expected to favor ionization from orbitals that lie close enough to the HOMO, typically within an energy range smaller than the ionizing photon energy.[45]

This trend is confirmed by decreasing the photon energy from 3.1 eV to 1.55 eV (Fig. 6(c) and (h)). A single frequency comb appears in the ARPES and PECD, corresponding to ionization from the HOMO. The PECD remains quite strong, in the $\pm$5% range even for the highest detected peak associated to the absorption of 10 photons. When the laser intensity increases, the ARPES becomes more sharply peaked along 90°, *i.e.* around the laser polarization plane, and the PECD decreases. This decrease is the signature of a stronger influence of the laser field on the outgoing scattering electron compared to the chiral potential.

The pure tunnel-ionization regime is reached by further increasing the laser wavelength to 1850 nm (Fig. 6(e) and (j)). The ARPES is very sharp, and a significant PECD is measured over the whole energy range, up to energies corresponding to the absorption of more than 30 photons. No contribution from the inner orbitals appears, since their ionization probability decays exponentially with the ionization potential. This measurement demonstrates that PECD can be observed even far away from the ionization threshold. This regime is usually described by the strong-field approximation, which consists in neglecting the influence of the ionic potential on the electron dynamics after tunneling. Our results show the failure of this approximation and reveal the influence of the electron scattering in the chiral molecular potential, as recently predicted by Dreissigacker and Lein.[46] We have recently shown that this could be understood within a purely classical picture of ionization through classical trajectory Monte-Carlo simulations.[43]

2.4 Conclusions

Photoelectron circular dichroism can be measured in all ionization regimes, from the single-photon to tunnel ionization, using ultrashort light pulses. The number of open channels depends on the ionization regime, offering complementary pictures of the chiral molecules. All energetically permitted channels are open in XUV one-photon ionization, and the direct transition to the continuum enables the probing of the vibrational state of the ion.[11] In the perspective of dynamical measurements, for instance in the case of a vibrational wavepacket in the ground electronic state, the high number of ionization channels can be considered as an advantage, since it would enable measuring the response of different molecular orbitals to the nuclear dynamics. On the other hand the channel identification in the ARPES and PECD can be complex. Coincidence electron-ion detection is a solution to this issue, enabling (partial) separation of the different channels by their different dissociative ionization pathways. In comparison, REMPI has the advantage to strongly favour resonant channels, even though several resonant channels can co-exist as we have seen in fenchone. The drawback is the complexity of the interpretation: the resonant character of the ionization may be modulated by the ongoing molecular dynamics, introducing an additional parameter to the sensitivity of the experiment. As the driving wavelength and intensity increase towards the tunneling regime, the number of open channels decreases, reflecting the exponential decay of tunnel ionization with increasing ionization potential. In the tunneling regime a weak PECD remains despite the 10^8 V cm^{-1} electric field. This PECD purely originates from the HOMO. This