Supporting information for Pulse Length Dependence of Photoelectron Circular Dichroism

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1 Additional figures related to fenchone PECD

The effective fitting results for LPECD in Fig. 3 (c1–c4) in the main text were obtained by dividing Gaussian fitting results for PES×LPECD by Gaussian fitting results for PES. The Gaussian fitting results for PES×LPECD, which were not shown in the main text, are provided in Fig. S1.

We also provide the raw data of c_1 and c_2, the coefficients for the two lowest order Legendre polynomials, in Fig. S2. c_1 and c_2 are the dominant contributions in our fenchone results. The higher-order terms have bad signal-to-noise ratios and are not presented.

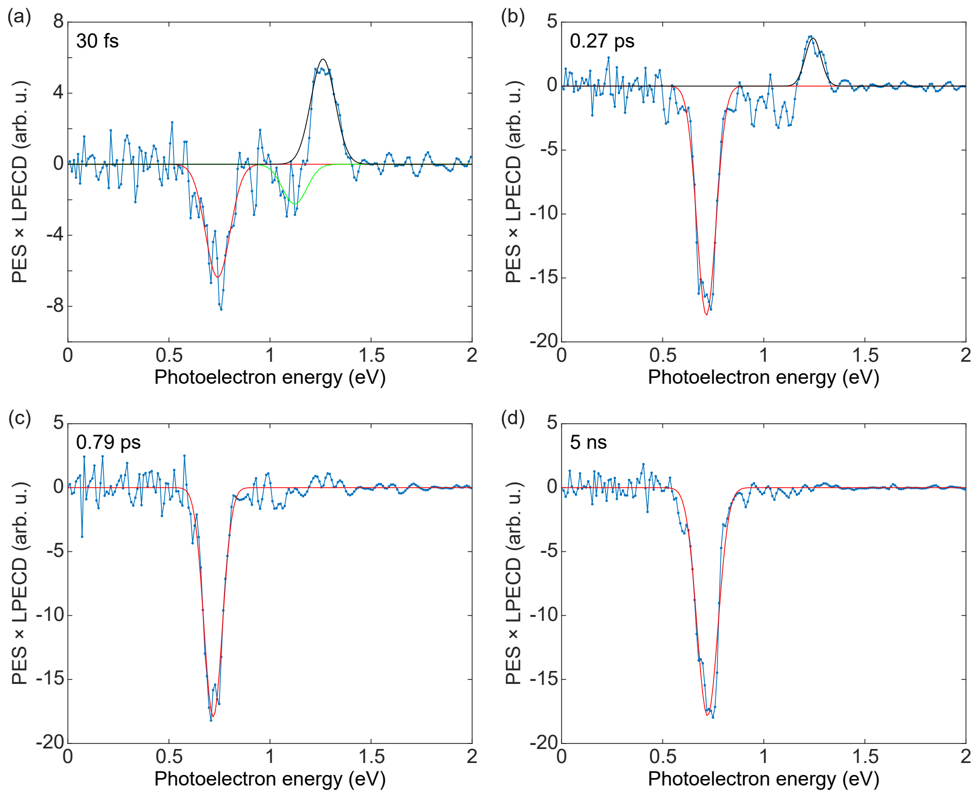

Figure S1: Gaussian fitting results for PES×LPECD corresponding to (a) 30 fs, (b) 0.27 ps, (3) 0.79 ps, and (d) 5 ns measurement with (*S*)-(+)-fenchone. We used (a) three Gaussians (b) two Gaussians, and (c, d) a single Gaussian for the fitting, depending on the shape and the strength of the 3p contribution. In each plot, the experimental result is shown as blue dots connected by blue solid lines and fitted Gaussians are shown as red, black and green solid lines.

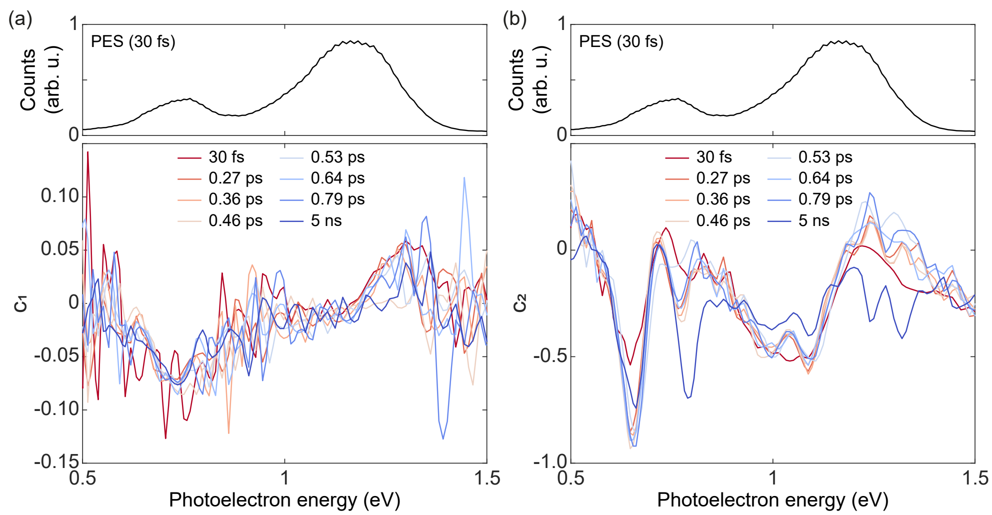

Figure S2: (a) c_1 and (b) c_2, the coefficients for Legendre polynomials of degree 1 and 2, for various pulse durations. The measurements were done with (S)-(+)-fenchone and the results are given as functions of the photoelectron energy. In the upper part of each plot, the PES from the 30 fs measurement is presented to show the positions of the 3s and the 3p contributions. c_1 and c_2 for different pulse durations seem to follow each other.

2 PES and lifetime estimation data

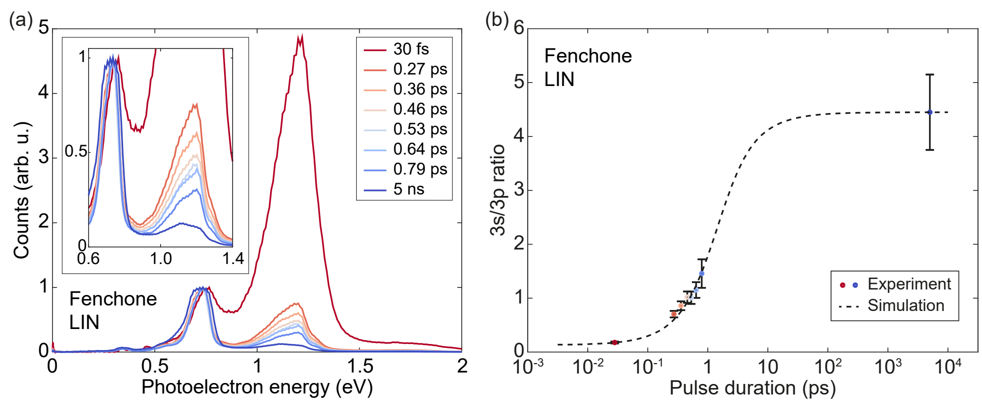

Figure S3: (a) Photoelectron energy spectra of fenchone, measured by using linearly polarized pulses with various pulse durations. The measurement was done with (S)-(+)-fenchone. The difference in the shape of the PES for the linear and circular polarization (see Fig. 2 (b) in the main text) is attributed to the polarization dependence of the absorption cross section and the internal conversion lifetimes. (b) Experimental and simulation results of 3s/3p ion population ratios of fenchone as functions of the pulse duration. The measurement was done with (S)-(+)-fenchone and linearly polarized pulses. Circles indicate the experimental results, and the dotted line shows the simulation result. The same color code as in (a) was employed to distinguish the data points for different pulse durations. The simulation was done in the same way as described in the model calculation section of the main text. This specific simulation result was obtained with an assumption of $\left[\mu^{(2)}_{\mathrm{3s,g}}/\mu^{(2)}_{\mathrm{3p,g}}\right]^2 = 0.2$

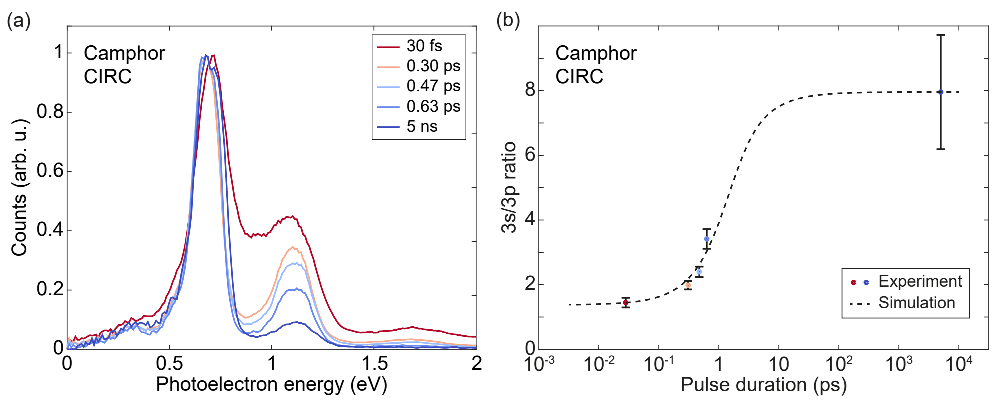

Figure S4: (a) Photoelectron energy spectra of camphor, measured by using circularly polarized pulses with various pulse durations. The measurement was done with (S)-(−)-camphor. (b) Experimental and simulation results of 3s/3p ion population ratios of camphor as functions of the pulse duration. The measurement was done with (S)-(−)-camphor and circularly polarized pulses. The details of the figure are the same as Fig. S3. This specific simulation result was obtained with an assumption of $\left[\mu^{(2)}_{3s,g}/\mu^{(2)}_{3p,g}\right]^2 = 2.0$

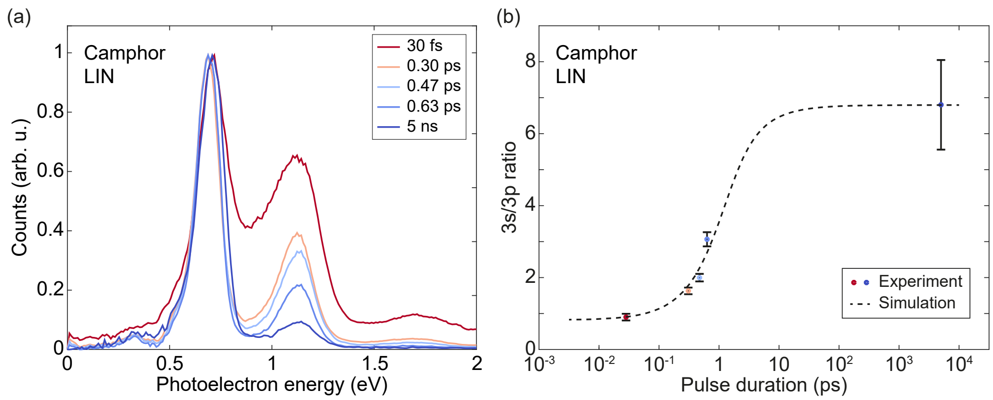

Figure S5: (a) Photoelectron energy spectra of camphor, measured by using linearly polarized pulses with various pulse durations. The measurement was done with (S)-(−)-camphor. (b) Experimental and simulation results of 3s/3p ion population ratios of camphor as functions of the pulse duration. The measurement was done with (S)-(−)-camphor and linearly polarized pulses. The details of the figure are the same as Fig. S3. This specific simulation result was obtained with an assumption of $\left[\mu^{(2)}_{3s,g}/\mu^{(2)}_{3p,g}\right]^2 = 0.30$