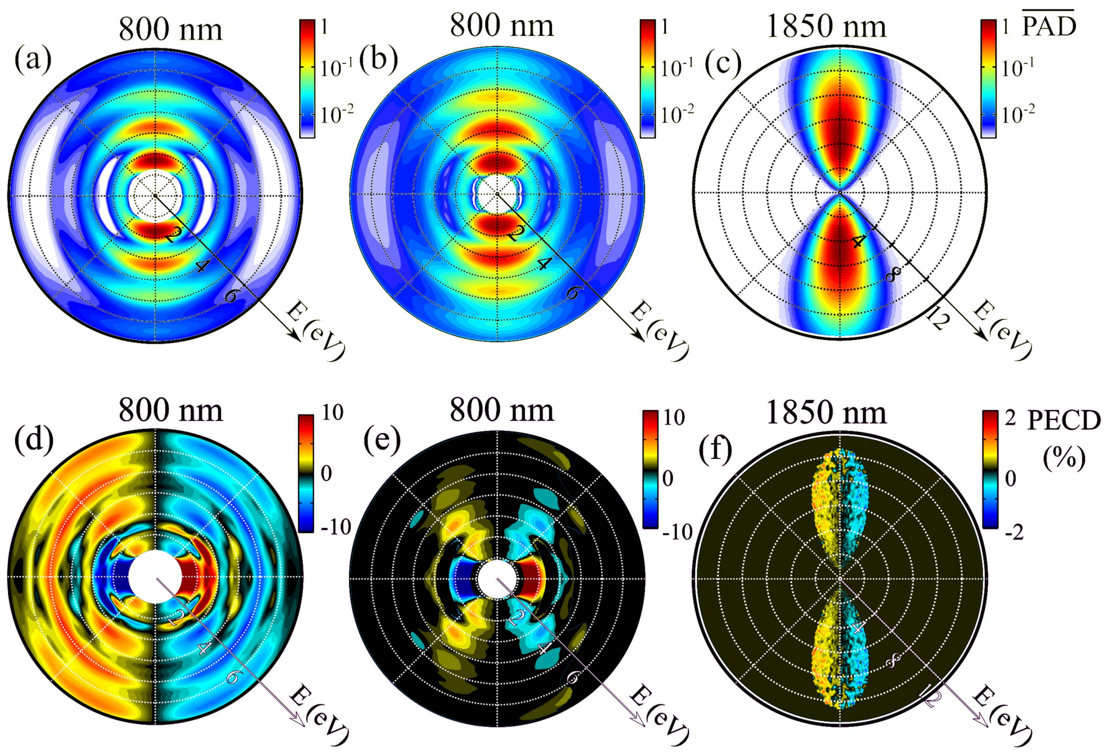

Figure 3. Photoionization of (+)-fenchone from ATI to the tunneling regime. $\overline{\mathrm{PAD}}$ and PECD for $\lambda = 800$ nm pulses with $I \sim 9 \times 10^{12}$ (a), (d) and $I \sim 1.2 \times 10^{13}$ (b), (e) W cm^{-2}. (c)–(f) Raw projection of the $\overline{\mathrm{PAD}}$ and PECD for $\lambda = 1850$ nm pulses with $I \sim 4 \times 10^{13}$ W cm^{-2}. The light propagation axis is horizontal and the radius extends from 0 to 7 eV in (a), (b), (d), (e) and 0 to 12 eV in (c), (f).

photons from the HOMO. There is no contribution of inner orbitals to the signal because of the exponentially decaying rate of tunnel-ionization with increasing IP. A clear PECD is observed, in the 1%–2% range. This demonstrates that PECD still persists in the tunneling regime, even high above the ionization threshold.

3. A classical outlook on PECD

In order to understand how the chiral potential succeeds in imprinting asymmetry in such a high intensity regime, we performed Classical Trajectory Monte Carlo (CTMC, [26, 27]) calculations. We focused on electron dynamics from the HOMO of fenchone, in the fixed-nuclei approximation. We used an approximate point-charges description of the ionic potential, where effective charges located on the nuclei of fenchone are set so as to reproduce the quantum mechanical potential issued from Hartree–Fock calculations [28]. For a given molecular orientation, the final $(E,\ \theta)$-distribution of freed electrons is simply defined by counting among the $\mathcal{N} = 10^6$ independent electron trajectories those with positive energy at the end of the interaction. A typical ionizing trajectory is displayed in figure 4(a). We then mimic random alignment of the experimental gas samples by repeating the CTMC calculations for an ensemble of molecular orientations defined in terms of Euler angles with regular spacing $\Delta\alpha = \Delta\beta = \Delta\gamma = \pi/8$ rad.

Figure 4(b) illustrates the $\overline{\mathrm{PAD}}$ and PECD obtained at the end of a half-cycle pulse with $\lambda = 800$ nm and $I = 10^{14}$ W cm^{-2}. Such irradiation conditions correspond to tunnel-ionization for the HOMO, with a Keldysh parameter of $\gamma \approx 0.85$. The classical simulations are in very good qualitative agreement with the experimental results of figures 3(c) and (f). In the commonly accepted picture of tunneling, the electron is freed at a distance $r \sim \mathrm{IP}/F_0$ from the center of the target, where F_0 is the maximum strength of the pulse electric field. Using Hartree–Fock results for IP, this yields $r \sim 7$ a.u. The ionic potential still presents small chiral anisotropy beyond such distances, which can thus induces small PECD. However our CTMC calculations reveal that the electron trajectories are submitted to significant multiple scattering on chiral nuclear structure before leaving the target, as examplified in figure 4(a). This is the main source to significant PECD within CTMC.

PECD from one-photon ionization can also be described classically, in the framework of the sudden approximation. Photon absorption is assumed to give the electron a kick resulting in instantaneous modification of its momentum at fixed position. Absorption occurs randomly within one laser cycle and the magnitude of the kick, in opposite direction to the driving field, is determined so that the electron suddenly