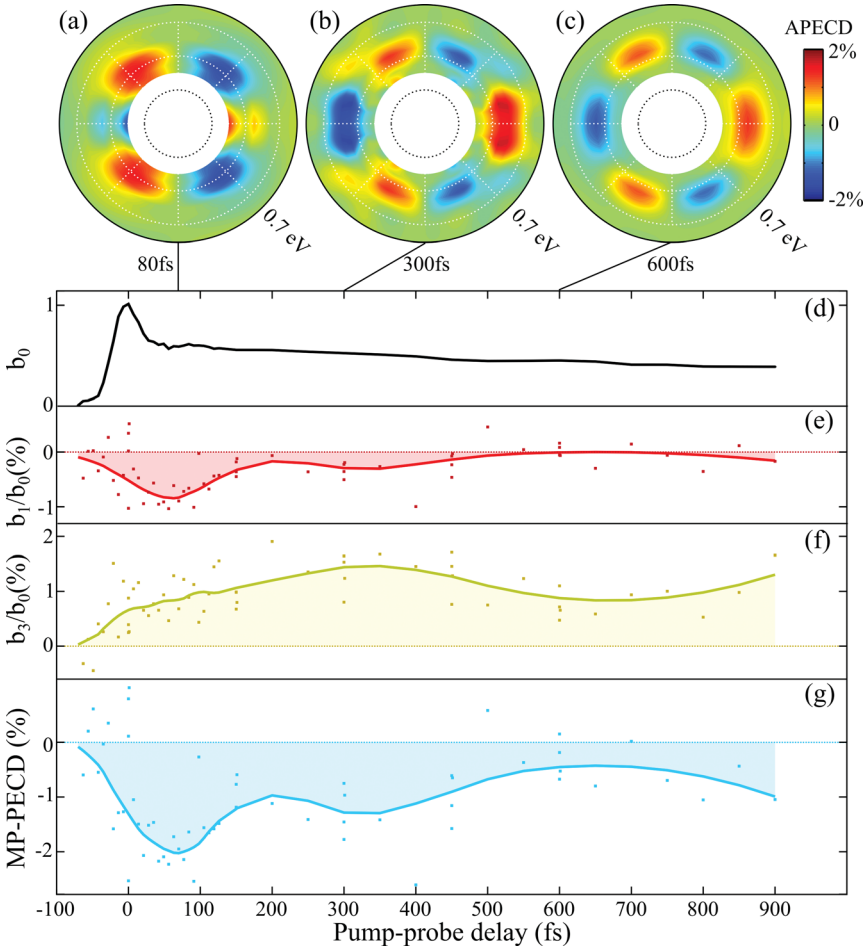


Fig. 10 Time-resolved PECD measurements in fenchone in a 2 + 2′ configuration (400 nm linearly polarized pump and 800 nm circularly polarized probe). (a–c) APECD averaged around three pump–probe delays: 80 ± 20, 300 ± 50 and 600 ± 50 fs. The light propagation direction is horizontal and the radius extends from 0 to 0.7 eV. The lower plots depict the temporal evolution of the PES $b_0$ (d), normalized Legendre coefficients $b_1/b_0$ (e), $b_3/b_0$ (f), and MPPECD (g).

### 3.3 Two-photon excitation and two-photon ionization

For this last TRPECD experiment, we used linearly polarized 400 nm pulses with 9 nm bandwidth to photoexcite fenchone molecules by resonant two-photon absorption, and ionized with circularly polarized 800 nm pulses. The cross correlation time was below 50 fs in this experiment. The fenchone source here was the continuous nozzle described in the static PECD measurements. The overall PECD is much weaker than in the 1 + 1′ and 1 + 2′ cases, reaching a maximum value of 2%. The angle-resolved APECD show a very fast evolution (Fig. 10(a)–(c)). This evolution can be quantified by monitoring the Legendre coefficients averaged over the width of the PES peak, as a function of pump–probe delay. The PES