

optical polarimeters including a dephazer element, the MP method is the only *in situ* approach providing the full determination of the (s_1 , s_2 , s_3) Stokes vector, equivalent to that of (ψ , ε , P), the orientation and signed ellipticity of the polarization ellipse, and the degree of polarization P . In particular, for partially polarized light, the MP method enables us to extract the light helicity s_3 separately from the degree of unpolarized light $s_4 = 1 - P$ (sometimes considered as a fourth Stokes parameter¹). One clear advantage of the method¹¹ is its validity across the whole VUV to X-ray range that offers a broad tunability for applications.

The remarkable development of advanced light sources delivering ultrafast XUV pulses has created unprecedented possibilities to address and possibly visualize the primary steps of photochemical dynamics in time-resolved studies of ultrafast processes at the natural time-scale of electron dynamics and fast nuclear motion, ranging from attoseconds to a few femtoseconds. Among these ultrafast sources free electron lasers (FELs) stand out, as well as secondary XUV sources based on high-order harmonic generation (HHG), resulting from the non-linear interaction between an intense infrared (IR) radiation and a gas medium.¹⁹ Ultrashort coherent XUV pulses may serve two types of application schemes: (i) an “external probing” scheme in time-resolved pump-probe PI experiments with controllable delay at the subfemtosecond scale,^{20–27} and (ii) a “self-probing” scheme in the case of HHG, where the three-step generation process (strong field tunnel ionization, laser driven electronic wave-packet motion, and recollision of the returning electron leading to photorecombination)^{28,29} probes the system on the time scale of half a laser cycle, a technique now referred to as high harmonic spectroscopy (HHS).^{30–35} In both approaches the molecular frame (MF) observables are a unifying concept for characterizing photochemical dynamics *via* snapshot mapping of the electronic density, either in photoemission or by molecular orbital tomography with femto to attosecond temporal resolution. For both approaches too, the occurrence of specific situations where symmetry breaking in the generation process results in the production of elliptically polarized coherent sub-femtosecond pulses has been emphasized in recent experiments, *e.g.*, HHG emission from aligned molecules^{36,37} or induced by an elliptical IR laser,³⁸ or HHG driven by a two-color field involving photons of opposite helicity.³⁹ This context motivates the development of methods for a complete characterization of the harmonic polarization state providing (i) sources with optimized ellipticities for time-resolved investigations of dichroisms, and (ii) access to the complex induced dipole vector and insight into the interactions occurring during the HHG process, in particular the dynamics of the electron–parent ion recombination.

In the work presented here, we report MFPAD studies based on electron–ion coincident 3D momentum spectroscopy in the context of attosecond molecular dynamics conducted at SLIC facility of CEA-Saclay, with the two perspectives addressed above: on the one hand, using the generated APT as a light source for PI of target systems characterized at the MFPAD level,^{40–42} and on the other hand, taking advantage of MFPAD-resolved PI of simple molecules, to perform a polarization analysis of the HHG emission in high harmonic spectroscopy.

(i) We demonstrate the measurement of complete MFPADs in dissociative photoionization of a chosen molecular target by an attosecond pulse train: this constitutes the first step towards the study of subfemtosecond time-resolved electron/nuclear dynamics in molecules where the reaction launched by the APT