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Argon ionization by laser of fundamental frequency and its second-harmonic: Phase delays

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Abstract

The ionization phases from argon atoms subject to a linearly polarized laser field ($\omega - 2\omega$) setting are calculated. We find excellent agreement between our results and measured phase delays [L. J. Zipp et al, *Optica* **1**, 361].

Since the 1990's, two-color ω - 2ω laser fields with well-controlled relative phases have been studied experimentally and theoretically. One key feature is that the broken inversion symmetry of the ω - 2ω field allows for interference between odd and even partial waves of the outgoing photoelectron. Zipp *et al* [1] extended the measurement of ionization phases and attosecond time delays to the strong-field multiphoton regime. The role of electron wave packets emitted by absorption of subsequent harmonics in the RABBIT is replaced in this ω - 2ω interference protocol by adjacent ATI peaks generated by a strong driving field of frequency 2ω . The concomitant weaker field opens up interfering pathways to sidebands in between neighboring ATI peaks by absorbing or emitting one ω photon. Measuring the ionization as a function of the relative phase between the ω and the 2ω fields, to some extent resembling the original RABBIT protocol, promises to offer insight into the ionization phase and, possibly, timing information of multiphoton processes [2].

In this paper, we present a theoretical study of the ionization phase in the multiphoton regime accessible by two collinearly polarized laser fields of the form $\vec{F}(t) = [F_{2\omega} \cos(2\omega t) + F_{\omega} \cos(\omega t + \phi)] \hat{z}$. In Fig. 1 we show *ab initio* results of the energy spectrum calculated within the time dependent Schrödinger equation (TDSE) in the forward direction as a function of the relative phase ϕ . Phase delays have been extracted by fitting the ATI and sideband with the perturbative prediction [4]. We find a very good agreement between our TDSE calculations and the experiments [1] but strong deviations from the predictions of the strong field approximation (SFA) in dashed vertical lines, clearly indicating that the atomic potential has a crucial influence on the ionization phase of ATI peaks even at energies well above the ionization threshold.

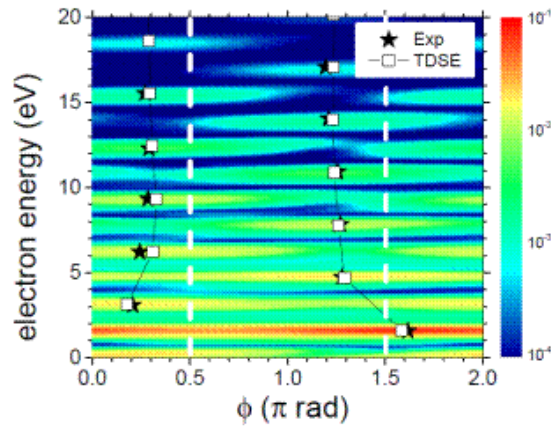


Figure 1: TDSE energy spectrum in the forward direction as a function of the relative phase ϕ . Squares: calculated phase delays and the stars: experiments in [1]. Vertical dashed lines: SFA.

In order to investigate the role of NIR probing field in the determination of the time delays, in Fig. 2 we show the delays calculated for decreasing probe laser intensities ($I_{2\omega}$). It can be noted that the shape of the delays of the ATI peaks substantially changes from the value $I_{2\omega} = 4 \times 10^{11} \text{ W/cm}^2$ to $I_{2\omega} = 1 \times 10^{11} \text{ W/cm}^2$, where the “shoulder” structure around $E = 20 \text{ eV}$ vanishes. As the emission energy increases and the probe intensity decreases, the global trend of ATI delays converges to the strong field limit $\delta = \pi$, corresponding to a time delay of $\delta/2\omega = 666.7 \text{ as}$, (horizontal dashed line). However, contrarily to the SFA predictions [1,4], a delay different from zero is reached for high emission energies at these probing intensities, and a no clear extrapolation can be proposed as a zero probing intensity. Maybe this non-zero limit can be due to delays produced by numerous ionization paths since unlike RABBIT, the present ω - 2ω protocol opens up a multitude of competing quantum paths, turning the extraction and interpretation of interference phases more complex.

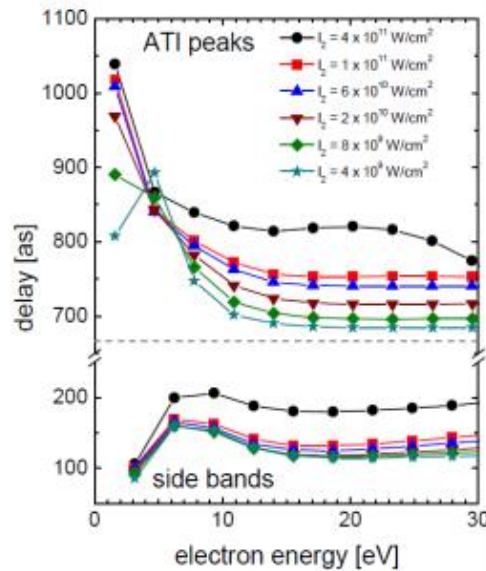


Figure 2: Time delays as a function of the emission energy calculated from asymmetries integrated over half spheres for decreasing NIR intensity.

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References

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