Comment on "Probing Single-Photon Ionization on the Attosecond Time Scale"

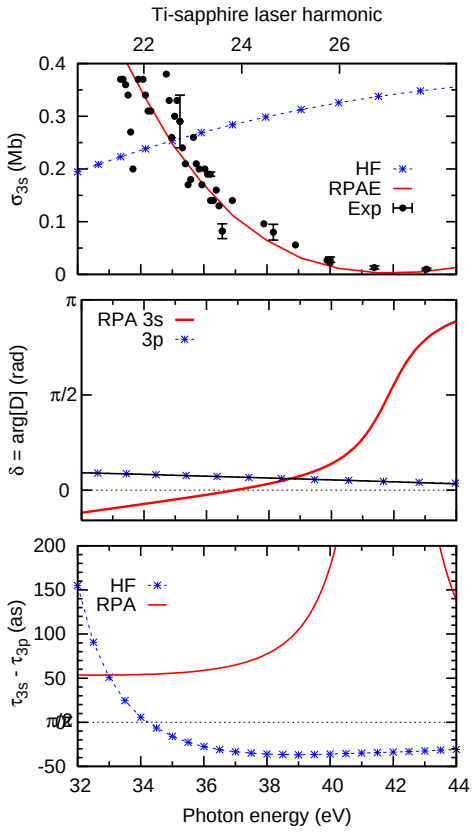
A. S. Kheifets
Research School of Physical Sciences, The Australian National University, Canberra ACT 0200, Australia
(Dated: June 22, 2011)

In a recent Letter, Klünder *et al* [1] connected their attosecond extreme-ultraviolet (XUV) time delay measurements in the valence shell of Ar with the Wigner time delay [2]. In this comment, we show that this interpretation may be too simplistic due to a rapid variation of the phase of the dipole matrix element. This variation is caused by a strong inter-shell correlation which modifies completely the photoionization process in Ar near the $3s$ threshold [3].

The concept of time delay was introduced by Wigner [2] in the context of s-wave quantum scattering. An extension to the total scattering amplitude leads to the concept of angular time delay $\tau_\ell = 2\hbar d\eta_\ell/dE$, where η_ℓ is the phase shift in the ℓth partial wave. One can also define a transmission group delay $\hbar\phi'$, where ϕ is the phase of the complex transmission amplitude $T(k) = t(k)\exp[i\phi(k)]$ [4]. With some modifications, similar concepts apply to photoionization [5]. As compared to particle scattering, the Wigner time delay should be halved as photoionization does not involve ingoing waves. The group delay of the outgoing electron wave packet can be defined as energy derivative of the phase of the complex photoionization matrix element $D(E) = d(E)\exp[i\delta(E)]$. In general, photoionization may involve several strongly interacting channels. Therefore, δ cannot be reduced to a phase shift η_ℓ in some particular ℓth partial wave. Thus, the photoionization group delay is a more general concept than the Wigner time delay. Nevertheless, in some special cases, the Wigner time delay can be conveniently used to characterize delay in photoemission. One such case is valence shell photoionization of Ne in the XUV range [5, 6]. Here there is no considerable coupling between the $2s \to \epsilon p$ and $2p \to \epsilon s$ or ϵd channels and ϵd is strongly dominant over ϵs.

However, the case of valence shell photoionization of Ar, considered by Klünder *et al* [1], is very different. The $3s$ photoionization is completely modified by strong inter-shell correlation with $3p$ [3]. In result, the photoionization cross-section σ_{3s} goes through a deep "Cooper" minimum at the photon energy $\omega \simeq 40$ eV, which is absent in the independent electron Hartree-Fock (HF) model. It is precisely this photon energy range where the measurement of Klünder *et al* [1] was performed (harmonics 22 to 26 of a 800 nm Ti-sapphire laser). It is for this reason that the relative time delay $\tau_{3s} - \tau_{3p}$ cannot be attributed solely to the difference of the Wigner time delays in the dominant $3s \to \epsilon p$ and $3p \to \epsilon d$ partial waves, as was done by Klünder *et al* [1].

We illustrate these findings in the Figure. On the top panel, we plot σ_{3s} calculated in the HF and random-phase (RPA) approximations, the latter taking full account of the inter-shell correlation with $3p$. The most recent experimental data are from Möbus *et al* [7]. On the middle panel, we plot the correlation-induced phase shifts of the dipole matrix elements $\delta_{3s} = \arg[D_{3s}(\omega + \epsilon_{3s})]$ and $\delta_{3p} = \arg[D_{3p}(\omega + \epsilon_{3p})]$ from the same RPA calculation. On the bottom panel, we plot the time delay difference $\tau_{3s} - \tau_{3p} = \hbar\frac{d}{dE}\Big[\eta_{\ell=1}(\omega+\epsilon_{3s}) - \eta_{\ell=2}(\omega+\epsilon_{3p})\Big]$ calculated with elastic scattering phases only (marked HF) and the correlation correction $\hbar\frac{d}{dE}\Big[\delta_{3s}(E) - \delta_{3p}(E)\Big]$ (marked RPA). Due to a rapid variation of the phase of D_{3s}, especially near the "Cooper" minimum, the RPA correction to the measured time delay is substantial. Had this correction been accounted for by Klünder *et al* [1], it would have certainly upset a seemingly good agreement between their measurement and theoretical interpretation based on the Wigner time delay.



[1] K. Klünder *et al*, Phys. Rev. Lett. **106**(14), 143002 (2011).
[2] E. P. Wigner, Phys. Rev. **98**(1), 145 (1955).
[3] M. Y. Amusia, V. K. Ivanov, N. A. Cherepkov, and L. V. Chernysheva, Physics Letters A **40**(5), 361 (1972).
[4] C. A. A. de Carvalho and H. M. Nussenzveig, Phys. Rep. **364**(2), 83 (2002).
[5] M. Schultze *et al*, Science **328**(5986), 1658 (2010).
[6] A. S. Kheifets and I. A. Ivanov, Phys. Rev. Lett. **105**(23), 233002 (2010).
[7] B. Möbus *et al*, Phys. Rev. A **47**(5), 3888 (1993).