

is used to encode the DLD time signals, providing the position for each particle of the ( $A^+$ ,  $e$ ) coincident events as well as the ion time of flight (TOF), while the electron TOF is encoded using a synchronized time to amplitude converter.

One issue for the present measurements was the 1 kHz repetition rate of the laser, which restricts to about 50 c/s the overall coincidence count rate. This imposed a few hours of stability for each measurement on the NO target. The subsequent statistics for these measurements is discussed in the following sections. This situation has motivated complementary measurements relying on PI of the He target which are not discussed here.

## 4. MFPADs for PI induced by an attosecond pulse train (APT)

In this section we report the first  $I(\theta_e, \phi_e, \chi)$  complete MFPADs subsequent to the interaction of an APT with a molecular target, based on the ( $V_{A^+}$ ,  $V_e$ ) vector correlation analysis of dissociative photoionization processes. Building on the experience in femtochemistry,<sup>57</sup> two strategies have prevailed so far in the few studies of MFPADs in PI of simple molecular targets by attosecond pulses. The first one relies on the impulsive alignment of small molecules such as  $O_2$ ,  $N_2$ ,  $CO$ ,  $CO_2$  using a near-IR laser and taking advantage of the periodical revival of the rotational wave packet to ionize the molecules at the maximum of field-free alignment or anti-alignment with a properly delayed APT.<sup>40</sup> The energy and  $I(\theta_e)$  angular distributions of the photoelectrons were obtained after processing an Abel inversion of the images recorded by Velocity Map Imaging (VMI) and could be assigned to the parallel and perpendicular transitions for the different ionic channels and photon energies in the harmonic comb. Secondly, despite HHG sources usually operate at a rather low repetition rate (1–10 kHz), they allow 3D momentum electron–ion coincidence spectroscopy: first complete measurements of  $I(\theta_e, \phi_e, \chi)$  MFPADs were demonstrated in dissociative photoionization of  $H_2$  and  $D_2$  induced by spectrally filtered single high harmonic, following resonant excitation of doubly excited auto-ionizing states,<sup>41</sup> and traced as a function of the kinetic energy release of the atomic fragments. This prototypical reaction features a dynamical interplay between the electronic and nuclear motions at the 1–5 femtosecond time scale, giving rise to quantum interferences between distinct reaction pathways and remarkable symmetry breakings, and it stands as a model process for state-of-the-art calculations.<sup>58,59</sup> A related study, based on a similar electron-ion coincidence technique and using a broad band APT, focused on the dependence of photoelectron emission asymmetries in the MF which also illustrates nicely those symmetry breakings.<sup>42</sup>

The present study is illustrated by DPI of NO molecules as motivated earlier, and more specifically on the prototype reaction (eqn (5)). We use for the ionizing radiation the APT generated by  $SF_6$  molecules driven by elliptically polarized IR laser, the polarization state of which will be characterized using the MP technique in Section 5. Inner-valence ionization of molecules usually involves several DPI processes corresponding to PI into distinct molecular ionic states and dissociation channels, each “reaction pathway” being assigned by the intermediate ionic state and the populated dissociation channel. The first step in the analysis of the ( $A^+$ ,  $e$ ) DPI coincident events consists of disentangling the reaction pathways,