

is either photoionization, or relaxation of a neutral excited state probed by photoionization, in different pump-probe schemes.⁴³

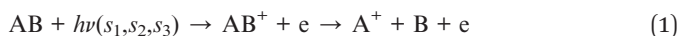
(ii) We use molecular polarimetry to determine the complete polarization state of harmonics generated in SF₆ molecules by an elliptically polarized IR field. SF₆ is a polyatomic molecule where it was found that strong field ionization involves contributions from multiple electronic channels, while the recombination step is influenced by resonant transitions due to the presence of autoionizing states and shape resonances⁴⁴ trapping the recolliding electron for typically hundreds of attoseconds before recombination to the ground state occurs. Our MP results are compared with data from optical polarimetry (OP) in the conditions of the MP study.

The paper is organized as follows. In order to ensure a self-contained content, we revisit in Section 2 the general expression of the MFPAD for PI of linear molecules induced by elliptically polarized light, and the subsequent key steps for the extraction of (i) the dynamical parameters of the PI reaction and (ii) the Stokes parameters, providing the grounds for MP. Section 3 describes the experimental methodology, where dissociative photoionization of small molecules induced by an attosecond pulse train is analyzed using 3D-momentum spectroscopy. In Section 4, we report the first complete measurements of MFPADs in PI from an incident APT. In Section 5, we apply the MP method to the complete characterization of the polarization state of the HH comb produced in SF₆ gas by an elliptically polarized driving laser. Conclusions and perspectives are given in Section 6.

2. MFPADs induced by elliptically polarized light: methodology for molecular polarimetry

The molecular polarimetry method, previously described,¹¹ relies on the remarkable properties of molecular frame photoelectron angular distributions (MFPADs) induced by elliptically polarized light^{45,46} as obtained in the study of dissociative photoionization (DPI) for primarily linear molecules using electron-ion coincident 3D momentum spectroscopy.

Briefly, for DPI of a molecule AB producing an ion fragment A⁺ and a photoelectron, MFPADs are determined from the correlated ion fragment recoil velocity V_{A^+} and photoelectron velocity V_e measured for each DPI coincident event (A⁺, e).^{47,48}



The extension to inner-shell ionization where PI is followed by Auger decay is straightforward.⁴⁹ For one-photon PI of a linear molecule, the general expression of the MFPAD in the dipole approximation is a function of three angles, $I(\theta_e, \phi_e, \chi)$, for linear⁵⁰ and circular⁴⁵ polarizations of the ionizing light, while it is function of an additional angle, $I(\theta_e, \phi_e, \chi, \gamma)$, in the general case where PI is induced by elliptically polarized light.^{11,46} (θ_e, ϕ_e) represent the polar and azimuthal electron emission direction in the molecular frame (MF) and (χ, γ) the polar and azimuthal ion fragment emission direction in the field frame (FF) or laboratory frame (LF), as indicated schematically in Fig. 1(a). In the LF, the z_{LF} reference axis for the (χ) polar dependence is parallel to the k