Angle-resolved RABBITT : from atoms to molecules

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Synopsis We report recent results of photoelectron angular distributions (PADs) for photoionization (PI) of atomic and molecular targets, resolved in the time domain at the attosecond scale, combining electron-ion coincidence momentum spectroscopy and the RABBITT scheme. We propose a unified formalism synthetizing the $PAD(\theta,\tau)$ results in terms of a reduced set of coefficients.

Combining the XUV-IR RABBITT [1] interferometric technique at the 10kHz Attolab-FAB10 beamline, with electron-ion coincidence momentum spectroscopy, we investigate PADs for atomic (Ar) and molecular (NO, O_2) targets.
In this scheme, an attosecond XUV pulse train ionizes the target and a superimposed IR field with angular frequency ω and delay τ induces continuum-continuum (cc) transitions, generating sidebands (SB) in the photoelectron (PE) spectra. Analysing the 2ω modulation of the SB signal as a function of τ paved the way to studies of PI dynamics in real time, leading to the determination of Wigner time delays τ_W of the PE wavepacket generated by one-XUV-photon absorption [2]. This requires deciphering, within the atomic time delay τ_A, the τ_{cc} contribution due to the cc transition [3]. Advanced developments address angularly resolved RABBITT studies, with the goal to retrieve Wigner delays as a function of the ejection direction [4-9].
This experiment gives access to the PAD $I_{SB}(\theta,\tau)$, where θ is the emission angle relative to the light polarization **P**, and to the MFPAD $I_{SB}(\theta_e,\phi_e,\chi,\tau)$ for a molecule oriented at any χ angle relative to **P,** where (θ_e,ϕ_e) defines the emission direction in the molecular frame (MF). In the RABBITT spectrogram:

$$I_{SB}(\theta,\tau) = A(\theta) + B(\theta)\cos\left(2\omega\tau + C(\theta)\right)$$

the atomic PE delays $\tau_A(\theta)$ relative to $\tau_A(0°)$ are extracted from the $C(\theta)$ phase. Here we display an example of measured $A(\theta)$ (static) and $B(\theta)$ (dynamic) amplitudes for PI of Ar. A unified formalism synthetizing $I_{SB}(\theta,\tau)$ in terms of a reduced set of coefficients, which allows for generating e.g. the $A(\theta)$, $B(\theta)$ and $C(\theta)$ functions, will be presented. First MFPADs, obtained taking advantage of dissociative PI, will also be reported for PI of NO and compared with MCSCI calculations.

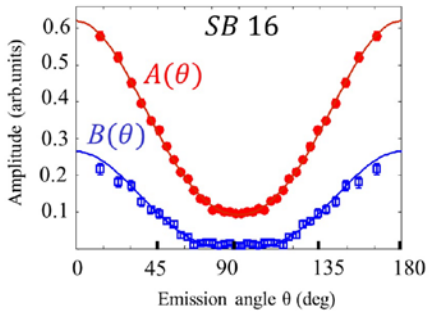

Figure 1 $A(\theta)$ and $B(\theta)$ for SB16 in XUV+IR PI of Ar.

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