

second, the significant discrimination of processes leading to more energetic N^+ fragments at the extraction field chosen for the HHG experiment to ensure a 4π collection of process (I) (eqn (5)). Furthermore, scrutiny of the one photon energy KECDs ensures that a unique selection of reaction (eqn (5)) induced by the APT is achieved when the $0.25 \text{ eV} \leq E_{N^+} \leq 0.55 \text{ eV}$ ion fragment selection is performed. As a result, in the photoelectron spectrum displayed in Fig. 4(c) corresponding to this selection, the resolved peaks reflect PI of NO into the $NO^+(c^3\Pi)$ ionic state for each of the HHs composing the APT: it features directly the HHG spectrum for harmonics H15 and higher, convoluted by the total photoionization cross section corresponding to eqn (5). Experimental and/or theoretical PI cross sections can be used as reference to restore the original HHG spectrum.^{63,64}

Fig. 5 displays $I(\theta_e, \phi_e, \chi)$ complete MFPADs, subsequent to the interaction of an APT with the NO molecular target, derived from the measured $I(\theta_e, \phi_e, \chi, \gamma)$ angular distribution for selected processes in the KECD, after integration over the (γ) angle. Here the APT was generated in SF_6 gas by an elliptically polarized IR laser $\varepsilon_{\text{fun}} = 0.2$, the elliptical polarization of the HHs being unknown at this level. Selecting as an example the peak assigned to PI by the H17 harmonic, at photon energy $h\nu = 26.35 \text{ eV}$, the complete MFPAD is determined by the extraction of the five $F_{LN}(\theta_e)$ functions implemented in (eqn (2)). The four $F_{00}(\theta_e)$, $F_{20}(\theta_e)$, $F_{21}(\theta_e)$, $F_{22}(\theta_e)$ are displayed in Fig. 5(a) providing the $I(\theta_e, \phi_e)$ MFPAD for any orientation of the molecular axis relative to the axis of linearly polarized light, shown in Fig. 5(b) for three meaningful orientations $\chi = 0^\circ$, 90° featuring the parallel and perpendicular transitions, and $\chi = 45^\circ$, where a coherent superposition between parallel and perpendicular orientations occurs. In Fig. 5(a) the $F_{LN}(\theta_e)$ functions are compared with those measured at SOLEIL at photon energy $h\nu = 26.35 \text{ eV}$. Despite the rather low statistics in the HHG measurements at the 1 kHz laser rep-rate, reflected by the statistical error bars (e.g. here 3000 events in the selection), the comparison shows that the main characteristics of the MFPADs are fairly well determined. This is a striking demonstration of the potential of the F_{LN} -based data analysis methodology, since all events collected for a given process are used to generate the MFPAD for each selected orientation of the molecule. The 3D plots of the $I(\theta_e, \phi_e)$ MFPADs for the selected orientations illustrate remarkably well the partial wave expansion different for the parallel and perpendicular transitions resulting from the linear combinations of $F_{00}(\theta_e)$ and $F_{20}(\theta_e)$, and including the azimuthal dependence described by the $F_{21}(\theta_e)$ and $F_{22}(\theta_e)$ as soon as the cylindrical symmetry is broken ($\chi \neq 0^\circ$). Expanding the F_{LN} functions in Legendre polynomials enables us to extract the complex dipole matrix elements for the PI transition. Extraction of the $F_{11}(\theta_e)$ function multiplied by the s_3 Stokes parameter, completing the dipole matrix elements, is conveniently presented in the next section where the MP method is applied.

As an illustration of the PI dynamics in this region, Fig. 6 displays the evolution of the parallel transition with the HH order, scanning the photon excitation energy across the σ^* shape resonance of the $NO^+(c^3\Pi)$ ionization channel,^{45,64} very well predicted by multichannel Schwinger configuration interaction method (MCSCI) when the effects of the dynamic electronic correlation, *i.e.*, the correlated motion of the photoelectron and electrons of the target are included.^{45,50} The dominance of the f partial wave and the π increase of the phases of the matrix elements can therefore be discussed in this context where significant inter-channel coupling occurs. At the maximum of the shape resonance, around 30 eV,