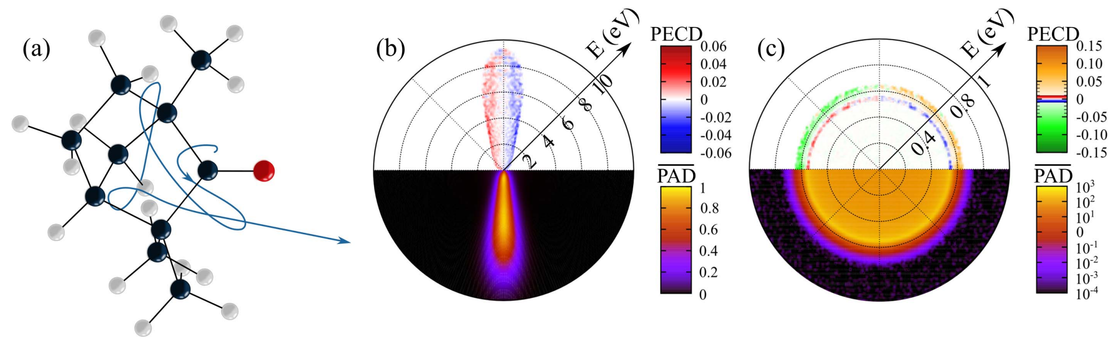

Figure 4. Typical ionizing CTMC electron trajectory in fenchone embedded in a circularly polarized laser pulse with $\lambda = 800$ nm and $I = 10^{14}$ W cm^{-2} (a). $\overline{\mathrm{PAD}}$ (bottom half) and PECD (top half) from CTMC calculations in the tunneling (b) and single-photon (c) ionization regimes. The light propagation axis is horizontal and the radius extends up to 10 eV in (b), and up to 1 eV in (c).

reaches the energy $\hbar\omega$-IP. The subsequent dynamics of the freed electron is calculated over three laser cycles, during which the electric field amplitude linearly decreases down to 0. The $\overline{\mathrm{PAD}}$ and PECD associated to absorption of 9.3 eV photons are presented in figure 4(c). A ring structure is obtained for the $\overline{\mathrm{PAD}}$, as expected. The PECD is weak, of the order of $\sim$1% at the maximum of the PES. However it reaches very significant and opposite values, of the order of $\pm$15%, in the tail of the PES. This PECD shape shows that electrons with different kinetic energies have experienced significantly different scattering on the chiral potential. Similar features are observed in quantum mechanical descriptions [4, 29].

4. Conclusion

To sum up we have demonstrated that PECD is a universal effect in the photoionization of fenchone molecules by circularly polarized radiation. Molecular chirality is clearly encoded in the PADs whatever the ionization regime, ranging from one-photon to tunnel ionization. To what extent can these results be generalized to other chiral systems? One-photon VUV PECD has been measured in a broad range of chiral molecules. Its value varies a lot from one system to another, and depending on the photon energy. Nevertheless, PECD can be considered as a general effect in XUV photoionization. On the other hand, laser-based PECD was up to now only measured in a REMPI scheme. This sets quite a limitation on the range of systems that can be studied, and makes data interpretation challenging for example when multiple resonances are hit by the excitation process. Nevertheless, REMPI-PECD is considered as a promising technique and a great metrology tool, enabling for instance accurate determination of enantiomeric excess [30, 31]. In this work, we have bridged the gap between the generality of one-photon XUV PECD and the versatility of femtosecond laser sources by demonstrating that PECD could be observed in non-resonant multiphoton/tunnel ionization. The fact that PECD still exists in the strong-field regime is indeed not so intuitive, because this regime is often successfully described using the strong-field approximation, in which the influence of the chiral molecular potential is neglected. Our CTMC calculations have revealed that even in a strong laser field, the chiral potential was able to significantly affect the ionizing electrons, imprinting a forward/backward asymmetry. We have shown that this effect was qualitatively similar to the one at the origin of one-photon PECD. Thus, strong-field PECD should be a general effect, as XUV PECD is. Its magnitude will of course depend on the molecules, and it may be very weak in some specific cases.

The variety of ionization regimes in which PECD can be measured offers interesting perspectives to obtain complementary pictures of the chiral response. Tuning the wavelength and intensity of the driving laser in the MPI and ATI ranges enables probing different electronic ionizing states, from one-electron (Koopmans) to two-hole-one-particle configurations. Vibrationally resolved PECD is also accessible in the MPI regime. All of this opens the route to more sophisticated multi-color ionization experiments, in which the control of different laser pulses will enable coherent control of the electron asymmetry, or to reach chiral information on the attosecond dynamics underlying molecular photoionization [32].

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