sufficiently high frequency.

A single electron moving in an atomic potential V and interacting with a laser electric field is described by the Hamiltonian

$$H = p^2/2m + V - e\mathbf{r}\cdot\mathbf{E}_0(t)\sin(\omega t)\,. \qquad (1)$$

Here, we examine the question of stabilization in terms of the Kramers-Henneberger (KH) frame [3], using the one-dimensional potential considered earlier [4], namely, $V(x) = -1/(1+x^2)^{1/2}$. The Kramers-Henneberger frame transforms the electron motion to a frame oscillating at the laser frequency, so that $V(x)\rightarrow V(x+\alpha(t))$, where $\alpha(t) = \alpha_0\sin(\omega t)$. We numerically solve the time-dependent Schrödinger equation directly using finite-difference techniques and employ appropriate projections for interpretation. We expand the potential in a harmonic series

$$V(x+\alpha(t)) = \sum_{n=-\infty}^{\infty} V_n(\alpha_0,x)e^{-in\omega t}\,, \qquad (2a)$$

where the average over an optical period $T = 2\pi/\omega$ is

$$V_n(\alpha_0,x) \equiv \frac{1}{T}\int_{-T/2}^{T/2} V(x+\alpha(t))e^{-in\omega t}dt\,. \qquad (2b)$$

Previous work has focused on the stabilization of single initial states, of either the field-free or the KH time-averaged potential. In frequencies large compared to the binding energy $W_0(\alpha_0)$ in the presence of the field (i.e., substantially less than the field-free energy) ionization is suppressed [2]. Suppression is linked to the relative unimportance of the time-dependent terms (V_n, $n\neq0$) of the KH potential. We discuss in this paper the effects of the following: What if the pulsed excitation produces a superposition of many KH eigenstates, and what if the frequency is not excessively large? Throughout, we adopt the viewpoint that we are discussing dynamics rather than statics, and expect the pulse shape and the nature of the field turn-on to be important [5]. We do not adopt a Floquet approach which only addresses the question of how an atom survives in a superintense field [6], but instead we also ask how the atom gets into such an environment. We find that the pulsed excitation controls *which* KH states are accessed, and can, for very fast pulse excitation rise times result not in suppression but exceedingly rapid ionization.

We adopt the same grid integration method as in previous papers [7] and directly integrate the time-dependent Schrödinger equation for the full KH Hamiltonian. In Fig. 1 the eigenfunctions of V_0 are plotted for $\alpha_0 = 18.49$, a high frequency of $\omega = 2$, and $E_0 = 73.96$. The dichotomous ground-state wave function with low probability density at the center of the potential [2] is shown in Fig. 1(a). However, we should note that the excited states shown in Figs. 1(b)–1(d), which we show play an important role in the ionization, are multilobed. Our full time-dependent wave function can be projected onto field-free or KH states depending on the relevant observables.

It is instructive for interpretative purposes to examine the behavior of an initial state $|\psi_1\rangle$ *suddenly* exposed to a constant superintense field. At later times t we can write $|\psi(t)\rangle$ in terms of the discrete $|l\rangle$ and continuous $|E\rangle$ *Kramers-Henneberger* states as

$$|\psi(t)\rangle = \sum_l C_l e^{-iE_l t}|l\rangle + \int dE\, C(E)e^{-iEt}|E\rangle\,. \qquad (3)$$

The amplitudes C_l and $C(E)$ are time independent if the higher Floquet terms V_n in Eq. (2b) are unimportant, so that the evolution is merely that of a superposition of stationary states. It is wrong to infer that no ionization can then take place, as some authors have, just on the basis of

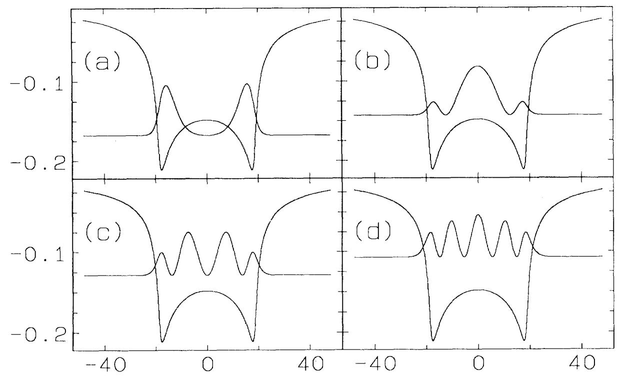

FIG. 1. Spatial dependence in atomic units of time-averaged dressed potential for $\omega=2$, $E_0=73.96$, $\alpha_0=18.49$, and the spatial KH wave-function probability density for (a) the ground state, and (b) the third, (c) the fourth, and (d) the fifth excited KH states. The probability density asymptotes are drawn at the appropriate height to indicate their energies in atomic units. For this value of α, the $n=2$ and $n=1$ probability densities are essentially identical.

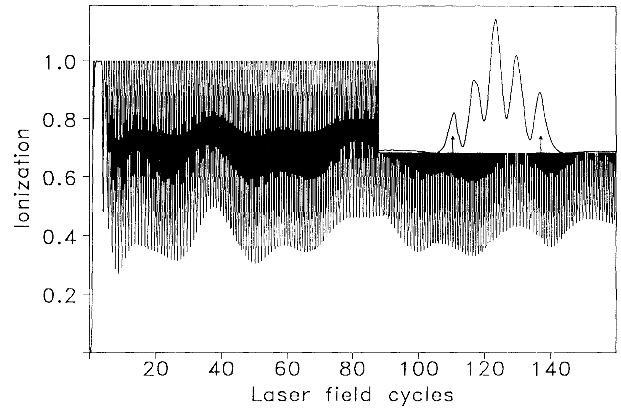

FIG. 2. Ionization probability of a field-free ground state excited by a smoothly turned-on field with a rise time of 5.25 cycles, with $\omega=2$, an asymptotic field amplitude of $E_0=73.96$, $\alpha_0=18.49$. The minima of the rapid oscillations demonstrate the residually stable packet (a superposition of KH eigenstates) persisting after approximately 40% ionization. Inset: A snapshot of the probability density as a function of x for this case after 76 cycles; the arrows indicate $x=\pm\alpha_0$.

the Hermiticity of the Hamiltonian. Ionization can still take place if sufficient population is transferred into the KH continuum states by the turn-on process, and depends on the energy mismatch between initial and continuum states compared with the reciprocal of the turn-on time. The probability amplitudes are written in terms of the overlap with the initial state, so that $C_l=\langle l|\psi_1\rangle$, and these overlaps are crucial in determining the distribution of KH states in the time-evolving superposition. We define the survival amplitude $A(t)=\langle\psi(0)|\psi(t)\rangle$ and from Eq. (3) find

$$A(t)=\sum_l |C_l|^2 e^{-iE_l t}+\int dE|C(E)|^2 e^{-iEt}. \qquad (4)$$

We note that if $C(E)$ is a square-integrable function, the last term tends to zero as $t\to\infty$ and represents ionization.

We have observed in our numerical computations that the KH ground state in an ultraintense field of *constant* amplitude at *high* frequencies ($\omega=2$, $E_0=73.96$, so that $\alpha_0=18.49$) is stable by calculating the ionization probability P_I, defined as 1 minus the total field-free bound-state probability as a function of time, starting with an initial, dichotomous KH ground-state wave function [2(a),7]. The wave packet oscillates rapidly, reconstructing its initial shape periodically as it traverses the nucleus with very little ionization loss (less than 1% during the twenty-cycle evolution).

We have examined the stability of the ground state for lower frequencies, $\omega=0.52$, used previously in studies of two-photon ionization of the ground state [2(b)]. The initial state *is* now coupled to KH continua by the higher-order time-dependent terms in the dressed potential [8] and does in fact ionize (by 30% in a twenty-cycle evolution), indicating partial ionization suppression rather than stabilization and confirming the importance of the high-frequency limit.

Next we need to address the question of how low-lying KH discrete states can be populated given pulsed excitation of a field-free ground state. In Fig. 2 we show the results of a numerical integration to give the ionization probability as defined above of a field-free initial ground state by a high-frequency field ($\omega=2$). This is turned on smoothly in 5.25 cycles of the laser field and then held constant so that questions of stability can be addressed by comparison with the results discussed above for a constant-amplitude field. The laser electric field is given by $E(t)=E_0\sin(\omega t)\sin^2(\pi t/\tau)$, $0<t<\tau</2$, $E(t)=E_0\sin(\omega t)$, $t>\tau/2$, where $\tau/2$ is the pulse rise time. We take a value of $E_0=73.96$ so that $\alpha_0=18.49$, and calculate the bound eigenfunctions (shown in Fig. 1) of the time-averaged potential appropriate for the constant part of the laser field. At this value of α_0, the dichotomy of the potential is marked and the binding energy of the ground state is reduced to -0.16 a.u. (from its field-free value of -0.67 a.u.), so the condition $\omega\gg W_0(\alpha_0)$ for the high-frequency limit of the potential is satisfied. We find that the pulse rise time is sufficiently short that a number of KH states are in fact populated; so it is *not* sufficient to analyze the stability of the KH ground state. Indeed, the population in each KH state $|l\rangle$ is determined in part by the overlap $|\langle l|\psi_1\rangle|^2$ and as we will show, this overlap is small for the KH ground state and leads to dramatic modifications to the wave-packet evolution beyond those expected from considerations of the ground state alone. A similar time evolution of the ionization probability can be calculated for $\omega=0.52$, and $E_0=5$ so that α_0 remains

18.49. Again an oscillatory wave packet is produced, but which ionizes *slowly*.

When the ionization probability as a function of time is examined (Fig. 2), a great deal of structure is observed. The continuum population oscillates at twice the laser frequency, between 1 and some approximately constant lower value, in this case 0.4. In a one-dimensional calculation, in the laboratory frame the electron wave packet moves through the nucleus twice per cycle. When the wave packet is centered on the nucleus, the overlap with the bound states is large and hence the continuum population is small. A coherent wave packet has been formed containing 60% of the total probability and if the laser were turned off, this wave packet would collapse to the bound states of the atom and not into the continuum. Therefore the lowest points on the continuum population graph actually represent the (true) ionized continuum population. From the lower extrema of this graph, it can be seen that the ionization is substantially suppressed and during the time of the pulse shown, there is a negligible amount of ionization in the constant electric field.

When the KH-frame wave function is examined at different times in the pulse, it can be seen from the inset in Fig. 2 that it is *not* simply dichotomous, but has roughly five lobes, the relative strengths of which vary as a function of time. The wave function is approximately symmetric and the time scale of the time variation is of the order of tens of cycles of the field.

The shape of the wave function and its time-varying nature contrast strongly with the dichotomous wave function predicted by Gavrila and others [2,7]. The explanation for the nondichotomous nature lies with the short turn-on of the field. The $l=1$ and the even-number eigenstates have very low probability amplitudes near the center of the potential where the initial wave function is concentrated, and so their overlap with the initial state is very small. The KH-frame ground state will therefore have a very low initial population and a dichotomous shape would *not* be expected. Instead, one would expect large initial populations in the states with large probability amplitudes at the center of the potential, primarily the $l=3$ and 5 states. This is confirmed numerically by direct projection on to KH eigenstates. Even when the atom sees a constant field amplitude, the bound-state populations continue to change. In spite of the $W_0(\alpha_0) \ll \omega$ condition being fulfilled, the time-dependent ($n \neq 0$) parts of the potential expansion are not negligible (especially for $\omega=0.52$) and cause transitions between bound states. These time-varying KH bound-state populations explain the time-varying wave-function shape; it is due to the trapped population moving between the bound eigenstates. Population moves into the other states that are not initially populated by the turn-on of the field. However, the longer time constant for the dynamics of the wave function for the $\omega=2$ case compared to the $\omega=0.52$ case implies again that the V_n's are more significant in the $\omega=0.52$ case.

The turn-on of the field results in a spread of population across those KH-frame bound states with a large amplitude at the center of the potential. As the pulse progresses, the KH-frame bound-state population becomes distributed among all the bound states because of the time-varying parts of the Floquet expansion of the KH-frame potential which cause multiphoton transitions to occur and results in a time-varying electron wave function with lobes that correspond to those of the predominant excited states. The outermost nodes of this wave function are approximately at $\pm\alpha_0$, as expected, because these are the positions of the potential walls, but the wave function is far from the simple dichotomous shape that would result if only the ground-state KH-frame eigenstate had been populated and no transitions between states had occurred.

We have also studied a much more rapid 0.25-cycle turn-on of the field as an extreme case of nonadiabatic excitation. Such a rapid turn-on populates directly the KH continuum and we find that this leads to essentially complete ionization in a few laser cycles. Hence we have shown that while the KH frame is useful for direct wave function evolution, working in the KH frame does not remove all the time-dependent dynamics of the system and although ionization suppression does most definitely occur, it is not due to the formation of a pure dichotomous ground-state wave function, but instead involves a dynamic wave packet of many bound eigenstates in the KH frame. We conclude that the field rise time can dominate the ionization: If it is too slow the atom is always ionized by normal multiphoton processes, and if it is too fast we see direct population transfer to KH continua. Stability is really a delicate balance between turn-on, intensity, and frequency.

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Note added.—After the submission of this work for publication, we learned of the work of Kulander, Schafer, and Krause [9] on stabilization of atomic hydrogen in three dimensions. They also stress the importance of the turn-on in populating superpositions of KH states.

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