



Fig. 8 Time-resolved PECD measurements in fenchone in a $1 + 1'$ configuration (201 nm linearly polarized pump and 404 nm circularly polarized probe). The top polar plots show the APECD recorded at three pump probe delays: 0.5 ps (a), 2 ps (b) and 6 ps (c). The light propagation direction is horizontal and the radius extends from 0 to 0.7 eV. The lower contour plots depict the evolution of the Legendre coefficients b_0 (d), b_1 (e) and b_3 (f) as a function of electron kinetic energy and pump probe delay.

Enantiopure fenchone molecules were injected by a pulsed Even-Lavie valve operating at a 1 kHz rate and backed with 12 bars of He. For the probe, we generated 3.08 eV (404 nm) pulses with 35 meV bandwidth by second harmonic generation, and sent them in a zero order quarter waveplate to convert their polarization state to circular. The pulses were focused into the VMIS at $\sim 1 \times 10^{11} \text{ W cm}^{-2}$ with an independent lens. The temporal resolution of the experiment was limited by the dispersion of the focusing optics and vacuum window, leading to a 170 fs cross-correlation time between the pump and probe. For each pump-probe delay, the probe laser helicity was switched every 45 000 laser shots to record a series of 8 left and 8 right images. The pump-probe delay was then scanned consecutively ten times to smooth out slow drifts in the experiment. Background images corresponding to the probe pulse alone were recorded for each helicity and subtracted from each image.

Fig. 8 shows the evolution of the PES and APECD (eqn (4)) as a function of pump-probe delay. The PES is centered around 0.52 eV, which corresponds to a $1 + 1'$ transition in which all the excess energy goes to the electron ($\text{KE} = E_{\text{pump}} + E_{\text{probe}} - I_{\text{p}} = 0.53 \text{ eV}$, where $I_{\text{p}} = 8.72 \text{ eV}$ is the experimental vertical ionization