are then refocused in the ion-electron coincidence 3D momentum spectrometer CIEL[52] using a 11.5° grazing incidence angle, 22 × 6 cm size 60 cm focal length toroidal gold mirror. Special care has been taken to characterize the transmission of the polarized harmonics by the toroidal mirror: its action has been calibrated using the MP method, providing s_1, s_2, s_3 Stokes parameters for a series of known linear polarizations of the incident harmonics, that is probing the complex reflectivity for the s and p components for the four studied harmonic energies using Mueller matrix formalism, as briefly discussed in the data analysis of the measured Stokes parameters.

A motorized gold mirror can be inserted upstream from the toroidal mirror. It intersects the light beam at 45° and directs the beam toward a second gold 45°-incidence mirror and an XUV spectrometer (composed of variable groove spacing grating, micro-channel plates and phosphor screen detector). This allows direct HHG monitoring in order to optimize efficiency and beam steering, but also to perform optical polarimetry, *i.e.*, Malus' law type polarization analysis using the two 45° reflections as an analyzer. Indeed, the difference of reflectivity between the s and p polarization components amounts to a factor 20 in the 15$^{\rm th}$ to 25$^{\rm th}$ harmonic spectral range (23.25–38.75 eV). Instead of turning the analyzer, we use the property that HHG is a field-driven process, the polarization of which in an isotropic target gas is determined by the driving laser. We thus rotate continuously the laser polarization ellipse by an angle θ using a half-wave-plate inserted before the focusing lens, and record the HHG yield $I_q(\theta)$ for the different harmonic orders q on the spectrometer. For a perfect analyzer, this would give the following dependence, also called Malus' law:

$$I_q(\theta) = \frac{1}{2}[S_0 + S_1 \cos(2\theta) + S_2 \sin(2\theta)] \tag{6}$$

The fit of the HHG yield variation with a Malus' law taking into account the imperfect analyzer provides the values of the two normalized Stokes parameters: $s_1 = S_1/S_0$ and $s_2 = S_2/S_0$, from which one obtains the direction of the harmonic ellipse (ψ), and an upper bound for the magnitude of the ellipticity $\varepsilon_{\rm ub}$ (see Section 2). Indeed, this incomplete OP cannot disentangle the circular part of the polarization (measured by s_3) from the unpolarized part. Performing a complete OP would require measuring in addition the variation of the HHG yield $I_q(\theta)$ in the presence of a dephasing element, such as an XUV quarter-wave plate which is not yet available, or using the dephasing induced by reflection on a metallic mirror, that is accompanied with a strong signal attenuation. In most of the published work except for a few studies[53,54] the harmonic emission was considered to be fully polarized, so it is assumed that $\varepsilon_{\rm ub} = |\varepsilon|$.

The XUV light beam directed into the CIEL 3D momentum spectrometer induces PI of the gas target, here mainly NO molecular and He atomic targets, produced by a two skimmer supersonic expansion. The supersonic jet and ultrahigh vacuum chambers originate from a previous version of the COLTRIMS type apparatus[55,56] which combines electric and magnetic fields to guide ions and electrons. The advanced version of the spectrometer based on the two delay line time and position sensitive detectors (DLD PSDs RoentDek), including an electrostatic focusing lens for the ion trajectories,[48] ensures a 4π collection of both particles for the studied DPI processes. An eight-channel time-to-digital converter