Coherent Intense Laser Pulses Lead to Interference in the Time Domain: Dynamic Interference of Electron Waves

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Abstract: We investigate how the time dependence of the field-induced couplings between electronic states of a system, exposed to strong laser pulses, influences spectroscopic observables. It is demonstrated that the time-envelope of a coherent laser pulse gives rise to two waves emitted with a time delay with respect to each other at the rising and falling sides of the pulse, which interfere in the time domain. By considering two show-case examples, the ionization of an atom by a high-frequency intense laser pulse and the sequential two-photon ionization of an atom by a resonant laser pulse, we show that dynamic interference is a very general phenomenon in the spectroscopy of strong laser fields.

1. Introduction

The interaction of an atom with intense laser fields has been widely studied. If the field is essentially monochromatic, the physics is well described by a time-independent Hamiltonian in the basis of `dressed' electronic states or Floquet states [1]. The inclusion of relaxation mechanisms, such as spontaneous emission, autoionization, or subsequent ionization, gives a `dressed' state a finite width, and it becomes unstable [2]. The concept of dressed states is applied in practically every branch of spectroscopy of optical lasers operating in the nano and picosecond regimes.

If the laser pulses are shorter, a Floquet basis is still useful, but one has to take the time-dependence of the pulse explicitly into account. Many new phenomena arise due to the impact of this time-dependence [3,4]. The situation becomes particularly interesting by the promise of the new generation of light sources, like femtosecond [3] and attosecond [4] lasers, high-order harmonic generation sources [5,6], and free electron lasers [7,8] to produce ultrashort and intense coherent laser pulses of high frequencies. Due to the high carrier frequencies, much of the physics follows the evolution provided by the pulse envelope nearly adiabatically up to rather short pulse durations. The pulse envelope enters explicitly the coupling between the electronic states and determines the evolution induced by the field [9,10].

In the present work we discuss a fundamental consequence of the finite nature of intense coherent laser pulses of a high frequency on spectroscopic observables. We demonstrate that the time-envelope of a coherent laser pulse gives rise to two waves emitted with a time delay with respect to each other at the rising and falling sides of the pulse, which interfere in the time domain [11,12]. Dynamic interference is a very general consequence of the finite nature of intense high-frequency laser pulses, and leads to pronounced patterns observable in the spectrum of the emitted particles (see also Figs. 1 and 2). As soon as the field-induced couplings between electronic states of a system possess the nontrivial time dependence provided by the pulse, dynamic interference takes place. In order to prove these statements, we consider two show-case examples of the ionization of an atom by a high-frequency intense laser pulse and of the sequential two-photon ionization of an atom by a resonant laser pulse.

2. Bound-continuum transition

We, first, discuss the ionization of an atom by a high-frequency intense laser pulse, where the energy of a single-photon is sufficient to ionize the system. It is shown that the ‘dressed’ ground state embedded into the electronic continuum experiences a time-dependent AC Stark shift [11]. This shift arises from the indirect coupling of nonresonant (nonessential) states that do not participate directly in the ionization process. As a consequence, the energy of the ‘dressed’ ground state and also the energy of the emitted photoelectron adiabatically follow the envelope of the laser pulse. This is demonstrated to result in strong dynamic interference of the photoelectrons of the same kinetic energy emitted at different times, when the pulse arrives and expires, respectively [11].

Numerically exact computations on the hydrogen atom demonstrate that the dynamic interference spectacularly modifies the photoionization process and is prominently manifested in the photoelectron spectrum by the appearance of a distinct multi-peak pattern. The photoelectron spectrum computed exactly for the hydrogen atom exposed to Gaussian-shaped pulse is shown in Fig. 1 as an example. The general theory is shown to be well approximated by explicit analytical expressions, which allow for a transparent understanding of the discovered phenomena and for making predictions on the dependence of the measured spectrum on the properties of the pulse.

3. Bound-bound transition

Now, let us consider two bound electronic states of an atom coupled resonantly by a short strong laser pulse. The dynamics of these states can be understood in terms of the decoupled resonances scenario [12]. These are the solutions of a 2×2 Hamiltonian for the ground and excited states coupled by the resonant laser pulse. This field-induced coupling between two initially degenerate `dressed' electronic states adiabatically follows the pulse

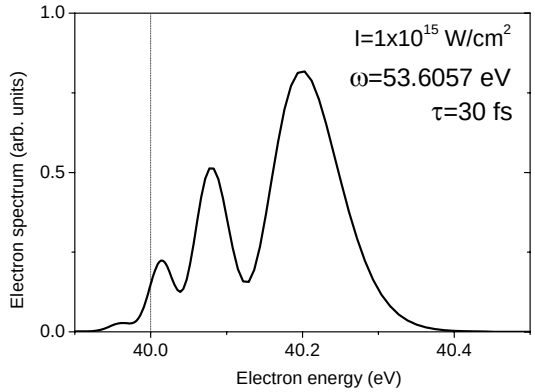

Fig. 1. Single-photon ionization spectrum of H exposed to a Gaussian pulse of 30 fs duration with carrier frequency 53.6057 eV and peak intensity of 5×10^{15} W/cm^2. In weak-field single-photon ionization, the photoelectron line appears as a Gaussian peak centered around the energy of $\varepsilon_0 = \omega$ - IP = 40 eV (indicated by the vertical line) and a width of about 52 meV FWHM for the 30 fs pulse. The presently computed strong pulse ionization spectrum is, however, much broader (by several times), shifted to higher energies from the electron energy of 40 eV, and posses distinct modulations of the electron yield, which are owing to dynamic interference.

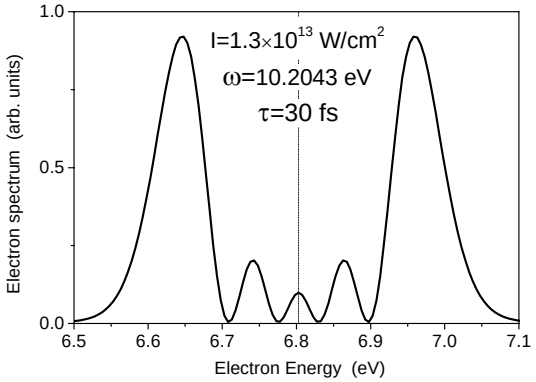

Fig. 2. Sequential two-photon ionization spectrum of H exposed to a Gaussian pulse of 30 fs duration with carrier frequency 10.2043 eV and peak intensity of 1.3×10^{13} W/cm^2. At this intensity, the atom manages to complete five Rabi cycles between the ground and excited states during the time the pulse is on. The presently computed sequential two-photon ionization spectrum is very different from that expected in the weak field regime. It is much broader than the width provided by the pulse and, owing to dynamic interference, exhibits a distinct multiple peak pattern which is symmetrically centered around the electron energy of $\varepsilon_0 = 2\omega$ - IP = 6.8029 eV.

envelope. When the pulse is already on, these solutions describe two decoupled time-independent resonances with time-dependent energies given by the field-induced coupling. Their energies move apart as the pulse arrives (the energy splitting between them adiabatically increases), and then move towards each other as the pulse expires (the energy splitting adiabatically decreases). If the atom emits particles during its exposure to the pulse (photoelectrons, Auger electrons, photons), these particles emitted when the pulse rises have the same kinetic energy as those emitted at a later time when the pulse decreases. The respective two waves emitted with a time delay with respect to each other will interfere and their spectrum will exhibit a pronounced interference pattern [12].

As an example, we consider the sequential two-photon ionization of an atom by strong coherent pulses of resonant frequency. Here, the coupled two-level system is probed by a second photon of the same pump pulse. Fig. 2 depicts the photoelectron spectrum computed for the sequential two-photon ionization of hydrogen atom exposed to the Gaussian-shaped pulse of the resonant carrier frequency, which fits to the energy of the H(1s)-H(2p) excitation. One can see that the dynamic interference spectacularly modifies the sequential two-photon ionization process and is prominently manifested in the spectrum by the appearance of a distinct multi-peak pattern.

4. Conclusions

We are convinced that dynamic interference is a very general and fundamental effect, which is best manifested in the observable spectrum of the emitted particles by prominent multiple-peak patterns. In many cases the dynamics of states coupled by an intense laser pulse follows the time envelope of the pulse. This dynamics can be probed by particles emitted in the process of this dynamics, either by employing an additional probe pulse, or as we discussed above, by the same pulse. The emitted particles do not have to be photoelectrons. They can be, e.g., Auger electrons or photons. We also briefly discuss the effect of dynamic interference in other branches of laser spectroscopy, such as resonant Auger effect, autoionization, and resonant X-ray emission. Our results pave the way for experiments on dynamic interference by available laser pulse sources.

5. References

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