

Excess-photon Ionization Spectra and Atomic Structure in Intense Laser Fields

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Abstract. Floquet states represent intrinsic modes of ionization of an atom in a monochromatic field of constant intensity. To describe atomic wave packets evolving in realistic laser pulses, linear superpositions of Floquet states are required ("multistate Floquet theory"). This gives the possibility of following the evolution of wave packets in terms of the Floquet states that are populated during the pulse. We study here the way in which the Floquet states present in the representation of the wave packet manifest themselves in the excess-photon ionization spectra (EPI/ATI). For the purpose of illustration we choose a 1D atomic model with a soft-core Coulomb potential. We calculate the totality of the Floquet states, at all intensities needed, and generate the corresponding "Floquet map". We then calculate the EPI spectra for wave packets evolving from the ground state under different types of pulses. By analyzing the location of the lines in the spectrum, and their shapes, we show that they can be associated, in a clear cut and predictable way, to Floquet states responsible for the emission. The understanding of the underlying physics can lead to tailoring laser pulses, such as to obtain EPI signals in a controlled way. Whereas our analysis is applied to theoretical spectra, it would apply, just as well, to experimental ones.

INTRODUCTION

Weak-field atomic spectra reflect the unperturbed structure of the atom. The location, multiplet structure, and intensity distribution of the experimentally determined lines give information on the atomic energy levels and eigenfunctions. These can be then compared to theoretical results derived from the Schrödinger eigenvalue equation.

In intense laser fields the situation is complicated by the fact that atomic structure gets distorted by the field and cannot be defined independently from ionization. At fixed ω and I the theoretical building blocks for the description of ionizing atomic

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structure in the field are the Floquet states, which are analogs of the unperturbed stationary states of the Schrödinger eigenvalue equation. The unperturbed energy of stationary states corresponds to the (complex) quasienergy of Floquet states, and the wave function of the stationary states, to the (infinite) set of components of Floquet states. Floquet states represent intrinsic ionization modes of the atom, and for CW laser fields, the "excess-photon ionization" (EPI/ATI) spectrum is related to one of these modes. When dealing with laser pulses, however, one may expect that the complete set of Floquet states, for *all* intensities involved, is needed. This set represents under the circumstances the "structure" of the atom. An analysis of EPI spectra in terms of this generalized set is conceivable, although it is not obvious a priori that one can thereby identify the states involved in the emission process. The result will obviously depend on the laser pulse features (shape, turn-on/off, etc).

A typical example of a pulsed-laser EPI spectrum is shown in Fig. 1. Is it possible to read the Floquet ionization modes involved, from such a spectrum? Can one identify the effects of the pulse shape? Can one modify the pulse shape so as to influence the EPI spectrum in a controlled way?

We want to show in the following that theoretical EPI spectra can be, indeed, interpreted in terms of atomic structure in the (varying amplitude) field, represented by the instantaneous Floquet states generated by the pulse. This opens up control possibilities. To this end we shall adopt a three-step procedure: we

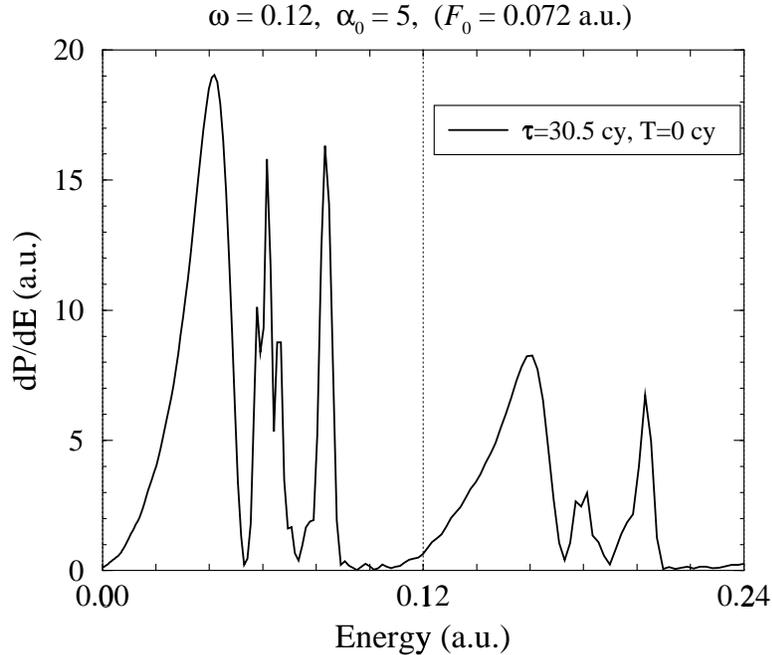


FIGURE 1. EPI/ATI spectrum computed for a laser pulse of \sin^2 shape and 61 cycle duration. The frequency is 0.12 a.u., and peak field strength is 0.072 a.u. We show the spectrum from zero energy to $2\hbar\omega$.

first obtain the Floquet states needed as a basis of interpretation, at all intensities involved, by means of a time-independent calculation; we then derive the EPI spectrum from a time-dependent wave packet description of the ionization with various pulses; finally, we carry out the Floquet analysis of the spectrum. Our approach has a conceptual, purely theoretical character. It indicates, however, that a similar analysis should be possible for experimental EPI spectra as well.

For illustration we shall use a 1D atomic model, with a "soft-core" Coulomb potential,

$$V(x) = \frac{-1}{\sqrt{a^2 \exp[-(x/a)^2] + x^2}} . \quad (1)$$

This is finite at the origin, and its Coulomb tail supports an infinite set of Rydberg states. By choosing the parameter $a = 1.6$, the ground-state energy is -0.50 a.u.

FLOQUET STATES

At fixed laser intensity and frequency, the time-dependent Schrödinger equation admits quasistationary solutions of the Floquet type, defined in terms of boundary conditions. For ionization, the appropriate boundary conditions are of the Gamow-Siegert form, and lead to an eigenvalue problem, having as complex eigenvalue the Floquet "quasienergy" parameter. The problem has an infinite set of eigensolutions, most of them being unacceptable physically. The "physical" Floquet solutions are specified by specially chosen boundary conditions: in the energetically "open channels" one requires outgoing asymptotic waves, with non-zero electron currents at infinity; in energetically "closed channels" one requires exponentially damped asymptotic behavior, with no currents at infinity. This is connected with the intuitive interpretation of the physical Floquet states as possible ionization modes of the atom. However, from the point of view of quantum mechanics, they have built in conceptual difficulties from their very definition (such as their non-square integrable character, which is a consequence of their asymptotic behavior), which preclude their acceptability as true physical states (for a general discussion of Floquet states, see [1]).

It was discovered that physical Floquet states may become unphysical when the intensity is increased continuously, and vice versa [2]. The first case is described broadly as "channel closure", whereas the latter case, as the materialization of a "light-induced state" (*LIS*). The discovery of *LIS* was first made for 1D model atoms [3], [4], and then for 3D physical systems, such as atomic H [5], and H⁻ [6]. It was, however, the analysis in [2] which has clarified the circumstances of materialization and disappearance of *LIS* at energy thresholds $n\omega$ ($n = integer$).

Although *LIS* have been known for more than a decade now, their physical significance has remained uncertain because of their peculiar features. The known difficulties of interpretation of Floquet states are compounded by new ones: the

fact that *LIS* cannot be followed in intensity from a field-free limit, that they often exhibit strange transient behavior [2] (e.g., *LIS* may materialize as some intensity, only to become unphysical again at a higher intensity), etc. Such behavior could hardly be regarded as physically acceptable, on the basis of Floquet theory solely.

Nevertheless, in a recent study based on wave packet dynamics we have concluded that *LIS* have *physical reality*, because they leave a clear signature on observable phenomena, such as EPI spectra [7]. Our study was followed by an instructive discussion [8], [9].

The first step in carrying out the approach mentioned in the Introduction is

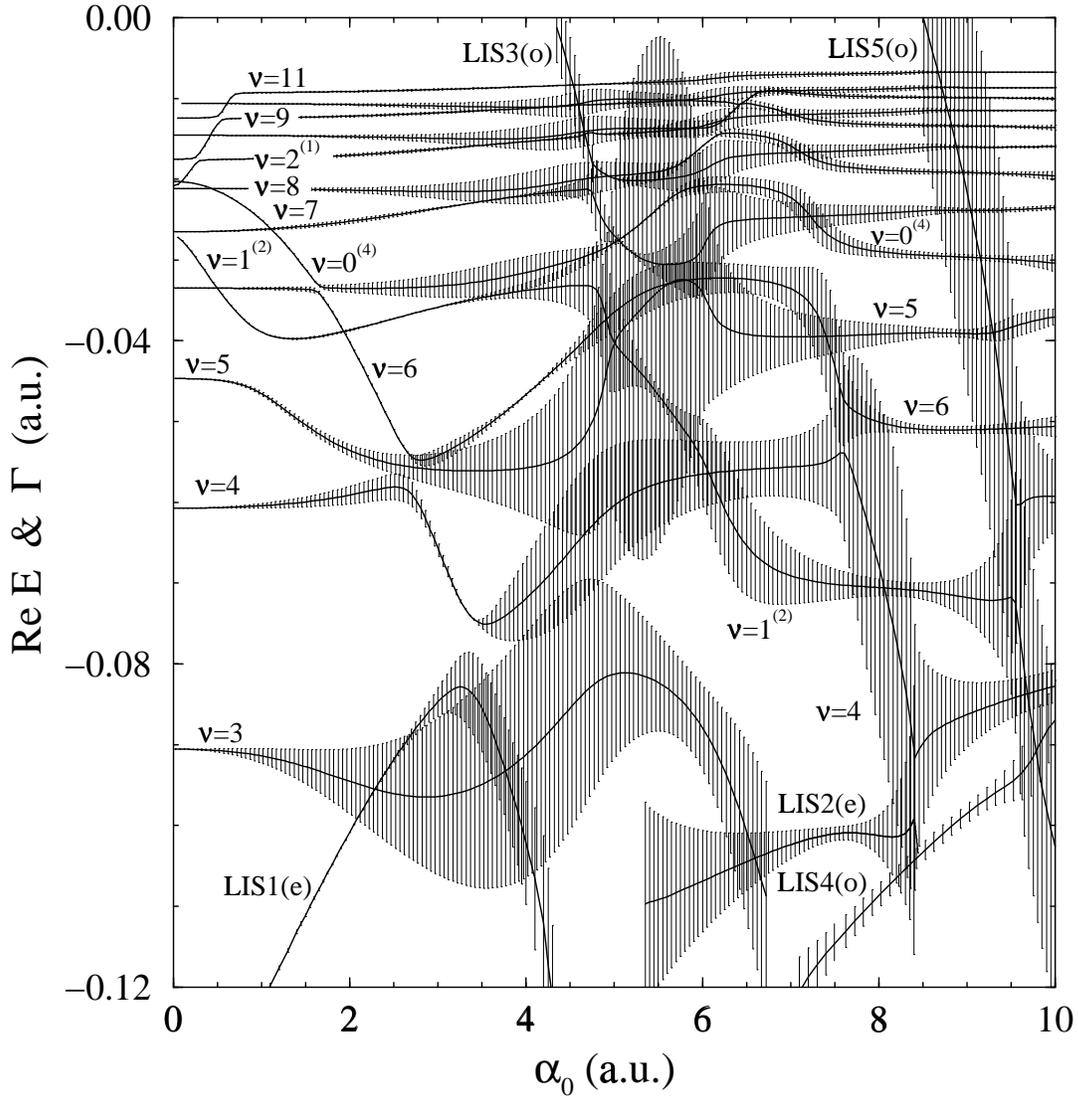


FIGURE 2. Floquet quasienergies $\Re(E)$, $\Gamma = 2\Im(E)$ at $\omega = 0.12$ a.u. $\Re(E)$ is represented by the solid lines. Γ is represented by the length of the vertical segment associated with $\Re(E)$.

the calculation of a complete set of (relevant) Floquet states of the system, for all intensities needed, at the fixed frequency considered. Thus, in Fig.2 we present computed results for the Floquet quasienergies of our 1D model atom, defined by Eq.(1) at $\omega = 0.12$ a.u. We plot the complex quasienergies E as a function of the parameter $\alpha_0 = I^{1/2}/\omega^2$, where α_0 is the quiver amplitude of a free electron in a monochromatic laser field. Five photons are needed to ionize the atom at small intensity. The real part of the quasienergies $\Re(E)$ is plotted modulo ω in the first energy band $(0, -\omega)$. We have this freedom due to the periodicity of these solutions. Quasienergies of regular states (i.e., which exist at $I = 0$), shifted into this band in Fig. 2, are labeled with a superscript denoting the integer multiple of ω required to bring this solution into this first energy band. For example, the ground state $\nu = 0$ requires $n = 4$ photons to be lifted into this band, and is thus labeled $\nu = 0^{(4)}$. For *LIS* we have given their parity, even or odd. $\Re(E)$ provides, however, only part of the information contained in the quasienergy, it has to be complemented by $\Gamma = 2\Im(E)$. We have introduced in [7] a way of visualizing Γ in the same figure as $\Re(E)$, by representing it as the length of the vertical segment associated with $\Re(E)$. The "Floquet map" Fig. 2 is rich in features, which will only be mentioned here: presence of "persistent" states (remaining in the energy band), channel closures ($\nu = 1, 3$), five *LIS* (*LIS1-LIS5*, some of them transient), and many avoided crossings. The figure also displays some of the bizarre features of *LIS*.

WAVE-PACKET DYNAMICS

Wave packets Ψ representing physical states can be analyzed in terms of Floquet states. A first step has been to try to represent Ψ by one Floquet state only, $\psi^{(\nu)}(x, t; \omega, E_0)$: this is single-state Floquet theory, the customary way to practice Floquet theory. Such a treatment has significant limitations: an adiabaticity assumption needs to be made concerning the turn-on/off of the field; the ionization rate Γ needs to be sufficiently small; one cannot describe multiphoton resonances; etc.(for a discussion see [1]). Some of these conditions are met in actual experiments, some not.

The next step is to extend the analysis by trying to represent physical states as superpositions of Floquet states (see [10]):

$$\Psi \propto \mathbf{S}_\nu C_\nu(E_0) \psi_\nu(x, t; E_0, \omega) . \quad (2)$$

The $C_\nu(E_0)$ are complex-valued coefficients, and \mathbf{S}_ν should be interpreted as a summation over discrete Floquet states, and an integration over continuum states, carried out along a contour in the complex energy plane. In a constant amplitude field E_0 , C_ν are constant, in a variable amplitude field they are time-dependent due to $E_0(t)$. This "*multi-state Floquet theory*" has its unsolved mathematical problems, one of them being the lack of a completeness proof for Floquet states defined *off* the real-energy axis. Some of them have been enumerated and discussed

in [10]. We have taken the pragmatic approach of using Eq.(2) for our analysis as long as it works; it works so far.

One question which arises is: should the expansion Eq. (2) include *LIS* ? Our answer is yes, and a proof of this statement was given in [7]. More discussion was given in the contribution to this conference by I. Simbotin, J.C. Wells, and M. Gavrilin: “*LIS* behavior in Intense Laser Fields”.

Let us analyze in terms of Eq.(2) the evolution of an atom exposed to a laser pulse of the form $E(t) = E_0(t) \sin \omega t$, with a smooth envelope $E_0(t)$. We will discuss \sin^2 type turn-on/turn-off of $E_0(t)$ over a duration τ_0 , with possibly a “flat-top” segment with duration T in between. So, the duration of our pulses is $\tau = 2\tau_0 + T$. We start with the atom in a field-free state, for example in the ground state. For sufficiently slow turn-on, one should achieve an adiabatic evolution, and only the initial $\nu = 0$ Floquet state will be populated during this stage (i.e., $|C_0| \simeq 1$, and all other $|C_{\nu \neq 0}| \simeq 0$). Note however that, in practice, some measure of “shake-up”, i.e., excitation to several other Floquet states may occur from the very beginning, for a more rapid turn-on of the pulse. In general, adiabatic evolution will continue until a multiphoton resonance with some state $\nu = 1$ is encountered, i.e., until $\Re(E_0) \simeq \Re(E_1)$, modulo ω . We recall that a resonance is associated with an avoided crossing (AvCr) of the quasienergy trajectories in the complex plane. There are two basic alternatives *at resonance*: (1) *diabatic transition* to the encountered state ($\nu = 0 \rightarrow \nu = 1$), or (2) *adiabatic passage* through the AvCr, the system remaining in the same state $\nu = 0$. The branching ratio between these alternatives depends on the laser pulse. The evolution of the wave packet appears on the Floquet map as a progression along a path made of portions of Floquet curves, interrupted at avoided crossings by possible jumps from one curve to another. We call one such possible path a “*diabatic path*” (DP). Several DP may start from a given initial state, branching off successively with certain probabilities, at various avoided crossings. A DP may extend over several energy bands, as is clearly illustrated in Fig. 2. In the general case the system evolves concomitantly along several DP’s (a “coherent superposition of DP’s”).

INTERPRETATION OF EPI SPECTRA

We now present EPI spectra for one particular DP and discuss how the structure of the atom in the laser field manifests itself in the photoelectron spectra emitted. These EPI spectra present many features, and only some of them will be discussed here. A more detailed account will be given elsewhere.

The DP we consider (to be designated as DP0) starts from the field-free ground state $\nu = 0$, descends at increasing intensity along the curve for $\nu = 0^{(4)}$, passes to curve $\nu = 6$ at the AvCr occurring at $\alpha_0 \approx 1.6$, passes to curve $\nu = 4$ at the AvCr at $\alpha_0 \approx 2.5$, passes to the descending branch of *LIS1* at the AvCr at $\alpha_0 \approx 3.3$, continues (*modulo* ω) on *LIS3* which descends from the top of the energy band, passes to $\nu = 7$ at the AvCr $\alpha_0 \approx 4.7$, passes to $\nu = 1^{(2)}$ at the broad AvCr

$\alpha_0 \approx 5$, etc; DP0 has many secondary branchings. In Fig.3 we show spectra for four pulses of increasing peak intensity α_0 , and fixed shape with $\tau_0 = 15.5$ and $T = 0$. Only photoelectron kinetic energies smaller than 2ω are represented. The vertical markings give the photoelectron energies for the Floquet states indicated at the *peak-intensity* α_0 . Note that the lines of these spectra are not located precisely on the markings. This is because in general photoelectron emission occurs all *along* the DP, not just at its end point of peak intensity. In panel (a) of Fig.3, at $\alpha_0 = 1$, one observes signals from the first two states, $\nu = 0$ and $\nu = 6$, the latter being excited by shake-up from the very beginning. Panel (b), shows that as the peak intensity is increased to $\alpha_0 = 2.4$, the wave packet clearly has accessed state $\nu = 4$, on the other side of the AvCr at $\alpha_0 \simeq 2.5$, but there is still a strong remnant signal

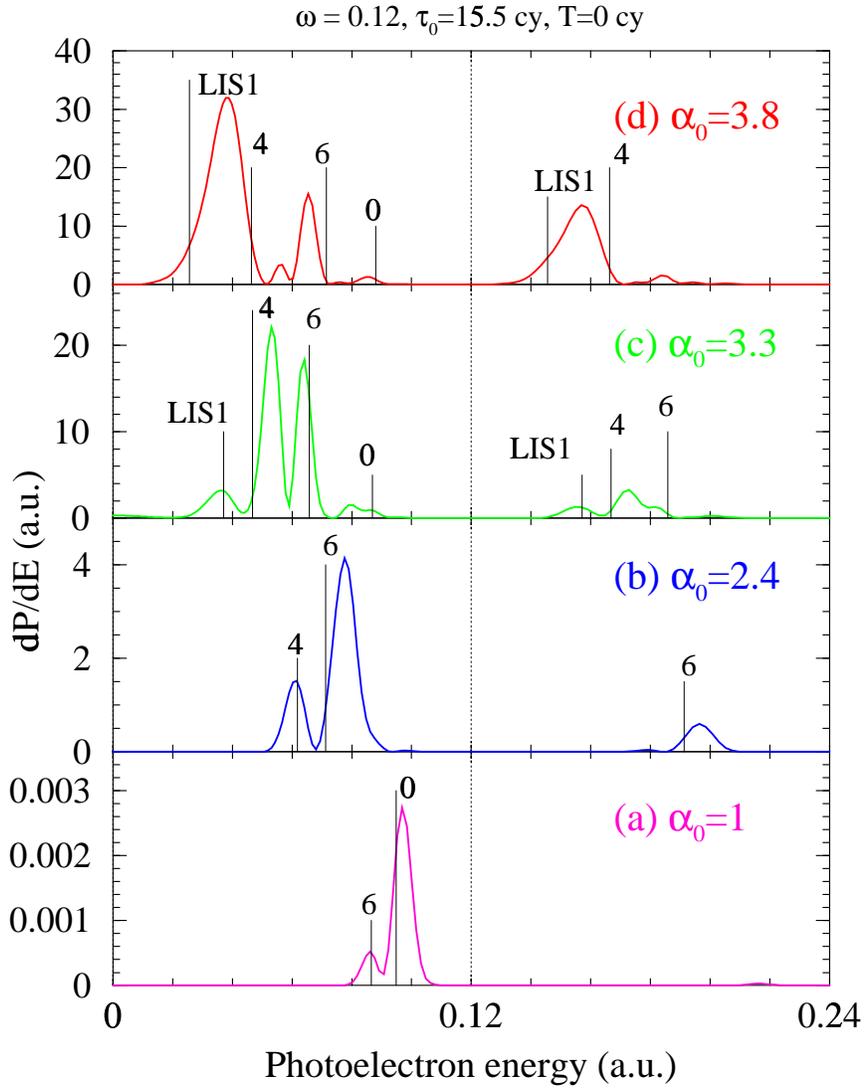


FIGURE 3. EPI/ATI spectra at $\omega = 0.12 \text{ a.u.}$, initial state $\nu = 0$, $\tau = 31$ cycles.

from $\nu = 6$. At $\alpha_0 = 3.3$, panel (c) shows clear signals from all of the first four states along DP0, $\nu = 0, 6, 4$, and the first light-induced state, *LIS1*. Some of the signal from state $\nu = 0$ is coming from initial shake-up to $\nu = 6$ followed by a diabatic transition to $\nu = 0$ at $\alpha_0 \simeq 1.6$. At $\alpha_0 = 3.8$, shown in panel (d), one observes a strong signal overlapping the markings for $\nu = 4$ and *LIS1*, but no individual signals from these two states.

Figure 4 shows a comparison of spectra from pulses of the same peak intensity, but of differing shapes. One of the results in the top panel is from a rapid turn-on of the pulse with $\tau_0 = 4.5$ cycles, while the one in the lower panel is turned on with $\tau_0 = 60$ cycles. Both of these pulses have a $T = 20$ flat top. Note in the upper panel that, when the pulse is turned on quickly to peak value, the diabatic

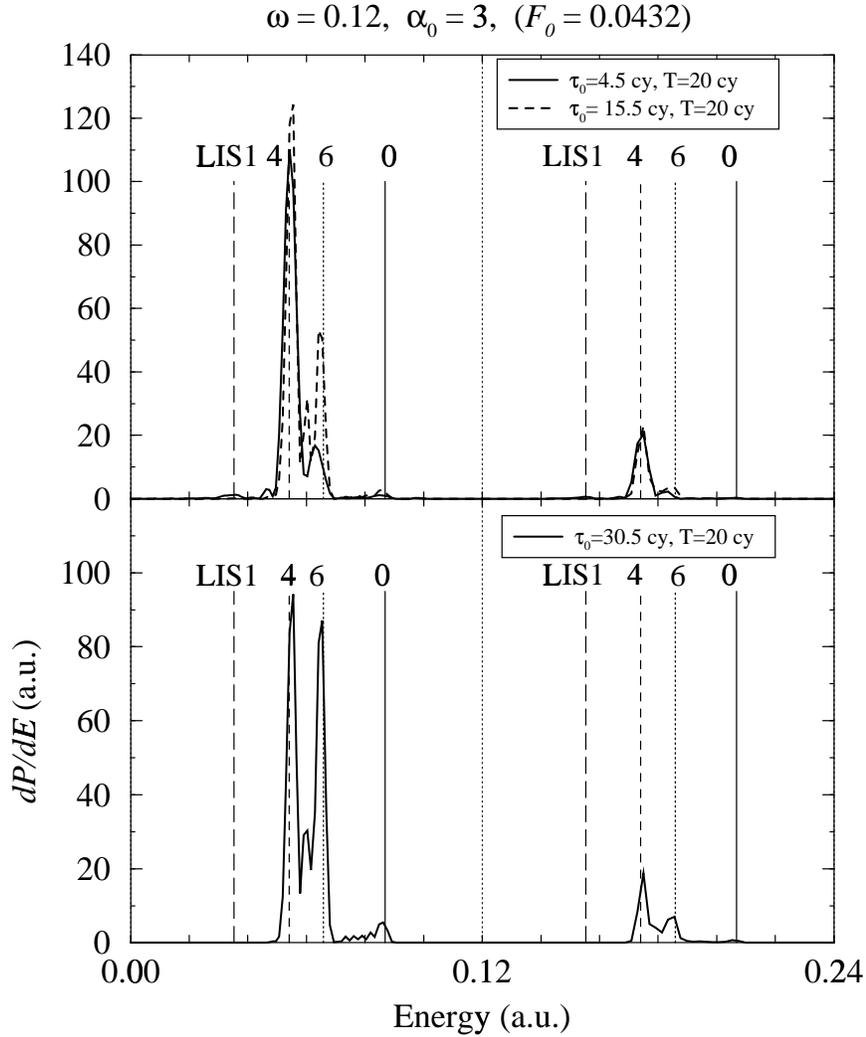


FIGURE 4. EPI/ATI spectra at $\omega = 0.12$ a.u., initial state $\nu = 0$, comparing the effect of varying the pulse length.

transition $\nu = 6 \rightarrow \nu = 4$ is favored at the AvCr $\alpha_0 \approx 2.5$, whereas, as shown in the lower panel, at slow rise of the pulse, there is a large probability of the system staying in $\nu = 4$. The case $\tau_0 = 15.5$ in the top panel is intermediate. This is a simple example that the evolution of the atom along the diabatic path may effectively be controlled by the pulse (in contradistinction to the statements made in [8]).

In Fig. 5 we return to the value of $\alpha_0 = 3.8$ a.u., shown previously in panel (d) of Fig. 3, where one could not distinguish between the contributions of $\nu = 4$ and *LIS1*. Along with the pulse shape considered in Fig.3, given as reference, we consider a faster turn-on of $\tau_0 = 4.5$ cycles, and a slower one, $\tau_0 = 30.5$ cycles. The shorter turn-on demonstrates the channeling of population at the AvCr at

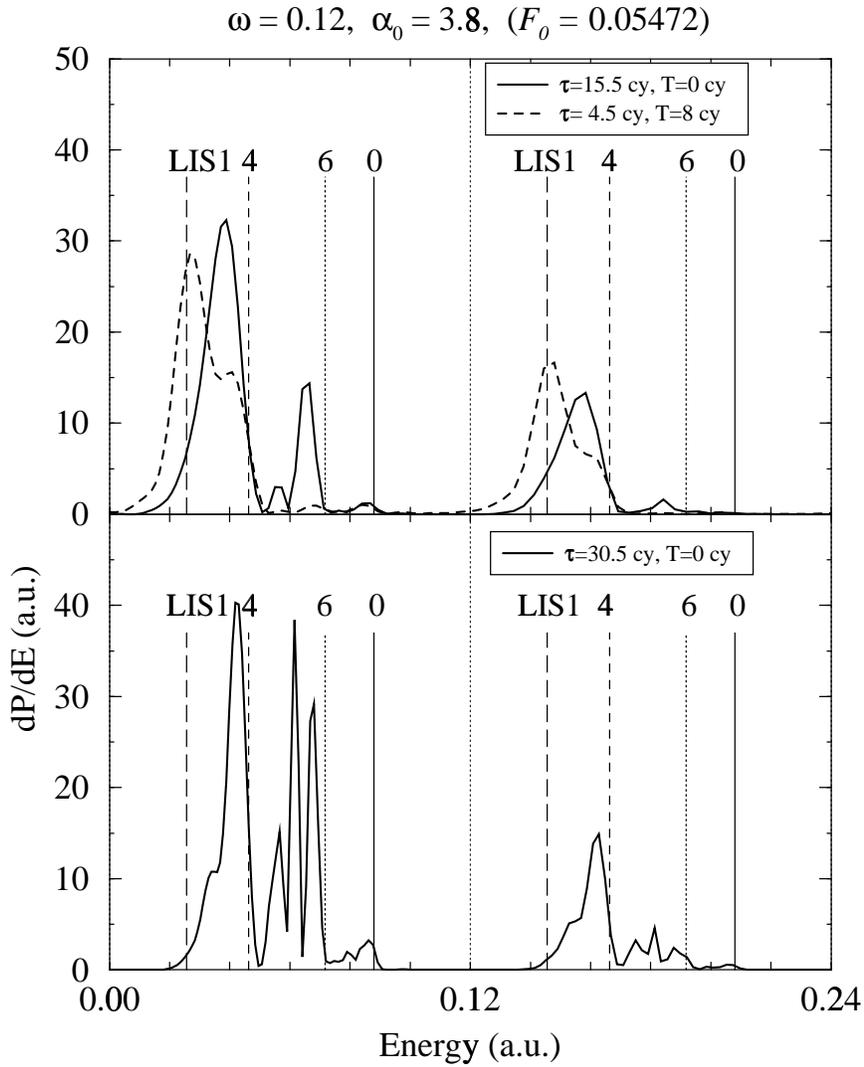


FIGURE 5. EPI/ATI spectra at $\omega = 0.12$ a.u., initial state $\nu = 0$, comparing the effect of varying the pulse length.

$\alpha_0 \approx 3.3$ into the diabatic transition, whereas the slower pulse results in a preference for passing the AvCr adiabatically. The two states $\nu = 4$ and $LIS1$ are clearly identifiable, whereas in the intermediate case $\tau_0 = 15.5$ they were obscured. The interference pattern appearing in the lower panel, at energies preceding the marking for $\nu = 6$, is due to the emission of electron de Broglie waves at the same kinetic energy, during the turn-on and turn-off of the pulse, a phenomenon long-known in the literature [4], [11].

In the top panel of Fig. 6, we return to the spectrum at $\alpha_0 = 5$ shown originally in Fig. 1, but now included are markings for the location of the relevant emitting Floquet states (along with markings for some non-emitting states, for the record). Even at these intensities, the signals in the photoelectron spectrum are

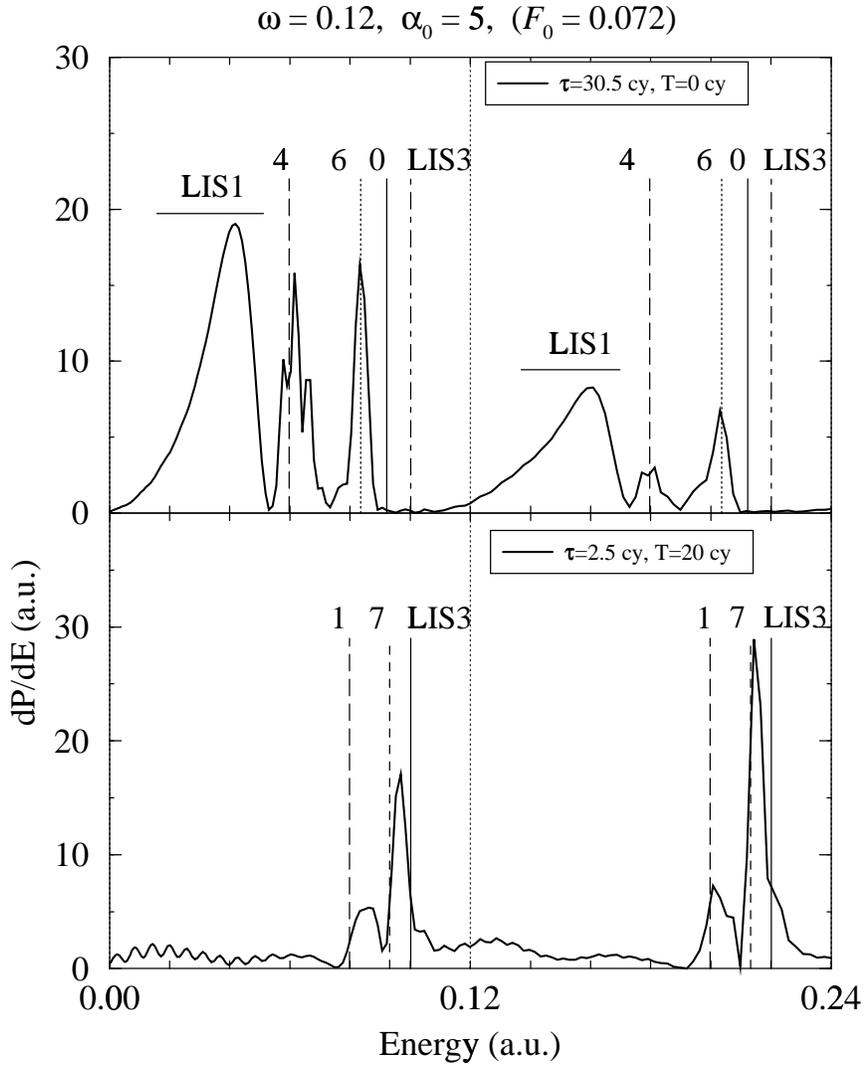


FIGURE 6. (*top*) EPI/ATI spectra as in Fig. 1, and (*bottom*) for a much shorter pulse.

clear. Floquet states $\nu = 4$ and $\nu = 6$ have strong signals showing a predominance for branching into these states along DP0 (several mechanisms can be invoked). A dominant feature is the broad, asymmetric line at the low-energy side of the first kinetic energy band ω , denoted by *LIS1*. Note that *LIS1* has already disappeared at $\alpha_0 = 5$, but the feature is nevertheless associated to *LIS1*, because the emission has taken place prior to the channel closure, due to the large rate Γ along the descending branch of *LIS1*. The asymmetric shape is due to variations in the state population and ionization rate along this branch prior to channel closing. Note that there is no signal from *LIS3*, or other states at the top of the energy band. This is because the pulse duration is long enough so that there are no more neutrals left by the time the peak intensity is attained. The bottom panel of Fig. 6 shows the result of turning the pulse on extremely rapidly ($\tau_0 = 2.5 c.$) at the same intensity. In this case, it is precisely the states reached at the peak of the pulse, such as $\nu = 7$ and $\nu = 1^{(2)}$, that are given the possibility to emit, especially at the long $T = 20 c.$ chosen. However, when ramping the field on this fast, the atom receives a strong kick projecting part of its population directly into the continuum ("shake-off"); part of the broad background, extending to zero kinetic energy, is due to this excitation mechanism.

CONCLUSION

We have shown that the representation of wave packets as superpositions of Floquet states ("multistate Floquet theory") is a very useful tool in the interpretation of EPI spectra. In this picture the atom evolves probabilistically along one or several DP's. Shake-up, shake-off, and diabatic transitions play are essential ingredients in this picture. The lines in the EPI spectra can be attributed quite clearly to emitting Floquet states, even at high intensities. The pulse shape plays an important role in steering the atom in a controlled way along the various DP's. Much remains to be done in this respect. Finally, it again follows from our discussion that *LIS* have physical reality, and participate on equal footing with the other Floquet states in atomic dynamics.

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