Chemical Physics 414 (2013) 53–64

Contents lists available at SciVerse ScienceDirect

Chemical Physics

journal homepage: www.elsevier.com/locate/chemphys

Theory of attosecond delays in laser-assisted photoionization

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ARTICLE INFO

Article history:
Available online 9 February 2012

Keywords:
Attosecond
Photoionization
Laser-assisted
High-order harmonics
Delay in photoionization
Phase of ATI
Above-threshold ionization
Attosecond pulse characterization
Atomic phase
Atomic delay
Steaking
RABITT
RABBIT
PROOF
Temporal characterization

ABSTRACT

We study the temporal aspects of laser-assisted extreme ultraviolet (XUV) photoionization using attosecond pulses of harmonic radiation. The aim of this paper is to establish the general form of the *phase* of the relevant transition amplitudes and to make the connection with the time-delays that have been recently measured in experiments. We find that the overall phase contains two distinct types of contributions: one is expressed in terms of the phase-shifts of the photoelectron continuum wavefunction while the other is linked to continuum–continuum transitions induced by the infrared (IR) laser probe. Our formalism applies to both kinds of measurements reported so far, namely the ones using attosecond pulse trains of XUV harmonics and the others based on the use of isolated attosecond pulses (streaking). The connection between the phases and the time-delays is established with the help of finite difference approximations to the energy derivatives of the phases. The observed time-delay is a sum of two components: a one-photon Wigner-like delay and a *universal* delay that originates from the probing process itself.

1. Introduction

The dynamics of photoionization can now be explored with unprecedented time resolution thanks to high-order harmonic-based sources that deliver pulses of XUV radiation with duration in the attosecond range. Recent measurements performed with single attosecond pulses have shown the existence of an unexpected time-delay between the single-photon ionization from the 2s and the 2p sub-shells of Ne atoms in gas phase [1]. The "streak camera" technique used in these experiments [2] implied nontrivial ejection times of the photoelectrons, depending on the sub-shell from which they originate. Similar delays between the ejection times from the 3s and 3p sub-shells in Ar have been measured also using trains of attosecond pulses [3], with the help of another technique based on interferometry called RABBIT (Reconstruction of Attosecond Beating By Interference of Two-photon transitions) [4–6]. In both cases, delays of several tens of attoseconds have been measured. As photoionization is one of the most fundamental processes in light-matter interactions, these results have motivated a large number of theoretical investigations [7–13].

The two kinds of measurements share many similarities since they involve a laser-assisted single-photon ionization process and they rely on a phase-locked IR laser field to probe the temporal aspects of the XUV photoionization. However, they differ in the analysis used to determine the time-delays and in the range of IR laser intensity.

The motivation of the present paper is to present an unified theoretical analysis of these processes. To achieve this goal, we shall expose first the theoretical background which has conducted us to conclude in [3], that in interferometric measurements, the measured delays arise from the combination of two distinct contributions: one is related to the electronic structure of the atomic target while the other is induced by the measurement process itself. The first one can be identified as a "Wigner time-delay" [14,15] that is directly related to the energy dependence of the different phase-shifts experienced by the photoelectrons ionized from

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doi:10.1016/j.chemphys.2012.01.017

distinct sub-shells in atoms. The other contribution is induced by the IR laser field that is used to probe the photoionization process. This latter contribution results from the continuum–continuum transitions induced by the probe IR laser field in the presence of the Coulomb potential of the ionic core. When simplifying the analysis to the cases when the process is dominated by the asymptotic form of the relevant second-order matrix elements, a characteristic measurement-induced delay can be identified, that is independent from the details of the electronic structure of the ionic core. This shows how the experimental signal can be related to the temporal dynamics of one-photon ionization.

Regarding the streaking measurements realized with a single attosecond pulse of XUV radiation [1], the experimental data were obtained for IR field intensities significantly higher than those obtained with attosecond pulse trains [3]. Understandably, the questions related to the role of the probe IR field on the photoelectron dynamics in streaking measurements have motivated several theoretical studies [7–13]; see also the earlier papers: [16–19]. Then, a natural issue arises which is to determine to what extent the "streaking delays" so obtained differ from those derived from the interferometric data. Although both the experimental techniques and the theory treatments differ, it is of interest to compare the two approaches. Indeed, as we shall show below, a link can be found when reducing the laser intensity of the streaking field so that one reaches the domain of applicability of the recently developed Phase-Retrieval by Omega Oscillation Filtering (PROOF) scheme, [20]. An interesting outcome of our analysis is to show the importance of the long-range Coulomb potential for understanding the absolute time-delays in the streaking experiments as well.

The interpretation of the attosecond delays in photoionization relies on our ability to determine the *phases* of the relevant transition amplitudes. Thus, before going into the details of the derivation of such phases, we shall outline the main features of the two techniques in Section 2. Then, Section 3 is devoted to the presentation of the general expressions for two-color, two-photon, complex transition amplitudes that are relevant for Above-Threshold Ionization (ATI) in single-active electron systems. The theoretical background is based on a perturbative approach and the emphasis will be on the derivation of a closed-form approximate expression that is of interest for evaluating the phase of the amplitudes. The basis of exact computations in hydrogen will be outlined, and a simplified classical treatment will be presented. Applications to the determination of the relation between the phases and the time-delays is presented in Section 4. Here we consider first ionization by an attosecond pulse train and then by a single attosecond pulse, in the presence of a relatively weak IR field. This discussion provides an interesting connection between the two types of measurements. Section 5 contains a comparison of the results extracted from the approximate evaluation of the delays to the ones deduced from exact calculations performed in hydrogen from different initial states. Also, we present our conclusions and perspectives.

2. Laser-assisted XUV photoionization: attosecond pulse train vs. single attosecond pulse

The principle of the measurements of the delays using an attosecond pulse train is illustrated in Fig. 1(a), which represents schematically the ionization of an atom in the simultaneous presence of a set of several XUV (odd) harmonics and of the IR field, used to generate the harmonics (atomic units will be used throughout the paper, unless otherwise stated). In the time domain, both pulses are "long", *i.e.* the IR laser pulse is multi-cycle, with typical duration of a few tens of femtoseconds, and the XUV harmonic field is constituted of a train of attosecond pulses (or equivalently of a comb of coherent odd harmonic frequencies $(2q+1)\omega : H_{2q+1}$). Under these conditions, the photoelectron spectrum consists of equidistant lines separated by 2ω that are associated to one-photon ionization of the target by each harmonic. In-between these lines are sidebands associated to two-photon transitions involving the absorption of one harmonic and the exchange of one IR photon. The signal intensities, S_{2q}, of the sidebands labelled $2q$ vary periodically with the delay τ between the IR and the harmonic pulses, according to a generic expression that involves the phases of the fields together with atom-dependent contributions:

$$S_{2q} = \alpha + \beta \cos[2\omega\tau - \Delta\phi_{2q} - \Delta\theta_{2q}], \tag{1}$$

where $\Delta\phi_{2q} = (\phi_{2q+1} - \phi_{2q-1})$ is the phase difference between the consecutive harmonics H_{2q+1} and H_{2q-1} and $\Delta\theta_{2q}$is an intrinsic atomic quantity, associated to the difference of the phases of the transition amplitudes associated to the distinct quantum paths leading to the sideband [5].

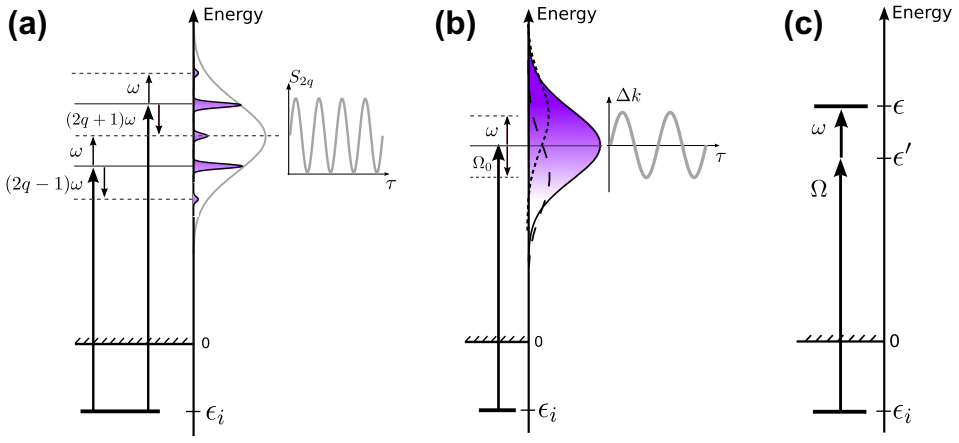

Fig. 1. (a) Laser-assisted photoionization by an attosecond pulse train, corresponding to odd XUV harmonics, H_{2q-1}, where q is a positive integer. The sideband S_{2q} can be reached by either absorbing H_{2q-1} and then absorbing a IR laser photon ω or by absorbing H_{2q+1} and then emitting ω. The sideband signal, S_{2q}, oscillates as a function of the subcycle-delay, τ, between the attosecond pulses and the IR laser probe field. (b) Laser-assisted photoionization by a single attosecond pulse, corresponding to a broad XUV continuum. To first order, the electron is ionized by absorbing one XUV photon, Ω, resulting in a wave packet centered at $\epsilon = \epsilon_i + \Omega_0$. To second order, the electron may absorb an additional laser photon ω resulting in an upshifted wave packet centered at $\epsilon = \epsilon_i + \Omega_0 + \omega$; or it may emit a laser photon resulting in a downshifted wave packet centered at $\epsilon = \epsilon_i + \Omega_0 - \omega$. The interference of these three wave packets leads to a modulation, Δk, of central momentum of the photoelectron as function of the subcycle-delay, τ, between the laser field and the attosecond pulse. (c) Two-photon XUV-IR Above-Threshold Ionization from an initial bound state with energy ϵ_i, to a final state with energy ϵ.