

Laser-induced resonance states as dynamic suppressors of ionization in high-frequency short pulses

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(Received 14 June 1999; published 8 December 1999)

An adiabatic-Floquet formalism is used to study the suppression of ionization in short laser pulses. In the high-frequency limit the adiabatic equations involve only the pulse envelope where transitions are purely ramp effects. For a short-ranged potential having a single-bound state we show that ionization suppression is caused by the appearance of a laser-induced resonance state, which is coupled by the pulse ramp to the ground state and acts to trap ionizing flux.

PACS number(s): 32.80.Rm

I. INTRODUCTION

The interaction of atomic electrons with intense laser fields is an active field of research for over 30 years and has been recently reviewed by several authors [1–3]. Perhaps the most exciting aspect is that every scale of laser frequency and intensity leads to phenomena caused by “different physics.” At weak fields, the perturbation theory of bound-free transitions is sufficient to explain most phenomena [4]. At stronger fields, it is necessary to correct for Stark shifts and level broadening. At even stronger fields, the Coulomb potential is severely distorted to a metastable well form; the electron either tunnels through or crosses over a potential barrier towards ionization [5,6].

A different theory is needed for understanding the interaction of a bound electron with high-frequency continuous-wave (CW) laser fields. Gavrilin and co-workers developed a high-frequency Floquet theory within the Kramers-Henneberger gauge [7,8] where now the atomic potential distortion does not lead to a metastable well. Instead, at high frequencies a stable dichotomous potential emerges [7,9–12], which is sometimes also reflected in a dichotomy of the electron density [12,13]. As the electric field amplitude E_0 of the laser increases, the distance α_0 between the dichotomous wells grows and the potential well-depth decreases. In its simplest (zero-order) form, the theory predicts stabilization merely because the dichotomous well supports bound states and light-induced ionization-resistant states. A first-order correction for lower frequencies accounts for some ionization from the dressed states, which now acquire a complex quasienergy [14]. Based on this picture electron lifetime is predicted [15] to increase with laser intensity, contradicting common spectroscopic intuition. This type of stabilization was coined “adiabatic stabilization” and corresponds to a CW high-frequency limit.

Intense-field short pulses also exhibit a suppression of ionization termed “dynamic stabilization” [16]. Here the

pulses have a short rise time [17,18]. Adiabatic stabilization is predicted by theory and dynamic stabilization by numerical simulations. Due to experimental difficulties, only a limited number of experiments and subsequent theoretical treatments [19–22] provide evidence for existence of stabilization.

The relation between the dynamic and adiabatic stabilization is an important issue. The practical possibility of measuring the light-induced atomic states, for example, depends on this issue [23]. Adiabatic stabilization can be considered a limiting case of dynamic stabilization [24]. This fact can be exploited to calculate Floquet-type quantities from finite time-dependent wave-packet computations [25]. The shape of the pulse envelope is known to have an effect on the dynamic stabilization. For example, it is established that a sudden turn-on of the intense field will generate complete ionization [26]. However, the interplay between frequency-induced ionization and envelope-induced ionization is still not fully understood.

In this paper we study dynamic stabilization. The goal is to qualitatively understand ionization by intense high-frequency short pulses. Using the $t-t'$ formalism of Refs. [27–30] an adiabatic-Floquet theory is obtained forming a single framework for both short and long pulses. We use this approach to studying the high-frequency limit of ionization for an electron in a short-ranged potential. The high-frequency limit is where the theory assumes its simplest form, analogous to the CW case in Ref. [8]. A high-frequency limit is found to exist also for short pulses; however, here the ionization is not completely suppressed: it is a function of the shape of the pulse ramp. This is a “pure ramp ionization” limit, and ionization suppression is caused by the formation of a laser-induced resonance state that traps ionizing flux.

II. THEORY

The electron dynamics in an intense laser field with vector potential $A(t)$ is described by the Schrödinger equation in the Kramers-Henneberger [9] (KH) gauge:

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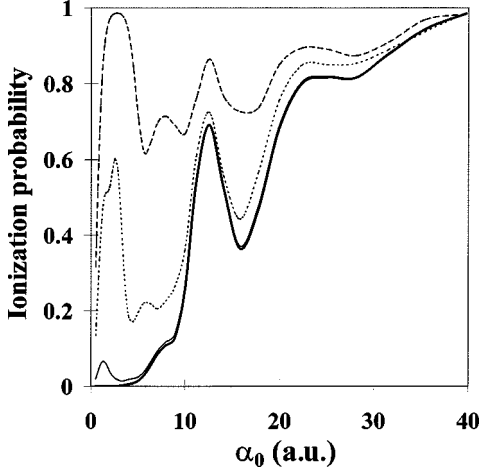


FIG. 1. Total ionization probability vs α_0 , for an Eq. (3) pulse with $T_{\text{on}}=94.25$, $T_{\text{flat}}=251.3$ a.u. and $\omega=0.25$ a.u. (dashed line), 0.5 (dotted), 1.0 (solid), and 2.0 (thick line).

$$i \frac{\partial}{\partial t} \psi(x, t) = \left(\frac{P^2}{2m_e} + V(x + \alpha(t)) \right) \psi(x, t) \quad (1)$$

where $P = -i\hbar\nabla$ and the instantaneous displacement $\alpha(t)$ is determined by $\dot{\alpha} = c^{-1}\mathbf{A}(t)$.

Consider a short electromagnetic pulse with displacement of the form $\alpha(t) = \alpha_0 f(t) \sin \omega t$. Here, α_0 is the maximal displacement and the function $f(t)$ depicts the pulse envelope of finite duration starting from $t=0$ ending at $t=T_f$. For an electron initially in the ground state $\psi_0(x)$, the high-frequency limit is studied by numerically integrating Eq. (1) with a one-dimensional short-range potential [31]:

$$V(x) = -B \exp(-x^2/2\sigma^2). \quad (2)$$

The potential is constructed to support a single bound state (the values of $B=0.187$ a.u. and $\sigma=1.8$ a.u. are chosen to yield a bound state energy of approximately -0.1 a.u.). The total ionization probability, as a function of α_0 and ω was calculated for a sine-square envelope [24], characterized by a ramp time T_{on} and flat time T_{flat} :

$$f(t) = \begin{cases} \sin^2(\pi t/2T_{\text{on}}), & t \leq T_{\text{on}} \\ 1, & T_{\text{on}} < t \leq T_r, \\ \cos^2[\pi(t-T_r)/2T_{\text{on}}], & T_r < t \leq T_f, \end{cases} \quad (3)$$

where $T_r = T_{\text{on}} + T_{\text{flat}}$. The wave-packet calculations were done within the KH gauge, using the Chebyshev expansion method of Kosloff and Tal-Ezer [32] with complex scaling of the Hamiltonian derived from Hoang, Kouri, and Hoffmann [33], using the Neuhauser-Baer negative imaginary potential [34] at the grid asymptotes. A grid spacing of $\Delta x = 2$ a.u. and time step of $\Delta t = 2\pi\omega^{-1}/4$ gave a solid four-digit convergence. The shifts of the potential in the KH frame are performed using fast-Fourier transform based on the identity $V(\hat{x} + \alpha) = e^{-i\hat{P}\alpha/\hbar} V(\hat{x}) e^{+i\hat{P}\alpha/\hbar}$.

The calculation results are shown in Fig. 1 where the end of pulse ionization is plotted as a function of the maximal displacement α_0 for several frequencies. All other envelope

parameters are kept constant. As the frequency is raised the ionization profile converges to a limit—the high-frequency limit. Unlike the CW case where the limit is zero, here there is a structure. The ionization initially rises monotonically with α_0 until it reaches a local maximum of 0.7 probability at $\alpha_0=12$ a.u. As α_0 is further increased beyond this value, the ionization probability rapidly decreases, reaching a local minimum at $\alpha_0=16$ a.u.. Beyond this the ionization increases once more as α_0 is increased.

III. NONADIABATIC PICTURE OF HIGH-FREQUENCY STABILIZATION

In order to understand these results let us follow Ref. [30] and generalize the $(t-t')$ formalism, treating the fast time t as a dynamic variable and defining a slow-time parameter τ and the τ -dependent Hamiltonian:

$$H(\tau) = \frac{P^2}{2m_e} + V(x + \alpha_0 f(\tau) \sin(\omega t)) - i\hbar \frac{\partial}{\partial t}. \quad (4)$$

With this Hamiltonian, just like in the usual $(t-t')$ formalism, the Schrödinger equation

$$i\hbar \partial \psi / \partial \tau = H(\tau) \psi(x, t; \tau) \quad (5)$$

is solved with the initial condition $\psi(x, t; 0) = \psi_0(x)$. The full solution of Eq. (1) is then obtained as $\psi(\tau) = \psi(t = \tau, \tau)$.

The extended Hamiltonian $H(\tau)$ is periodic in t , allowing the following Fourier expansion:

$$V(x + \alpha_0 f(\tau) \sin(\omega t)) = \sum_n e^{in\omega t} V_n(x, \tau), \quad (6)$$

where the expansion coefficients are

$$V_n(x, \tau) = \frac{1}{2\pi} \int_0^{2\pi} e^{-in\phi} V(x + \alpha_0 f(\tau) \sin(\phi)) d\phi. \quad (7)$$

An adiabatic approach is now used to treat the slow time τ . Consider the instantaneous eigenfunctions and eigenvalues:

$$H(\tau) \psi_j(x, t; \tau) = \varepsilon_j(\tau) \psi_j(x, t; \tau). \quad (8)$$

Because of periodicity in t the eigenfunctions are written as

$$\psi_j(x, t; \tau) = \sum_n e^{-in\omega t} \varphi_n^{(j)}(x; \tau), \quad (9)$$

where the components $\varphi_n^{(j)}(\tau)$ are the instantaneous Floquet states, satisfying

$$\sum_m \left\{ \left(\frac{P^2}{2m_e} - n\hbar\omega \right) \delta_{nm} + V_{n-m}(x; \tau) \right\} \varphi_m^{(j)}(\tau) = \varepsilon_j(\tau) \varphi_n^{(j)}(\tau), \quad (10)$$

where $\varepsilon_j(\tau)$ are the instantaneous quasienergies. The time-dependent wave packet is expanded in the instantaneous Floquet basis:

$$\psi(x,t;\tau) = \sum_j c_j(\tau) \exp\left(-i/\hbar \int_0^t \varepsilon_j(\tau') d\tau'\right) \psi_j(x,t;\tau), \quad (11)$$

Plugging this form into Eq. (5) leads to the equation

$$\dot{c}_j = \sum_k \left\langle \psi_j \frac{\partial}{\partial \tau} \psi_k \right\rangle_{x,t} \exp\left(i/\hbar \int_0^t (\varepsilon_k - \varepsilon_j) d\tau'\right) c_k(\tau). \quad (12)$$

Using the Hellmann-Feynman theorem and Eq. (9), one obtains for the nonadiabatic transition coefficients ($k \neq j$):

$$(\varepsilon_k - \varepsilon_j) \left\langle \psi_j \frac{\partial \psi_k}{\partial \tau} \right\rangle_{x,t} = \langle \varphi_m^{(j)} | \dot{V}_{n-m} | \varphi_n^{(k)} \rangle, \quad (13)$$

where

$$\dot{V}_l(x,\tau) = \frac{\alpha_0 \dot{f}(\tau)}{2i} (\nabla V_{j+1} - \nabla V_{j-1}). \quad (14)$$

Note that nonadiabatic transitions are of course possible *only when the envelope itself is changing*, as clearly seen in Eq. (14). The strength of transitions also depends on the gradient of the Fourier components. It is seen clearly that for a continuous wave pulse, where the pulse envelope is constant, the transition rate is zero, as is the case for the theory of Gavrilu. Note, however, that this observation does not mean there is no ionization, because in general the eigenvalues ε_j may be complex with negative imaginary parts. In the adiabatic picture it is meaningful to differentiate between ‘‘adiabatic ionization,’’ which is caused by a complex energy and nonadiabatic ionization caused by nonadiabatic transitions.

The theory above makes no specific assumptions and can be considered for our purposes exact. When the laser frequency ω is very high we can make simplifying approximations. Following Gavrilu [8] we neglect the rapidly oscillating parts of the eigenfunctions ψ_j . We therefore set

$$\psi_j = \varphi_0^{(j)}, \quad \varphi_m^{(j)} = 0 \quad (m \neq 0). \quad (15)$$

In this case the eigenvalues $\varepsilon_j(\tau)$ corresponding to bound states are all real. The equation for the coefficients is

$$\begin{aligned} \dot{c}_j &= \sum_k T_{jk} c_k \\ &= \sum_{k \neq j} \frac{\langle \varphi_0^{(j)} | \dot{V}_0 | \varphi_0^{(k)} \rangle}{\varepsilon_j - \varepsilon_k} \\ &\quad \times \exp\left(-i/\hbar \int_0^t (\varepsilon_k - \varepsilon_j) d\tau\right) c_k(\tau) \\ &\quad - \langle \varphi_0^{(j)} | \dot{\varphi}_0^{(j)} \rangle c_j(\tau), \end{aligned} \quad (16)$$

where

$$\dot{V}_0(x,\tau) = \dot{f}(\tau) \alpha_0 \nabla \left\{ \frac{1}{2\pi} \int_0^{2\pi} V(x + \alpha_0 f(\tau) \sin \phi) \sin \phi d\phi \right\}. \quad (17)$$

Equation (16) is an adiabatic equation for the slow Schrödinger equation:

$$i\hbar \frac{\partial}{\partial \tau} \Psi(\tau) = \left\{ \frac{P^2}{2m_e} + V_0(x,\tau) \right\} \Psi(\tau). \quad (18)$$

This equation is independent of the frequency and yields the ‘‘high-frequency limit’’ shown as the bold line in Fig. 1. Because the potential $V_0(x,\tau)$ is of the same parity as the atomic potential $V(x)$, only even-even and odd-odd transitions are possible in the high-frequency limit.

Equation (16) may be solved directly for the adiabatic population amplitudes. Here we ignored the odd adiabatic decoupled states. During the pulse rise and pulse decay we calculated the M lowest even eigenstates (the ‘‘adiabatic states’’) of the instantaneous Hamiltonian of Eq. (18). This need not be done by diagonalization (although for the present system size it was the most efficient way), since the lowest adiabatic states can also be calculated using filter-diagonalization [35] methods or a Chebyshev expansion of projection operator [36]. When the pulse flat time is very long, the adiabatic approach is more efficient than solving Eq. (18). If only the M lowest adiabatic states are important (where M is much smaller than the number of grid points), the transition matrix in Eq. (16) is a small $M \times M$ anti-Hermitian matrix and the evolution of the coefficients for the time step can be efficiently performed by diagonalizing it. We found that 12 states were important for quantitative agreement with the full wave packet results. However, as we will shortly show, only two states are needed for understanding the general features of the stabilization.

The numerical stability of the evolution within the adiabatic scheme is sensitive to the way one chooses the phases of the adiabatic states. The exact adiabatic equations can employ any phase choice but any numerical implementation should make sure that the phases change continuously. One way to do this is to choose phase of the n th eigenstates $\varphi_n(\tau + \Delta\tau)$ so that $\langle \varphi_n(\tau) | \varphi_n(\tau + \Delta\tau) \rangle = \rho$ is a real positive number. There is a possibility of accumulating geometric Berry phases [37] at each state. If this happens the eigenstate at the end of the pulse has a Berry phase $\Omega_B = \pi$ relative to the state at time $t=0$, in other words: $\langle \varphi_n(0) | \varphi_n(T_{fin}) \rangle = -1$. In the present context, since we have no degeneracies and we are only interested in adiabatic state *population*, the geometric phase is inconsequential. Also, we did not encounter such a phenomenon here. In a more general case of working within an adiabatic representation, a removal of such the discontinuity between the initial and final adiabatic states is possible by judicious choice of the phase assignment. For example, when a Berry phase does accumulate in state n it can be eliminated by including a counter-rotating phase in $\varphi(\tau + \Delta\tau)$, choosing $\langle \varphi_n(\tau) | \varphi_n(\tau + \Delta\tau) \rangle = \rho e^{i(\pi\Delta\tau/T_{fin})}$.

The population of the five lowest eigenstates for various times during the rise and decay of a $\alpha_0 = 16$ a.u. pulse is shown in Fig. 2. The prominent feature, becoming apparent

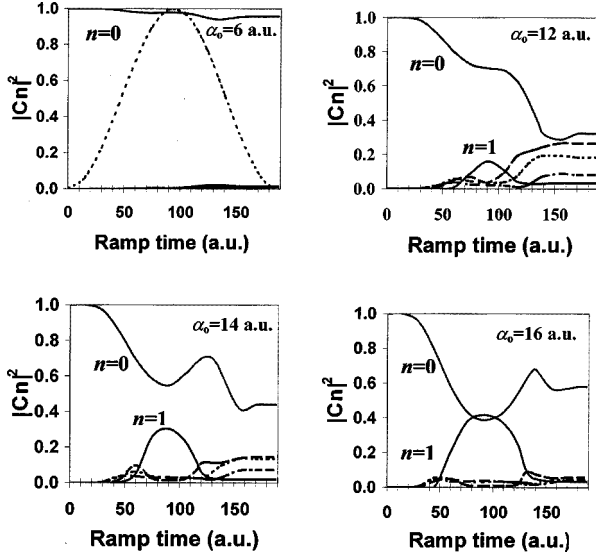


FIG. 2. Population of the lowest five adiabatic states as a function of pulse rise and decay time for four pulses with the shown maximal displacement α_0 . Pulse ramp forms are shown as a dotted line in the $\alpha_0 = 6$ a.u. figure. The $n=0$ and $n=1$ lines are captioned while the $n=2$ is a dashed line, $n=3$ is dotted, and $n=4$ is dot-dashed. In all cases the $n=0$ state starts with $|C_0|^2 = 1$ at $t = 0$ a.u. and loses population to excited states, which are all positive energy states.

at $\alpha_0 \approx 12$ a.u. is the strong population transfer between the $n=0$ and the $n=1$ states. This effect becomes dominant for yet larger α_0 's. Thus, under the influence of a strong laser field, the $n=1$ state acts as a trap against ionization, explaining the high-frequency stabilization starting at $\alpha_0 \approx 12$ a.u. and reaching a maximum suppression at $\alpha_0 \approx 16$ a.u. (see Fig. 1). Beyond $\alpha_0 \approx 16$ a.u. ionization suppression degrades as the $n=1$ states population is high and some transfer to continuum states occur.

The reason for the trapping effect is the creation of a laser-induced resonance state as shown in Fig. 3. It is possible to estimate the magnitude of displacement α_{res} at which the resonance appears. Define the α -independent quantity $A = \int_{-\infty}^{\infty} V_0(r, \alpha) dr$ and assume the dressed potential may be approximated as a square well of length $L = 2\alpha$. The resonance appears when the well depth A/L is equal to the energy of the second state in a square well $E_2 = 2\hbar^2 \pi^2 / m_e L^2$; thus $\alpha_{\text{res}} \approx \hbar^2 \pi^2 / m_e A$. In the potential function used in this paper, the equation correctly estimates the appearance of the resonance at $\alpha_0 = 12$ a.u.

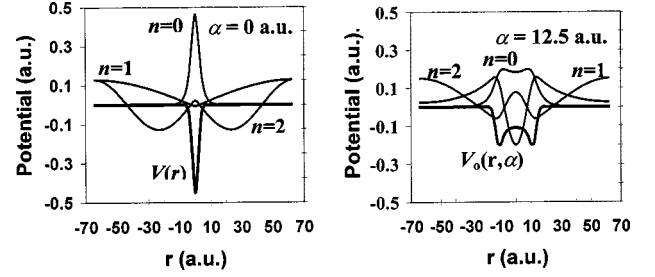


FIG. 3. The shape of the dressed potential at $\alpha_0 = 0$ a.u. and $\alpha_0 = 12.5$ a.u.. Superimposed, the amplitude of the first three even adiabatic states. It is seen that while the $n=0$ state is bound and $n=2$ is a continuum state in both cases, the $n=1$ state changes character from a continuum state to a localized resonance.

IV. CONCLUSION

The adiabatic framework has allowed a straightforward explanation of dynamic stabilization in short-ranged potentials caused by the appearance of a laser-induced resonance state that traps ionized population. A fundamental difference between adiabatic and dynamic stabilization is that the former is caused by the suppression of nonadiabatic transitions while the latter exhibits strong transitions and the suppression of ionization is caused by trapping into a laser-induced resonance state. Pulses must also be turned off for a complete understanding of dynamical stabilization. The pulse can also have a ‘‘flat’’ part where no nonadiabatic transitions occur. Still, the length of the flat part has an influence on the total ionization, because it determines the quantum phases at which components remix as the pulse is turned off. Dynamic stabilization in our case is a manifestation of light-induced atomic-resonance states. Two related questions deserve detailed treatment in future work. First, what is the role of laser induced resonance states when longer ranged potentials are present. Second, what is the analog of Fig. 1 when, instead of fixing the frequency and varying α_0 (i.e., laser intensity I), it is I that is kept fixed and α_0 that is controlled by varying ω^2 .

ACKNOWLEDGMENTS

We thank Ronnie Kosloff, Charlie Cerjan, Ken Kulander, Nimrod Moiseyev, and Avinoam Ben Shaul for illuminating comments. R.B. gratefully acknowledges support from the Fritz Haber Research Center at the Hebrew University of Jerusalem. A.E.O. and D.B. were supported by NSF Grant No. PHY-97-22136 and the Livermore National Laboratory under DOE Contract No. W-7405-Eng-48.

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