SPECTROSCOPY OF ATOMS IN INTENSE LASER FIELDS

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INTRODUCTION

The goal of atomic spectroscopy is to determine atomic structure from experimental spectra. In a weak-field environment, one determines the stationary, unperturbed structure of the atom. Theoretically, the Schrödinger equation provides the organizing principle, and the task is to solve this equation to obtain its energy eigenvalues and eigenvectors. In the intense-laser-field environment, the atomic structure is dependent on the laser field. For monochromatic fields, the atomic structure is parameterized by the frequency, ω, and intensity, I, of the field. For this case, the quasi-stationary properties of the atom in the field are given by Floquet theory. The Floquet states represent or describe the natural decay modes of the atom in the field, and are the generalization of the stationary energy eigenfunctions, whereas, the energy eigenvalues are generalized to the complex Floquet quasi-energies. These Floquet states are the building blocks of the atomic structure in the intense laser field, and understanding the EPI/ATI photoionization spectra means deciphering the presence or signature of the Floquet states in the spectra.

For example, consider a typical (theoretical) photoelectron spectrum (1-D model atom) presented in Fig. 1. Is it possible to extract information on the structure of the atom in the laser field? Certainly, the character of the laser pulse must be included in the analysis of the spectrum. This leads to a dual approach where we provide a wave-packet dynamics description of the ionization phenomena combined with interpretation from Floquet analysis. To define the model atom we wish to discuss for illustration, we choose a 1D potential for the soft-core type,

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

choosing the parameter $a = 1.6$ so that the ground-state energy is -0.50 a.u.

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FLOQUET STATES

Assume the laser frequency and intensity to be fixed, this results in periodicity for the field. As a result, the Floquet states are the quasi-stationary solution of the periodic time-dependent Schrödinger equation specified in terms of boundary conditions. For ionization, Gamow-Siegert boundary conditions are appropriate and lead to an eigenvalue problem for the complex quasi-energy. This eigenvalue problem has an infinite set of solutions, with most solutions being unphysical. The physical solutions are specified by adequate boundary conditions. That is, the open channels are represented by outgoing waves with non-zero currents at infinity. Closed channels are represented by exponentially damped functions, with no currents at infinity. This results in an intuitive interpretation of physical decay modes. These physical solutions are not without mathematical difficulties, such as their asymptotic divergence, non-square integrable character, etc.

However, we cannot simply discard the non-physical solutions because the physical and non-physical solutions may interchange at $n\omega$ energy thresholds as the intensity varies [1]. The significance of the interchange of the physical and non-physical states is as follows. When a physical solutions goes over to a non-physical solution as function of increasing intensity, we say that *channel closure* occurs, where the lowest open ionization channel changes from N photons to $N+1$ due to large Stark shifts across an energy threshold. When a non-physical solution

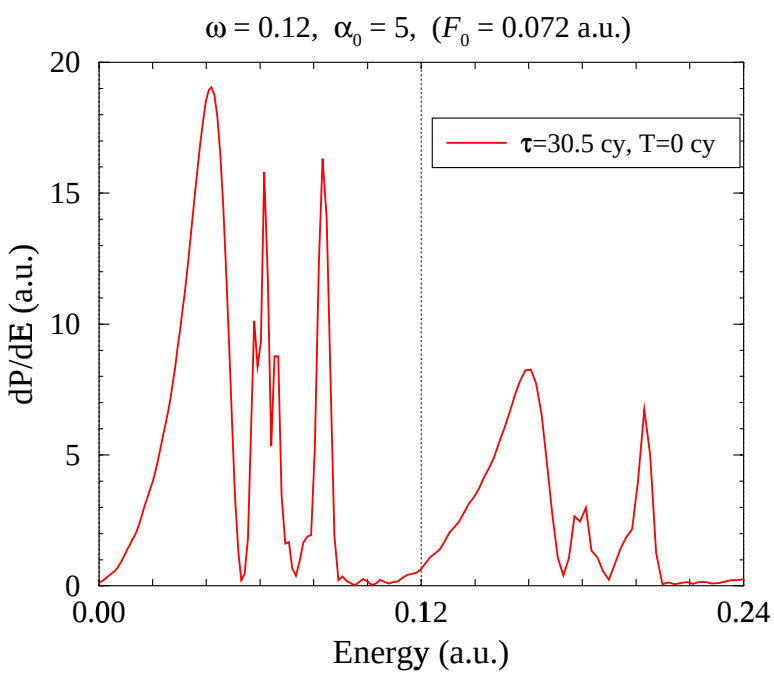

FIGURE 1. EPI/ATI spectrum computed for a laser pulse with a turn-on time of 30.5 cycles. The frequency is $0.12 a.u.$, and peak field strength is 0.072 a.u. We show the spectrum from zero energy to two times the laser frequency.

becomes a physical solution, the atom acquires a new state (a channel opening). This new state is called a *light-induced state* (LIS).

The LIS sometimes exhibits transient existence as a function of intensity. An unphysical solution may become physical as intensity increases only to become unphysical again at a higher intensity. Can these LIS be accepted as "physical states". The history of LIS extends over some 10 years with descriptions of LIS in model potentials appearing since 1988 [2], and in physical systems in the early '90's [3]. At the time, the physical reality of the LIS was an open question.

In Fig. 2 we present computed results for the Floquet quasienergies of our 1D, one-electron model atom defined by Eq. 1. We plot the complex quasienergies E

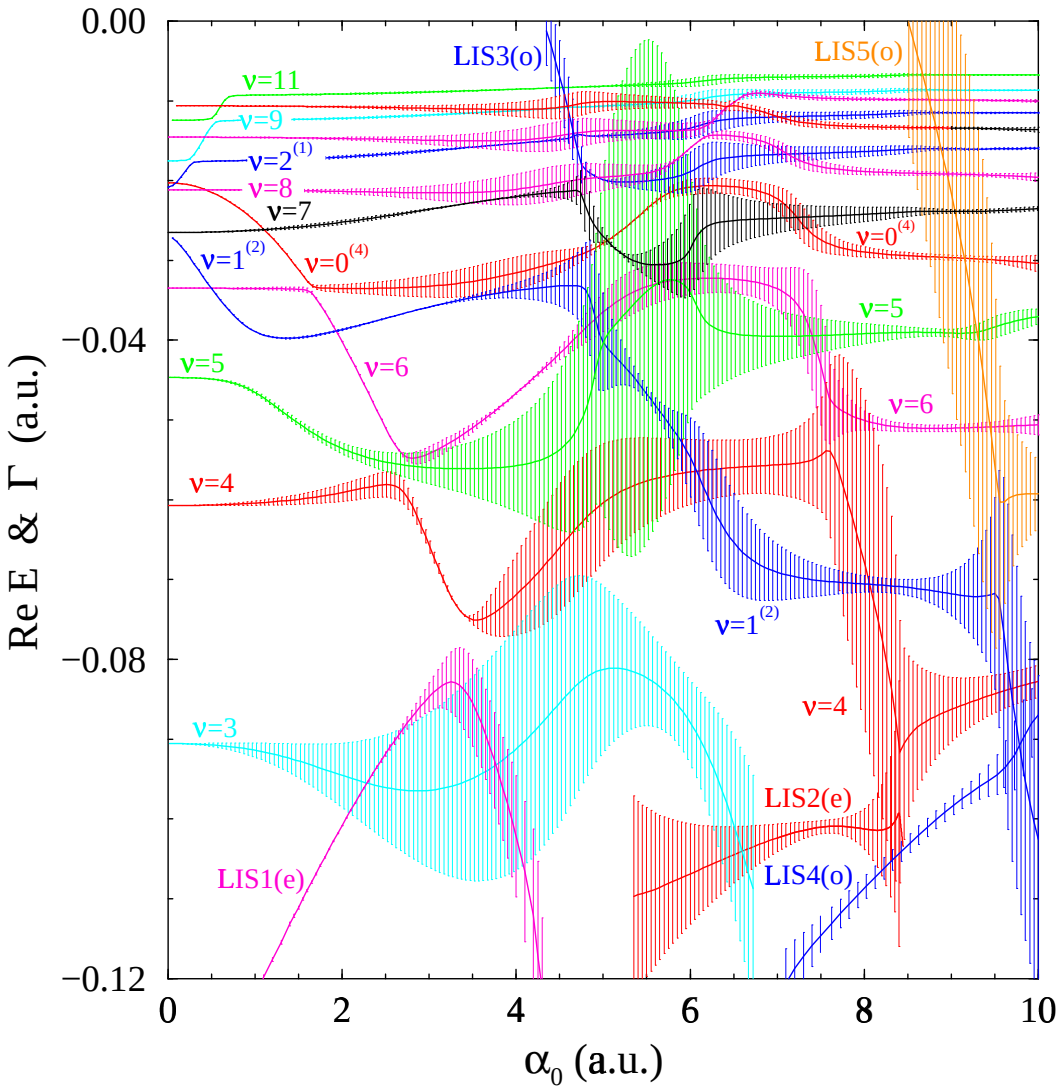

FIGURE 2. Floquet quasienergies $\Re(E)$, $\Gamma = 2\Im(E)$ at $\omega = 0.12 a.u.$ Γ is represented by the length of the vertical segment associated with $\Re(E)$.

as a function of the parameter $\alpha_0 = \sqrt{I}/\omega^2$, where α_0 is the quiver amplitude of a free electron in a monochromatic laser field. The quasienergies are plotted modulo ω in the first energy band $(0, -\omega)$. We have this freedom due to the periodicity of these solutions. Quasi-energies 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 lift it into this band, and is thus labeled $\nu = 0^{(4)}$. Since $\Re(E)$ provides only partial information on the quasi-energy, we have introduced Floquet quasi-energy "maps" visualizing both $\Re(E)$ and $\Gamma = 2\Im(E)$, where Γ is depicted in Fig. 2 as the length of the vertical segment associated with $\Re(E)$. The many features of this Floquet map include "persistent" states (remaining in the band), channel closures ($\nu = 1, 3$), five LIS (LIS1-LIS5, some transient), and many avoided crossings.

WAVE-PACKET DYNAMICS ANALYSIS

Let us consider wave-packet dynamics (WPD) analysis in terms of Floquet theory. A first step would be to consider only a single Floquet state, $\psi^{(\nu)}(x, t; \omega, I)$: Single-State Floquet Theory (SSFT). Our Floquet state ψ has an index ν which we use to refer to the field free state *when possible*. The Floquet state is parametrized by the frequency ω and intensity I of the laser field, and depends on space and time (x, t). For a laser pulse, one considers the intensity to have a time-dependent envelope. Such a treatment has significant and sever limitations: the adiabaticity assumptions, small ionization rates, inability to describe multiphoton resonances, etc.

A next step in improving this WPD analysis is to use a linear superposition of Floquet solutions for expanding wavepackets evolving in a laser pulse for all intensities and frequencies, i.e., Multi-State Floquet Theory (MSFT),

$$\Psi \propto \sum_{\nu} C_\nu(t) \psi_\nu(x, t; \omega, I) \ . \tag{2}$$

The $C_\nu(t)$ are the complex-valued expansion coefficients and the $\sum_\nu$ should be interpreted as a summation over discrete states and an integration over the continuum. This approach is also not without problems, not the least of which is the lack of a completeness proof for Floquet States defined off the real-energy axis. What should be the meaning of the association between the wavepacket Ψ and the Floquet-state expansion? Given a wavepacket, how would one compute the expansion coefficients? There are many open mathematical problems here which we do not have time or space to discuss [4]. Here we will take the approach of using Eq. 2) for our analysis as long as it works. It works so far.

On the issue of the LIS, should these Floquet solutions be included in Eq. (2)? Our answer is yes, and a description of our point of view is provided in Ref. [5]. An interesting and instructive discussion followed in Ref. [6] and Ref. [7]. For more discussion, see the contribution to this conference by Simbotin "LIS and Atomic Dynamics in Intense Laser Fields".

ATOMIC EVOLUTION ALONG DIABATIC PATHS

Consider laser pulses with smooth envelopes. We will discuss $\sin^2$ type turn-on/turn-off 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 allow the atom to begin in a field-free atomic state, for example the ground state. For sufficiently slow turn on, one will achieve an adiabatic evolution and only the $\nu = 0$ Floquet state will be populated during this stage, (i.e., $|C_0| \sim 1$, and $|C_{\nu\neq 0}| \sim 0$). Note however, in practice, some measure of "shakeup", i.e., the projection of amplitude into many states, is possible due to rapic turn-on of the pulse. In general, adiabatic evolution continues until a multiphoton resonance is encountered: $\Re(E_0) \sim \Re(E_\mu)$ modulo ω. Note here that resonances are associated with avoided crossing of the quasienergy trajectories in the complex plane. At resonance, one has two basic alternatives: (1) diabatic transition to another state,

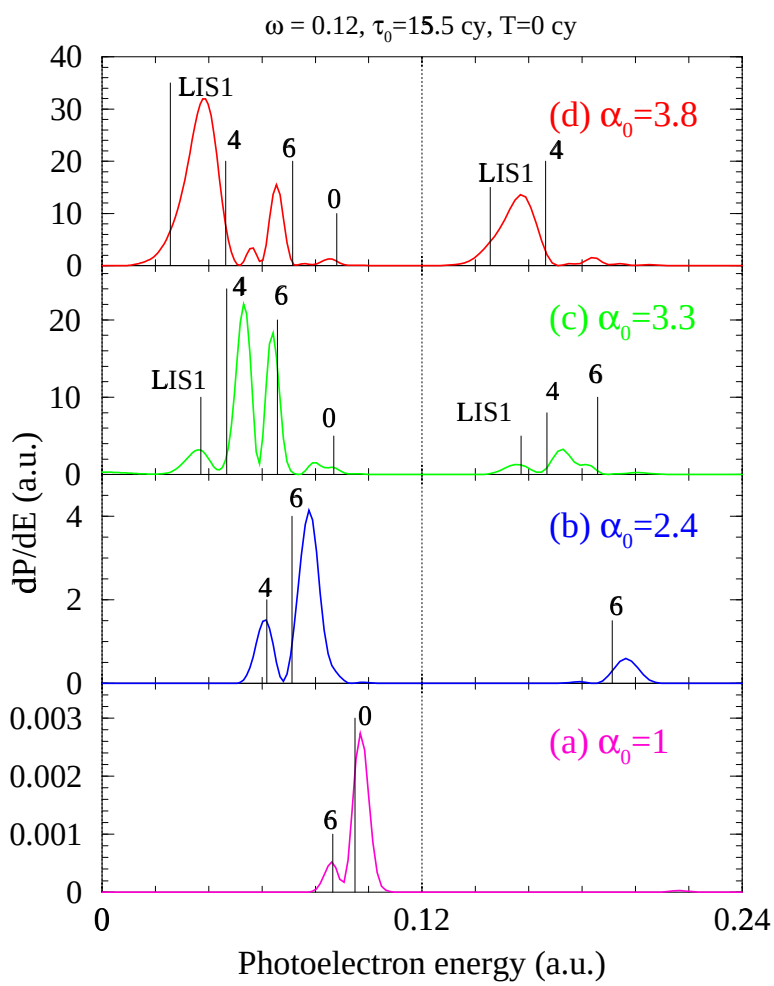

FIGURE 3. EPI/ATI spectra at $\omega = 0.12$a.u., initial state $\nu = 0$, $\tau = 31$ cycles.

or (2) adiabatic passage through the avoided crossing remaining on the same state. The branching ration between those alternatives depends on the laser pulse. There exists sequences of adiabatic evolutions along Floquet states interrupted by diabatic transitions, as can be seen in Fig. 2. We call these sequences "diabatic paths" (DP). Several DP may begin from a single initial state and DP may extend over several energy bands.

INTERPRETATION OF EPI/ATI SPECTRA

We present EPI/ATI spectra for one particular DP starting from the ground state $\nu = 0$, and discuss how the structure of the atom in the laser field manifests itself in photoelectron spectra emitted. In Fig. 3 we show spectra four pulses of increasing intensity. The frequency is $0.12 a.u.$ so five photons are required to ionize the field free ground state. This series of spectra show the evolution of the atom along the DP that begins at the field-free ground state (i.e., DP0) with its branches. The vertical lines mark the peak-field positions of the relevant Floquet states. We note that the features of these spectra are not located precisely at the peak-field positions of the Floquet states. This is because photoelectron emission occurs all along the DP, not just at the peak field intensity. Panel (a) shows the lowest intensity result, and one observes signals from only the first two states, $\nu = 0$ and $\nu = 6$, comprising DP0. Panel (b) shows that as the intensity is increased, the wavepacket clearly accesses the next state, $\nu = 4$, along DPO, as well as a strong signal from $\nu = 6$ remaining. At the next intensity, 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 shake-up to $\nu = 6$ followed by a diabatic transition to $\nu = 0$. At $\alpha_0 = 3.8$, shown in panel (d) of Fig. 3, one observes a strong signal near $\nu = 4$ and LIS1, but one cannot resolve the individual signals from these two Floquet states.

Figure 4 shows a comparison of two spectra from pulses of the same frequency and peak intensity, but of differing pulse duration. The result in the top panel is from a rapid turn-on of the pulse over 4.5 cycles, while the pulse used in obtaining the results in the second panel is ramped on over 60 cycles. Both have a 20 cycles period where the atom is exposed to the peak intensity of the laser. This shows a clear example of how the evolution of the atom along the diabatic path may be controlled by the pulse.

In Fig. 5 we consider again the value of $\alpha_0 = 3.8$ a.u., a case shown previously in Panel (d) of Fig. 3 when one could not distinguish between contributions of $\nu = 4$ and LIS1. Using a shorter pulse turn-on demonstrates the channeling of population into the diabatic transition, whereas the slower pulse results in a preference for the adiabatic transition. The interference fringes observed here are well understood in the literature [8], as they are interference fringes from electrons emitted into the continuum at different times of the pulse at the same energy, in analogy to "which path" type experiments.

In the top panel of Fig. 6, I return to the spectra shown originally in Fig. 1, but now included are marking for the location of the relevant Floquet states at peak fields. Even at these intensities, one observes clear signals in the photoelectron spectrum from the neutral atom. Floquet states $\nu = 4$ and $\nu = 6$ have strong signals showing a predominance for adiabatic branching along DP0. The broad, asymmetric structure at the low-energy side of the photon band is associated with LIS1. But these electrons are emitted prior to the closing of the channel. The asymmetric shape is doe to variations in the state population and ionization rate

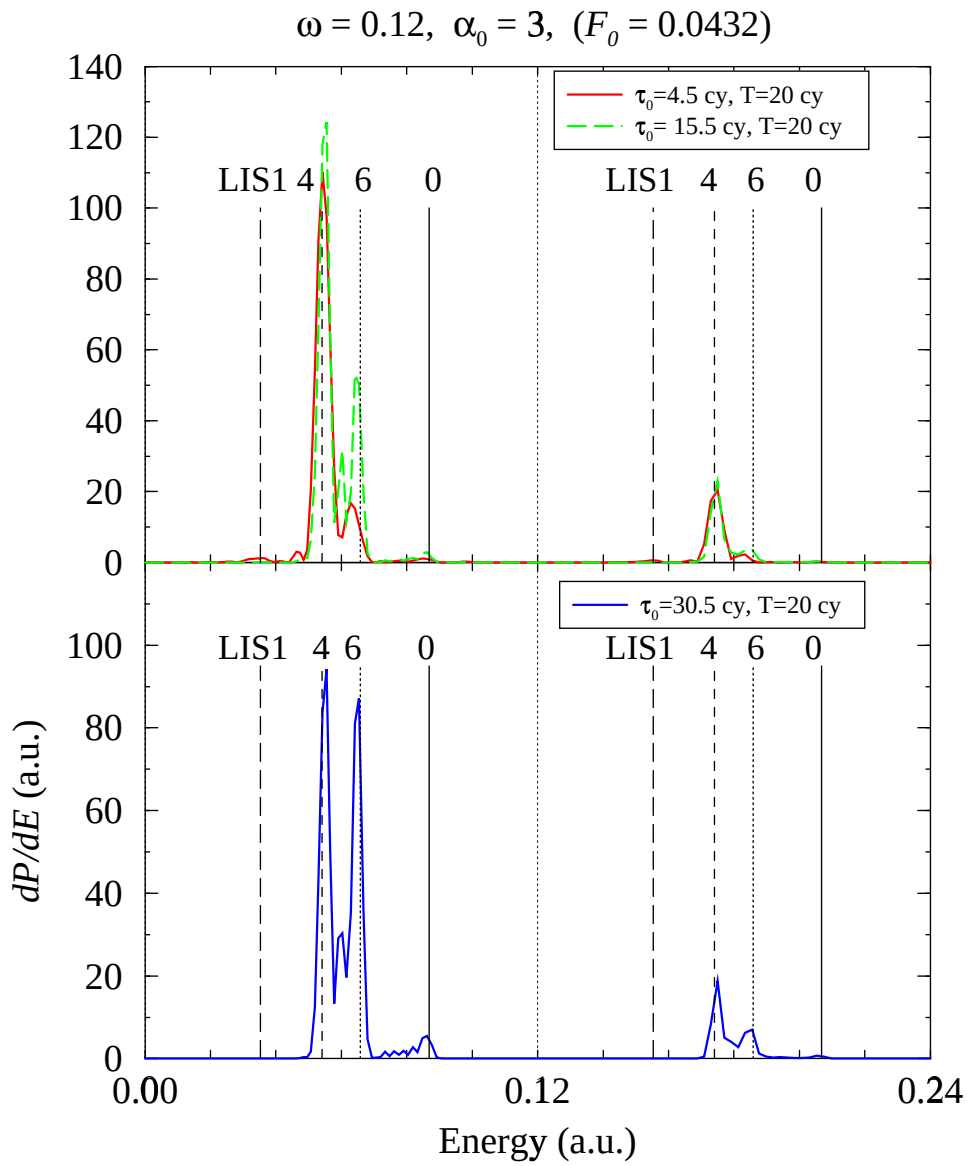

FIGURE 4. EPI/ATI spectra at $\omega = 0.12$a.u., initial state $\nu = 0$, comparing the effect of varying the pulse length.

along the DP0 prior to channel closing for LIS1. The bottom panel of Fig. 6 shows the result of turning the pulse on rapidly for this intensity. In this case, the diabatic branchings are followed predominately, and one observes signals from states $\nu = 1$ and $\nu = 7$. However, when ramping the field on this fast, the atom receives a strong kick projecting part of the probability amplitude into several states. Part of the broad background is from such a shake-up mechanism.

In conclusion, the dual approach of interpreting photoelectron spectra computing

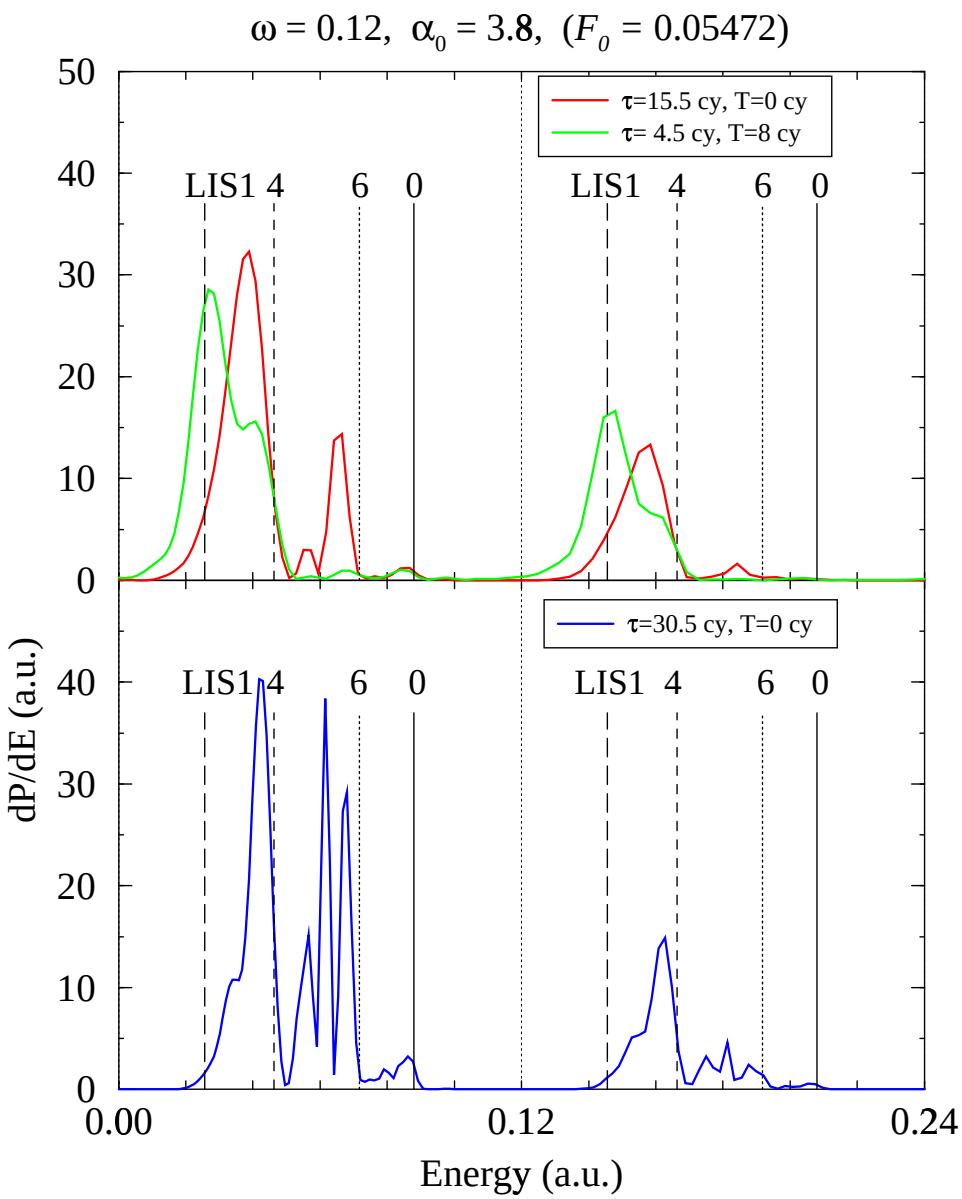

FIGURE 5. EPI/ATI spectra at $\omega = 0.12$a.u., initial state $\nu = 0$, comparing the effect of varying the pulse length.

via wavepacket dynamics with the use of the Floquet may has provided very illuminating, and is seen to have great promise in interpreting the EPI/ATI spectra produce in the strong-field regime. We have observed that the intense-field EPI/ATI spectra reveal the probabilistic evolution of the atom along diabatic paths with shake-up and diabatic transitions playing significant roles. We have demonstrated the possibility of controlling the evolution of the wavepacket along the diabatic paths by shaping the pulses. The EPI/ATI spectra are observed to reflect the structure of the atom during the pulse. For the case of intense fields, it would

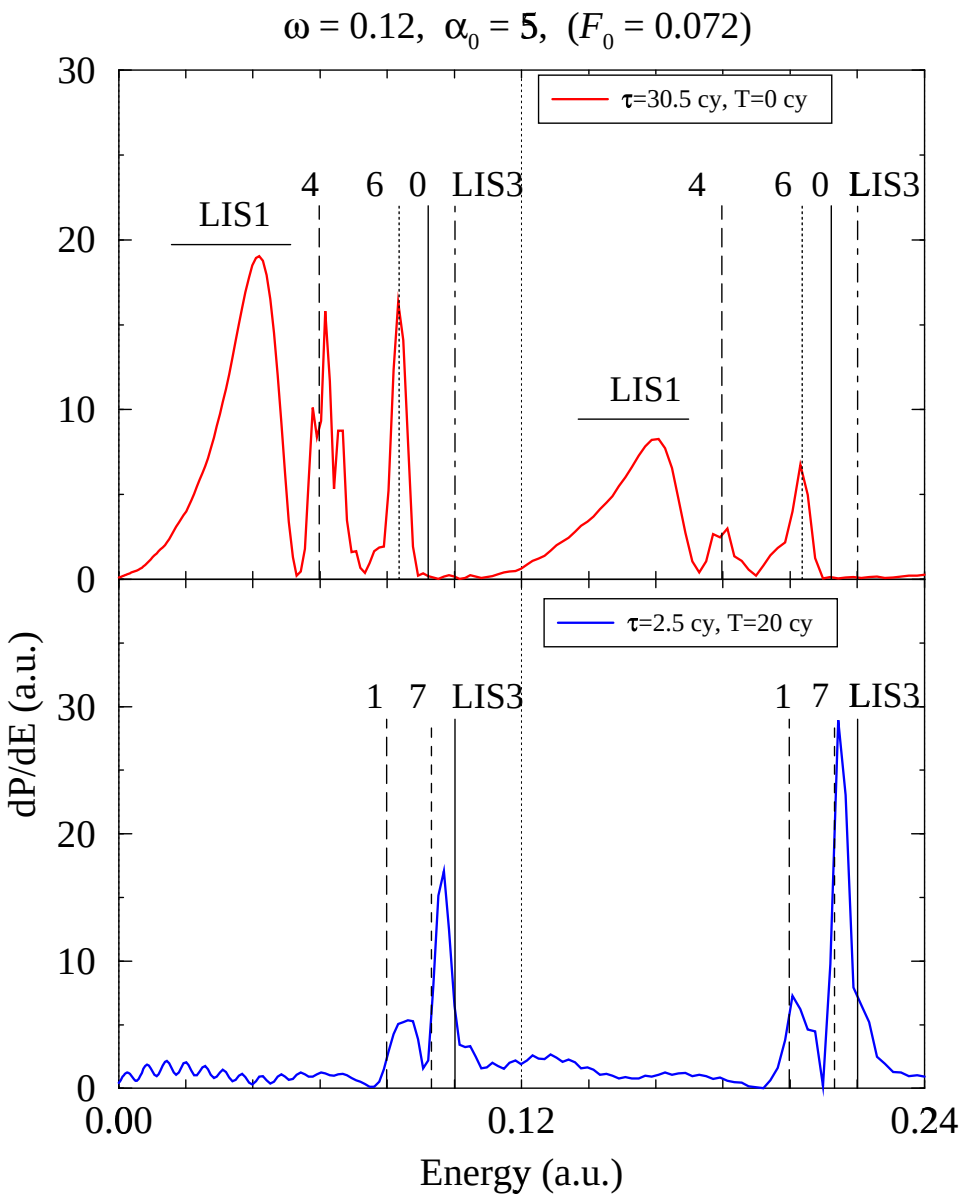

FIGURE 6. (*top*) EPI/ATI spectra as in Fig. 1, and (*bottom*) for a much shorter pulse.

be misleading to interpret the photoelectron spectra by referring to the field-free atomic states. Significant ionization occurs all along the diabatic paths, not only at multiphoton resonances. Moreover, we conclude that the LIS have physical reality, that is, they yield well-defined signals in the photoelectron spectra, and constitute part of (one or several) DP.

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