



Strong-Field Photoionization: Analysis of Overlapping Above-Threshold Ionization and Laser-Assisted Photoemission Structures [†]

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Abstract

We present a theoretical description of atomic strong-field photoionization. Specifically, we consider an atom driven by a combination of two electromagnetic fields: a high-frequency field assisted by an intense laser of lower frequency. We investigate the photoelectron spectrum (PES) as the sum of two contributions: direct ionization due to the laser field and the photoionization term associated with the high-frequency field. We identify the contributions of above-threshold ionization (ATI) and laser-assisted photoemission (LAPE) structures in the total spectra, even when they overlap. As a particular case, we investigate the situation where an ATI peak coincides with a sideband. Our theoretical scheme for hydrogen initially in the 1s quantum state and based on strong field approximation is general enough to be applied to other atomic species and field configurations.

Keywords: strong field; photoionization; ATI; multiphotonic; LAPE; sideband; coherent sum; incoherent sum; pathway interference

1. Introduction

The interaction of strong and short laser pulses with atoms and molecules has received renewed attention, mainly due to advances in laser technology that have enabled new experimental investigations of atomic and molecular processes on ultrashort time scales and under extreme-intensity conditions [1,2]. When an atom or molecule is exposed to an intense laser field, ionization can occur through above-threshold ionization (ATI), a process in which the system absorbs more photons than the minimum required for ionization [1,3,4]. On the other hand, when an extreme ultraviolet (XUV) pulse overlaps in space and time with a laser field in the presence of matter, laser-assisted photoemission (LAPE) processes take place. Depending on the duration of the XUV pulse, two distinct scenarios arise: the sideband and the streaking regimes. Each of these contributes specific structures to the electron emission probability [5,6]. It is important to note that, in the presence of LAPE, direct ionization processes induced by the laser field also occur, among which ATI is a prominent example. ATI and LAPE are fundamentally different processes and, under typical experimental conditions, give rise to well-separated structures in the photoelectron spectrum. In the usual scenario, ATI structures extend from the threshold up to twice the



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ponderomotive energy, while the XUV contribution is approximately centered around the XUV photon energy. By appropriately choosing the XUV frequency, both spectral domains can be kept apart.

However, depending on the parameters of both fields, these structures may overlap. In this work, we explore such overlapping regimes. Several questions arise: Under which conditions can each structure be studied separately? Is it valid to treat them as independent? When does their superposition become relevant? And must this superposition be treated coherently? To address these questions, we employ the strong-field approximation (SFA), a powerful analytical tool, and focus on the hydrogen atom H(1s) as a tested system.

The paper is organized as follows: In Section 2, we briefly resume the SFA theory of ATI and LAPE processes and analyze the properties of the photoelectron spectra (PES) for linearly polarized probe field and pump XUV pulses. In Section 2.3, we analyze the main features and behavior of the PES as the field parameters are varied, identifying the domains where each structure (either ATI or LAPE) dominates, as well as their overlapping region. Concluding remarks are presented in Section 3. Atomic units are used throughout the paper, except where otherwise stated.

2. Theory and Results

Let us consider the ionization of an atomic target by the combination of two EM fields: a high-frequency field (typically in the XUV regime, noted by X) assisted by another intense laser of lower frequency (L). The Hamiltonian of the system in the single active-electron approximation is $H = H_0 + H_{\text{int}}(t)$, where H_0 is the time-independent atomic Hamiltonian and the $H_{\text{int}}(t)$ describes the interaction of the atom with both time-dependent X and L EM fields in the dipole approximation and length gauge: $H_{\text{int}}(t) = \vec{r} \cdot [\vec{F}_X(t) + \vec{F}_L(t)]$.

The electron initially bound in an atomic state ϕ_i is emitted to a final continuum state with final momentum $\vec{k}$, energy $E = k^2/2$ and solid angle Ω . Within the time-dependent distorted wave theory, the energy photoelectron spectra (PES) can be calculated as $dP/dE = \int d\Omega \sqrt{2E} (dP/d\vec{k})$ where the differential emission probability is

$$\frac{dP}{d\vec{k}} = |T_{\text{if}}|^2, \quad \text{and} \quad T_{\text{if}} = -i \int_{-\infty}^{+\infty} dt \langle \phi_f^-(\vec{r}, t) | H_{\text{int}}(\vec{r}, t) | \phi_i(\vec{r}, t) \rangle \quad (1)$$

and where the T -matrix element corresponds to the transition $\phi_i \rightarrow \phi_f$ in the prior form [7,8]. The initial atomic state is $\phi_i(\vec{r}, t) = \phi_i(\vec{r}) e^{iI_p t}$, with ionization potential I_p , and $\phi_f^-(\vec{r}, t)$ is the distorted final state. Within the strong-field approximation (SFA), the Coulomb distortion in the final channel is neglected, as well as the influence of the laser field in the initial ground state. Hence, we can approximate the distorted final state with a Volkov function [1,9], i.e., $\phi_f^-(\vec{r}, t) = \chi_f^V(\vec{r}, t)$, where

$$\chi_f^V(\vec{r}, t) = (2\pi)^{-3/2} \exp(i\vec{\Pi} \cdot \vec{r}) \times \exp\left(\frac{i}{2} \int_t^\infty \vec{\Pi}^2 dt'\right), \quad (2)$$

$\vec{\Pi}(\vec{k}, t) = \vec{k} + \vec{A}(t)$, and the vector potential due to the total external field is defined as $\vec{A}(t) = -\int_{-\infty}^t dt' [\vec{F}_X(t') + \vec{F}_L(t')]$. Since the interaction Hamiltonian has two terms, the transition matrix can also be split into two contributions and written as:

$$T_{\text{if}} = T_X + T_L \quad \text{where} \quad T_{X(L)} = -i \int_{-\infty}^{+\infty} \vec{F}_{X(L)}(t) \cdot \vec{d}[\vec{\Pi}(\vec{k}, t)] e^{iS(\vec{k}, t)} dt \quad (3)$$

for the X or L pulse respectively. Here the generalized action S and dipole moment $\vec{d}$ are defined as

$$S(\vec{k}, t) = I_p t + \int_{-\infty}^t \frac{[\vec{\Pi}(\vec{k}, t')]^2}{2} dt' \quad \text{and} \quad \vec{d}(\vec{\Pi}) = \langle \frac{e^{i\vec{\Pi} \cdot \vec{r}}}{(2\pi)^{3/2}} | \vec{r} | \varphi(\vec{r}) \rangle. \quad (4)$$

To incorporate the Coulomb distortion, another common approximation known as the Coulomb–Volkov approximation (CVA) can be used. In this case, the plane wave is replaced by a Coulomb wave function in the dipole moment $\vec{d}$, as shown in Equation (4). The CVA has been successfully tested against ab initio time-dependent Schrödinger equation (TDSE) results under a wide variety of conditions, especially in the multiphotonic regime [8,10,11]. Since the CVA and the SFA share the same Volkov phase, both approximations predict the same emergence and positioning of multiphotonic structures in the spectra, albeit with a different modulation due to the Coulomb distortion factor that is absent in the SFA.

The total potential vector is the sum of two linearly polarized pulses of duration τ_i , polarization $\hat{\epsilon}_i$ and carrier frequency ω_i , where $i = X$ or L respectively, then $\vec{A} = \vec{A}_X + \vec{A}_L$, where each term is modeled as

$$\vec{A}_i = \frac{F_{i0}}{\omega_i} f_i(t) \sin[\omega_i(t - t_{0i})] \hat{\epsilon}_i. \quad (5)$$

The proposed envelope function is $f_i(t) = \cos^2[\pi(t - t_{0i})/\tau_i]$ only during the temporal interval $(t_{0i} - \tau_i/2, t_{0i} + \tau_i/2)$ and vanishes outside this. Then, the electric field peak amplitude F_{i0} is reached at the middle of the pulse, i.e., at time t_{0i} . Additionally, we consider each pulse duration as an integer number of its oscillation cycle: $\tau_i = N_i 2\pi/\omega_i$. Therefore, due to this finite duration, the time integration in Equation (3) also reduces from infinity to a finite interval of duration τ_i .

In the following, we first separately consider each term of the transition matrix in Equation (3) and its relationship with ATI and LAPE processes to finally address the total spectrum as a sum of both contributions simultaneously. In order to present specific results, we perform our calculations for the case of a hydrogen atom initially in the 1s state and for EM fields with the same polarization direction along $\hat{z}$ ($\hat{\epsilon}_L = \hat{\epsilon}_X = \hat{z}$). Our theoretical scheme can be applied, however, to other atomic species and field configurations as well.

2.1. ATI Review

By setting $F_{X0} = 0$ in the previous section, we recover the strong-field ionization process by the field L only. Then, we introduce the transition matrix for ATI as: $T^{\text{ATI}} = T_L|_{F_{X0}=0}$. It is well known that ATI takes place when the multiphotonic regime is achieved, then the atom can absorb more photons than the minimum needed to ionize. The PES presents the well-known ATI peaks at energy values given by:

$$E_n = n\omega_L - I_p - U_p \quad (6)$$

with $U_p = (F_{L0}/2\omega_L)^2$. There are several works that have studied the characteristics of ATI peaks (height, width, shape, position, etc.) as a function of the laser profile; see, for example, [1,3,4,8,10] for more details. One important point is that in perturbative conditions the n -photon absorption gives ionization rates proportional to the n th power of the laser intensity; then, by increasing the laser intensity, the height of each ATI peak not only increases, but more photons also become available to be absorbed, resulting in the appearance of more ATI peaks with an increasing number n in Equation (6).

As an example, we present the PES for the ATI process in Figure 1a for a particular case of ionization of H(1s). We compare our SFA model (orange solid line) with TDSE (black dots) and CVA (dark blue dashed line) results. For the three models, we observe the

presence of three ATI peaks corresponding to the absorption of two, three and four photons of frequency ω_L . The fifth photon absorption is only predicted by the TDSE, with a rate approximately 10^{-10} times smaller than the second one, owing to the different modulation of the structures in the PES. Nevertheless, a good qualitative agreement of the spectrum can still be observed, even across the different modulations of each model.

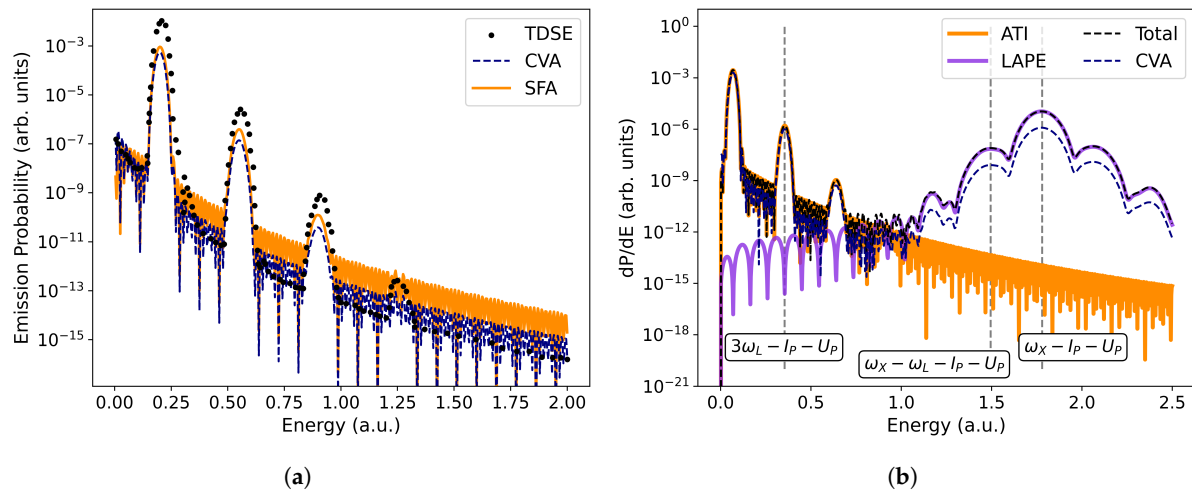


Figure 1. PES for H(1s) as a function of the photoelectron energy for $F_{L0} = 0.01$ a.u. and $N_L = 20$. (a) ATI for $\omega_L = 0.35$ a.u. within SFA (orange line), CVA (dark blue dashed line) and TDSE models (black dots from [11]). (b) SFA results considering only T^{ATI} (orange line), only T^{LAPE} (purple line) and the coherent sum in Equation (9) [idem Equation (10)] shown in black dashed line. Other field parameters are $\omega_L = 0.285$ a.u., $\omega_X = 8\omega_L$, $F_{X0} = 0.001$ a.u., $N_X = 24$ and $t_{0X} = t_{0L}$. Vertical dashed lines indicate the energy position corresponding to the boxed value. CVA results shown in dark blue dashed line.

2.2. LAPE Review

When the high-frequency field is also present, the PES presents specific structures corresponding to the absorption of the high-energy photon. In the sideband regime, it has been shown that the intercycle interference due to the coherent electronic emission at each optical cycle of the laser gives rise to the sideband peaks in the PES at energy values

$$E_m = \omega_X + m\omega_L - I_p - U_p, \quad (7)$$

corresponding to the absorption (positive m) or emission (negative m) of m laser photons, following the absorption of one X photon (see for example [5,6,12]). What is more, this intercycle structure is modulated by the intracycle factor restricting the spectrum to the energy domain established by the semiclassical model between limits $E_{\text{low,up}} = (v_0 \mp F_{L0}/\omega_L)^2/2$ (where $v_0^2/2 = \omega_X - I_p$ corresponds to the mean energy of the photoelectrons ionized by the X pulse in the absence of the laser). Similarly to the ATI process, the emergence of sideband peaks is related to the laser intensity: for stronger fields, more photon exchanges become possible, thereby increasing the maximum value of $|m|$ in Equation (7).

To analytically demonstrate these features, the authors of [5,6,12] have worked out the transition matrix T_X assuming that, as the frequency of the X pulse is much higher than the L one, and considering that the strength of the first one is much smaller than the laser one, the X contribution to the total vector potential can be neglected ($\vec{A}_X \simeq 0$). Furthermore, within the rotating wave approximation which accounts for the absorption of only one X photon and neglects the contribution of its emission, the X electric field becomes $\vec{F}_X(t) \simeq F_{X0}(t)e^{-i\omega_X t} \hat{\epsilon}_X$. Then, they have finally approximated T_X by

$$T^{\text{LAPE}} = T_X|_{\vec{A}=\vec{A}_L} = -i \int_{-\infty}^{+\infty} F_{X0}(t) \hat{\epsilon}_X \cdot \vec{d}[\vec{k} + \vec{A}_L(t)] e^{iS_{LX}(\vec{k},t)} dt, \quad (8)$$

where $S_{LX}(\vec{k}, t) = (I_p - \omega_X)t + \int_{-\infty}^t dt' [\vec{k} + \vec{A}_L(t')]^2/2$, and $F_{X0}(t)$ is the time-dependent amplitude of the X electric field.

In Figure 1b we show the LAPE spectra in a solid purple line for the pulse parameters specified in its caption. We observe that, for such laser intensity, only the sideband peaks corresponding to $m = 0, \pm 1, \pm 2$ can be clearly distinguished in the PES. Outside this range, only oscillatory background behaviour corresponding to the Fourier transform of the pulse appears. Furthermore, we also find that the energy domain of the LAPE process is well separated from the domain of ionization by the laser alone (orange line). In other words, the contribution of direct laser ionization is negligible in the energy domain where the absorption of one X photon takes place. In the following section we will vary the parameters of both pulses to study the superposition of structures in PES.

2.3. Overlapping ATI and LAPE Processes

As we have written in Equations (1) and (3), due to the combination of both X and L pulses, the ionization probability corresponds to the coherent sum of T_X and T_L . However, as we already discussed in the previous sections, with the appropriate choice of the X and L pulse parameters, each term can be approximated by T^{LAPE} and T^{ATI} respectively. Even more, if the spectra do not overlap in the energy domain, the sum can be made incoherently:

$$dP/d\vec{k} = |T_X + T_L|^2 \quad \text{coherent sum} \quad (9)$$

$$\simeq |T^{\text{LAPE}}|^2 + |T^{\text{ATI}}|^2 \quad \text{incoherent sum} \quad (10)$$

Provided the assumptions of the approximations made in Equation (10) are satisfied, both Equations (9) and (10) will yield equivalent results. This is shown, for example, in Figure 1b, where the dashed line indicates that the total ionization probabilities (sum of the ATI and LAPE spectra, represented by orange and purple lines, respectively) produce exactly the same curve using both Equations (9) and (10), both plotted with dashed lines. Here we have also included CVA results; although SFA and CVA models differ in the modulation of PES as a function of electron energy (as expected), the same qualitative description is obtained.

However, a reasonable question would be to inquire as to whether Equation (10) remains valid in other parameter regimes. In order to explore it beyond its range of applicability, we present in Figure 2 the PES as a function of the amplitude F_{L0} and frequency ω_X . The amplitude of the L field is related to the width of the ATI and LAPE structures, i.e., the energy range where the ATI peaks or sidebands arise. Therefore, increasing the amplitude F_{L0} also increases their energy domain. At a certain point, they may overlap. This is precisely what is observed in Figure 2a: as F_{L0} increases, more photons become available, giving rise to additional ATI peaks and sidebands. Meanwhile, for low amplitudes, ATI peaks are spectrally separated from the X photon absorption peak and its sidebands. On the other hand, it can also be seen that the position of the peaks does not remain at the same energy; instead, they shift quadratically toward lower energies due to the ponderomotive shift $-U_p$ [see Equation (6)].

Figure 2b shows the PES as a function of the frequency of the X field. Since the position of the sidebands depends on ω_X [see Equation (7)], decreasing it allows the spectra to overlap. This fact can be seen in the figure: the structures that follow diagonal lines correspond to the absorption peak of the X photon accompanied by its sidebands. In contrast, at low energies, the ATI peaks remain constant, appearing as horizontal lines.

A third line (white dashed line) with a steeper slope (2 a.u.) can also be observed. This maximum corresponds to the absorption of two X photons.

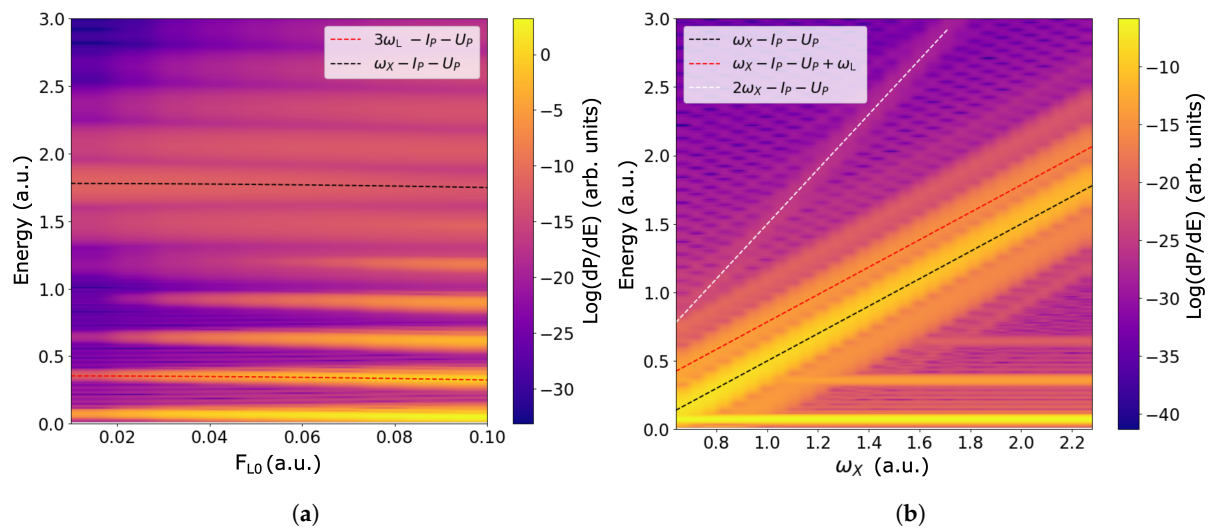


Figure 2. SFA H(1s) PES in color scale for angle integrated in Equation (9): (a) as a function of the laser peak amplitude F_{L0} and (b) as a function of the frequency ω_X . Other field parameters are idem in Figure 1b.

From Figure 2 we observe that, even when overlap occurs, ATI and LAPE structures remain unchanged. In this sense, the total spectrum resembles the incoherent sum. This seems to be the general rule: Equation (10) satisfactorily reproduces Equation (9) in most of the investigated range, except obviously for the two X photon zones.

In what follows, we look for particular conditions under which a noticeable difference between the PESs from Equations (9) and (10) arises. To this end, we aim for a scenario where a sideband peak coincides in the energy position of an ATI peak, and both have comparable heights. In Figure 3a, we show the spectra for an emission direction parallel to both polarization vectors. After carefully choosing specific parameters, we finally observe a case where the coherent and incoherent sums differ, even if only in a very localized region of the spectrum. In the coherently summed spectrum (purple line), one can observe the suppression of the peak at energy 0.64 a.u., which corresponds both to the ATI peak with $n = 4$ [see Equation (6)] and to the $m = -1$ sideband [see Equation (7)]. In contrast, Equation (10) yields a simple sum of both contributions. This could be interpreted as quantum interference between the different pathways leading to the same final energy: absorbing four photons of the laser, or absorbing one X photon and emitting one L photon. Figure 3b shows that upon angle integration, this destructive interference is erased. Moreover, a slight change in the delay between the two pulses also makes the interference disappear. This behavior closely resembles that observed in a RABBIT protocol, where sidebands exhibit a suppression pattern depending on the delay between pulses [13,14]. Apart from this, since the height of both ATI peaks and sidebands is very sensitive to the model employed, this quantum destructive interference within the CVA or TDSE approach is expected to be found for another set of slightly different laser parameters. This finding requires deeper analysis, which will be explored in a future work.

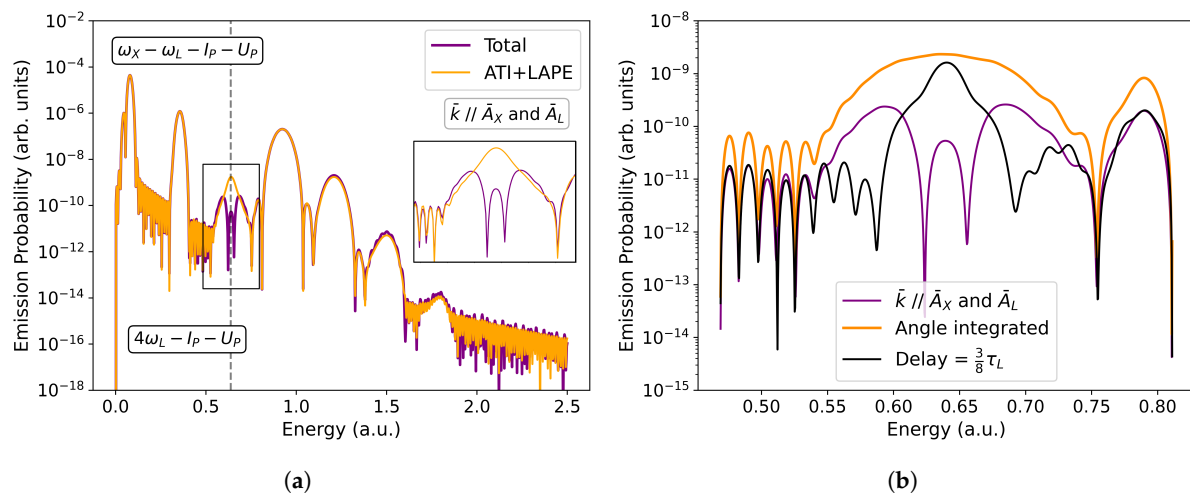


Figure 3. SFA differential emission probability for H(1s) corresponding to forward emission: $\vec{k} \parallel \hat{z}$. Laser pulse idem to Figure 1. For the X field, the parameters are $\omega_X = 5\omega_L$ a.u., $F_{X0} = F_{L0}/150$, $N_X = 25$ and $t_{0X} = t_{0L}$. **(a)** Equation (9) in purple line and Equation (10) in orange line. **(b)** Zoom of panel (a): Equation (9) in purple line idem to panel (a); coherent PES Equation (9) integrated in emission angles shown in orange line and black line shows coherent Equation (9) with delay $t_{0X} - t_{0L} = \frac{3}{8}\tau_L$.

3. Conclusions

We have studied strong-field photoionization within the framework of the strong-field approximation (SFA), considering the electromagnetic field as the sum of two pulses: a high-frequency contribution (typically in the XUV regime) assisted by an intense laser of lower frequency. From the analytic properties derived from the SFA transition matrix element, we have identified the contributions of the above-threshold ionization (ATI) and laser-assisted photoemission (LAPE) transition matrices and their corresponding structures in the total spectra.

After exploring a wide range of parameters, we conclude that, in general terms, the total spectrum corresponds to a simple superposition of both ATI and LAPE structures. However, upon closer inspection, in some very specific circumstances, quantum interference between different electron emission pathways can be observed, leading to destructive interference. In particular, we have analyzed the situation where an ATI peak coincides with a sideband, resulting in the suppression of the electron emission probability. Slight variations in the laser parameters cause this destructive interference to disappear. The detailed study of this interference phenomenon will be the subject of future research.

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