based on the resolving power of the electron-ion kinetic energy correlation (KEC) featured in KEC diagrams (KECDs),[47,60] and relying on the total energy conservation ($h\nu - E_D = E_e$ + KER), where E_D is the asymptotic potential energy at the dissociation limit, E_e and KER are the photoelectron energy and the kinetic energy release of the two heavy atomic/molecular fragments, respectively. Typical KECDs are displayed in Fig. 4(a) and (b) for one-photon PI corresponding to DPI of the NO molecule induced by (a) a one-photon-energy pulse at SOLEIL, here $h\nu = 23.65$ eV as discussed above, and (b) an APT produced by HHG from the SF_6 gas. In Fig. 4(a) three DPI processes are identified, which correspond (I) to PI into the first $NO^+(c^3\Pi,(4\sigma)^{-1})$ ionic state followed by dissociation into the $N^+(^3P) + O(^3P)$ + e limit and has the largest intensity, and (II) and (III) to PI into the $NO^+(B^1\Pi,(4\sigma)^{-1})$ and $NO^+(B'^1\Sigma^+,(1\pi)^{-1})$ ionic states dissociating into the $N^+(^3P) + O(^3P)$ + e limit (II) and the two close lying ones $N^+(^1D) + O(^3P)$ + e and/or $N^+(^3P) + O(^1D)$ + e (III).[47]

The KECD presented in Fig. 4(b) for DPI induced by the APT shows well resolved peaks corresponding to the same $E_{N^+} \approx 0.4$ eV ion fragment energy and different E_e electron energies, separated by 3.1 eV, *i.e.*, the energy difference between two successive odd harmonics in the APT. These structures are assigned to the dominant DPI process (eqn (5)) produced by the harmonics comb, with energies larger than the ionization potential of NO into the $NO^+(c^3\Pi,v = 0)$ ionic state $I_P = 21.7$ eV, namely from harmonic H15 up to H25 in the present experiment. Such a KECD reflects the superposition of as many one-photon-energy KECDs as there are contributing HHs in the APT. To ensure a valid interpretation of the KECD assigned to the APT, as well as to provide reference data for the MF circular dichroism at the H15–H23 photon energies, we have performed a series of complementary experiments on the DESIRS beamline at SOLEIL synchrotron using circularly polarized light at the same energies.[61,62] These results establish that there is no significant overlap between the resolved structures assigned to DPI into the $NO^+(c^3\Pi)$ ionic state, and other DPI processes that might result in comparable (E_{N^+}, E_e) positions in the KECD, so a proper selection of the processes enable us to proceed to the MFPAD analysis for each harmonic. The reason why weaker processes (II) and (III) resolved in Fig. 4(a), or the other DPI processes induced at the higher photon energies, do not provide any significant contribution in Fig. 4(b) is, first, the intrinsically weak probability of process (III) and,

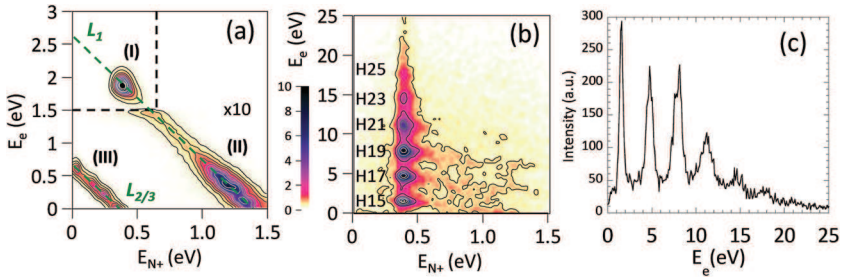

Fig. 4 KECDs characterizing DPI of NO induced by (a) synchrotron radiation at $h\nu = 23.65$ eV (b) an APT generated on the SF_6 gas medium; (c) photoelectron spectrum reflecting the HHG spectrum (H15 to H25) convoluted with the PI cross section for eqn (5) (4π collection of electrons and ions) extracted from the KECD shown in (b) after the 0.25 eV $\leq E_{N^+} \leq$ 0.55 eV ion-fragment energy selection.