derived from the analysis of the $I(\chi,\gamma)$ histograms for each harmonic according to the method illustrated in Fig. 1.

However one additional step has to be included in the data analysis, which accounts for the modification of the polarization state of the HHGs which is likely to be introduced by optical elements, in particular the toroidal gold mirror which focuses the XUV pulses at the center of the COLTRIMS spectrometer. As mentioned in the Experimental section, the action of this mirror has been characterized using the MP method, providing s_1, s_2, s_3 Stokes parameters for a series of known orientations of the incident linearly polarized XUV field (*i.e.*, that of the IR laser), and modeled with a designed Mueller matrix which includes transmission parameters for the four relevant harmonics. The other optical element crossed by the XUV pulses is the metallic Al filter which has a flat response in the region of interest and should not influence the polarization state. In the data reported in the following, the Mueller formalism was used to determine the complete polarization state for each harmonic in terms of the normalized Stokes vector upstream from the mirror, based on that directly measured after transmission by the mirror.

Fig. 7 illustrates the extraction of the s_3 Stokes parameters for the four harmonics downstream from the mirror: the MFPADs display significant right–left asymmetries which demonstrate that large positive s_3 values are obtained, in particular for H15 and H17.

The large values determined for the s_3 Stokes parameter, in particular $s_3 \approx 0.8 \pm 0.05$ for H15 and H17, demonstrate unambiguously the large degree of polarization and the quasi-circular character of the polarization state for these harmonics, corresponding to a positive ellipticity $\varepsilon \approx 0.70 \pm 0.06$. For H19 and H21 the sign remains positive while the magnitude of the ellipticity decreases.

In Fig. 8 we present the results obtained with the MP method for two ε_{fun} values of opposite sign close to $\varepsilon_{\text{fun}} \approx \pm 0.2$ (which we would rather attribute to

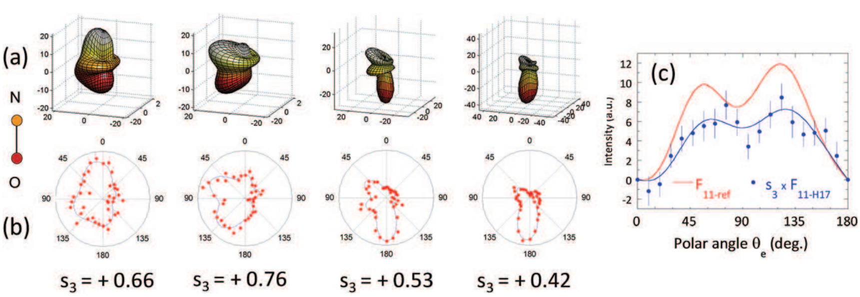

Fig. 7 (a) $I(\theta_e,\phi_e)$ MFPADs for the ($\chi = 90°$) orientation of the molecular axis relative to the propagation axis of the APT generated by $\varepsilon_{\text{fun}} \approx 0.23$ of the driving IR laser, for the H15 to H21 harmonics: the significant right-left emission anisotropies reflect the HH dependent s_3 Stokes parameter and F_{11} function (b) cuts of the MFPADs in the polarization plane ($\phi_e = 90°$ or 270°) displaying the largest CDAD anisotropy (c) $s_3 \times F_{11}(\theta_e)$ for H17 (blue dots and Legendre polynomial fit line) compared to the reference F_{11} function at $h\nu = 26.35$ eV (red line): the $s_3 = 0.76 \pm 0.05$ value is obtained as the ratio between the blue curve and the reference F_{11} function (red); the measured values downstream from the mirror are shown at the bottom of the figure, with ±0.05 error bars, while the values upstream from the mirror amount to $s_3 = 0.78$, 0.77, 0.51, and 0.22 (±0.05), respectively, using the Mueller formalism (see text).