large error bars. The case $\varepsilon_{fun} \approx -0.17$ does not show this trend, which could possibly be due to an imprecision in the determination of s_1 and s_2, in particular for H17. A higher repetition rate in future experiments will allow increasing the statistics and thus reducing these error bars in order to reach a final conclusion on the presence of depolarization in this case.

These polarization characteristics provide insight into the complex interactions which occur during the HHG process in the SF_6 molecule. It was proposed[44] that the first step, *i.e.* 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 resonances trapping the recolliding electron for typically hundreds of attoseconds before recombination to the ground state occurs. The specific behavior observed for H15 and H17 was related to the presence of the $5t_{1u} \rightarrow \varepsilon t_{2g}$ shape resonance in the A channel that dominates the harmonic emission in this spectral range, as shown by complex Kohn computations of valence PI total cross sections involving strong intercoupling effects.[72] The influence of resonances was then investigated using 2D-TDSE simulations for Ar atoms (see ESI of ref. 38), and it was found that they result in a structuring of the returning electron wavepacket and an increase of the perpendicular component of the dipole in the radiative recombination step of the HHG process. The high ε_{ub} values (assimilated to $\langle\varepsilon\rangle$) measured in SF_6 were then attributed to such an effect induced by the $5t_{1u} \rightarrow \varepsilon t_{2g}$ shape resonance. The reported MP measurements prove that the harmonic emission is indeed highly elliptical, which strengthens the proposed interpretation in terms of resonance. Furthermore, some degree of depolarization is expected in such a case, because the polarization characteristics should be highly dependent on the detuning from the resonance, inducing spectral variations inside the harmonic bandwidth.[38] This calls for more precise MP measurements using a high repetition rate laser. With the direct determination of the signed ellipticity and of the degree of polarization, these results complement those obtained previously for SF_6 and demonstrate the potential of the MP method to access the complete state of polarization of the HHs composing an APT. The interpretation of the striking fingerprints of the ε_{fun} and harmonic order dependent polarization state of the HHs composing the APT generated on SF_6 molecules, in particular the large ellipticities and strong variation of the ellipse orientation in the H13–H17 range, calls for additional theoretical studies.

6. Conclusion

In the work presented we have reported MFPAD studies based on coincident electron-ion 3D momentum imaging in the context of ultrafast molecular dynamics investigated at the SLIC facility of CEA-Saclay, with two perspectives, namely (i) using APT generated in atoms/molecules as a source for PI studies performed at the level of MFPADs, and (ii) taking advantage of MFPAD-resolved PI of simple molecules to perform a polarization analysis of the HHG emission in molecules, a major challenge of high harmonic spectroscopy.

The ability to measure $I(\theta_e,\phi_e,\chi)$ complete MFPADs induced by an APT opens perspectives for two-color pump-probe time-resolved MFPAD studies at the attosecond time scale, with the goal to probe the evolution of *e.g.*, the electronic density in a transient excited molecular state through photoionization. Entangled