Study of atomic delays in negative ions

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Synopsis The atomic delay in laser-assisted photoionization of negative ions is studied theoretically. It is found that the Wigner-like delay of the photoelectron can be studied directly with attosecond pulse characterization techniques.

Attosecond pulses are traditionally characterized by methods based on laser-assisted photoionization [1, 2]. Further experimental work with these methods have shown that new insight into the photoionization process can be achieved by performing relative delay measurements between different bands in solids [3], atomic orbitals [4, 5], atomic species [6] or angle of photoemission [7]. Theoretically, it has been found that the atomic delay ($\tau_A \approx \tau_W + \tau_{cc}$) can be interpreted as a sum of the photoelectron Wigner delay (τ_W) and a contribution from the laser–ion interaction (τ_{cc}) [8, 9]. The τ_{cc} depends on the effective charge Z of the photoionized target. In the special case of photodetechment from negative ions τ_{cc} is expected to vanish because the photoionized ion is neutral with $Z = 0$. Here, we present work on laser-assisted photoionization of negative ions with the aim to answer the question if it is possible to measure directly the photoelectron delay τ_W from targets that lack long-range interaction.

We have chosen to study the negative ions of fluorine and chlorine for two reasons (i) they both have large affinities and (ii) they are isoelectronic to the much studied rare gases atoms neon and argon. One important difference is that there are no bound excited states in most negative ions and few autodetaching resonances as well. In fact, only one autodetaching resonance in F^- has been found ($2p^4 3s^2$), and, similarly, only one is found in Cl^- ($3p^4 4s^2$) [10]. This is good news because it is understood that Fano resonances in laser-assisted photoionization will invalidate the atomic delay relation when the laser photon is larger than the resonance width [11]. In this work ionization from the outermost shell is performed using the Random Phase approximation with Exchange (RPAE) with a computer program that has been used in earlier studies to compute the atomic delay in noble gas atoms [12]. In Fig. 1 we present the difference $\tau_A - \tau_W$ for both negative ions and noble gas atoms. As expected, we find that the delay difference associted with negative ions are small at high kinetic energy of the photoelectron, so that $\tau_A \approx \tau_W$. However, below 8 eV the delay difference is increasing, thus making it impossible to directly extract τ_W from τ_A by laser-assisted photoionization. Finally, we have studied the effect of reversed time-order processes (IR+XUV). While the dominant process remains the XUV+IR process, the IR+XUV process is not negligible for negative ions. As an example, we find that the reversed process induces a delay of ∼5 as in F^- close to the affinity threshold, while in the noble gas atoms this effect is less than 1 as.

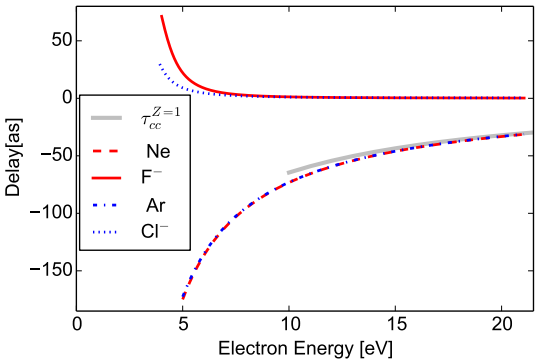

Figure 1. The difference between atomic delay and Wigner delay ($\tau_A - \tau_W$) is computed for negative ions and noble gas atoms.

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