

Supplementary Material:

Tracking electron motion within and outside of Floquet bands from attosecond pulse trains in time-resolved ARPES

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1. Additional Tr-ARPES spectra from polychromatic case probed along k_x

We provide here additional results complimentary to those presented in the main text in Figs. 8,9, and 10, but where instead of probing the ARPES spectra along the k_y axis, it is probed along k_x . The two sets of data together can help form a more complete picture of the charge dynamics driven in the system. The resulting spectra are presented in supplementary figures S1, S2, and S3, for varying probe arrangements. In general, the spectra present very similar physical phenomena to those discussed in the main text, verifying the generality of the conclusions.

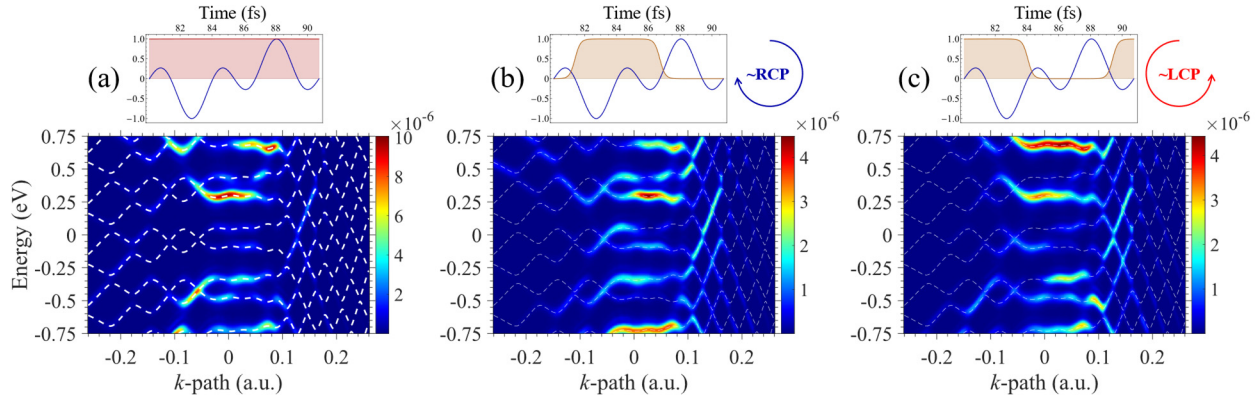


Figure. S1. Tr-ARPES spectra with poly-chromatic $\omega-2\omega-3\omega$ pump field (same as in Fig. 8 in main text, 3200nm, $I_0=5\times 10^{10}$ W/cm²) but probed parallel to k_x . (a-c) Same as in Fig. 8 in main text.

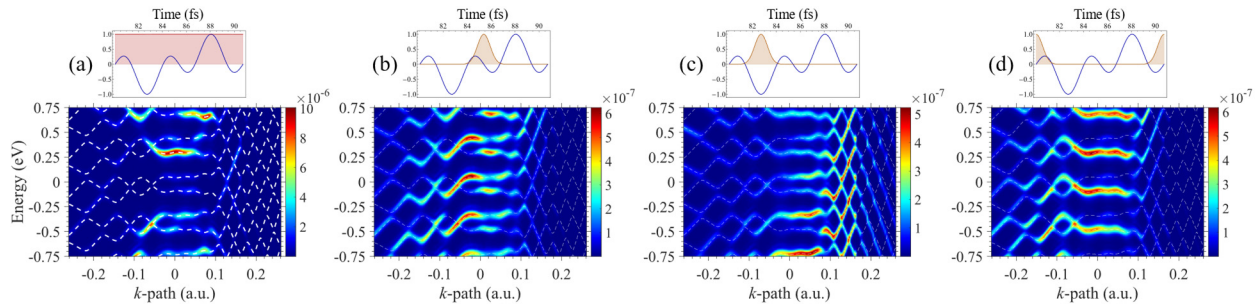


Figure. S2. Tr-ARPES spectra with a polychromatic $\omega-2\omega-3\omega$ pump field (as in Fig. 9 in the main text, eq. (7) in the main text, 3200nm, $I_0=5\times 10^{10}$ W/cm²) but probed parallel to k_x . (a-d) Same as in Fig. 9 in main text.

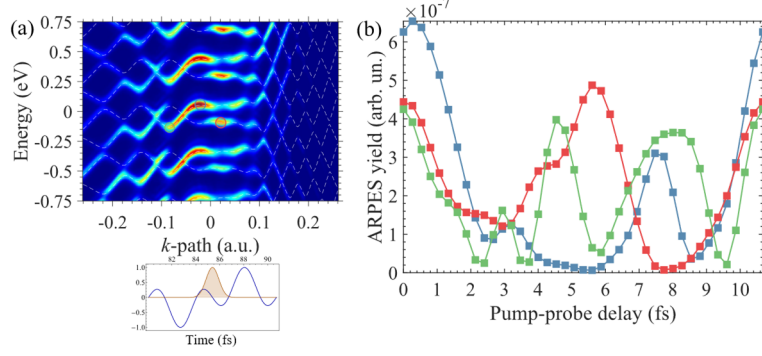


Figure. S3. Sub-laser-cycle charge dynamics in Tr-ARPES spectra a polychromatic $\omega-2\omega-3\omega$ pump field (as in Fig. 10 in the main text, eq. (7) in main text, 3200nm , $I_{\text{p}}=5\times 10^{10}\text{ W/cm}^2$) but probed parallel to k_x . (a-b) Same as in Fig. 9 in main text.

2. Tr-ARPES spectra for changing probe duration

We present here the full Tr-ARPES spectra corresponding to Fig. 6(c) in the main text, which explores the formation of non-Floquet bands and the strength of their ARPES signal with respect to the probe duration. Figure S4 shows the Tr-ARPES spectra for a fixed pump-probe delay, but for various probe pulse durations towards the single attosecond pulse regime. We emphasize here the regime where the Floquet bands ‘dissolve’ and are replaced by the corresponding instantaneous Houston bands when the probe pulse no longer overlaps with multiple laser cycles (whereby the Floquet phase is essentially not sampled by the probe). For further discussion see main text.

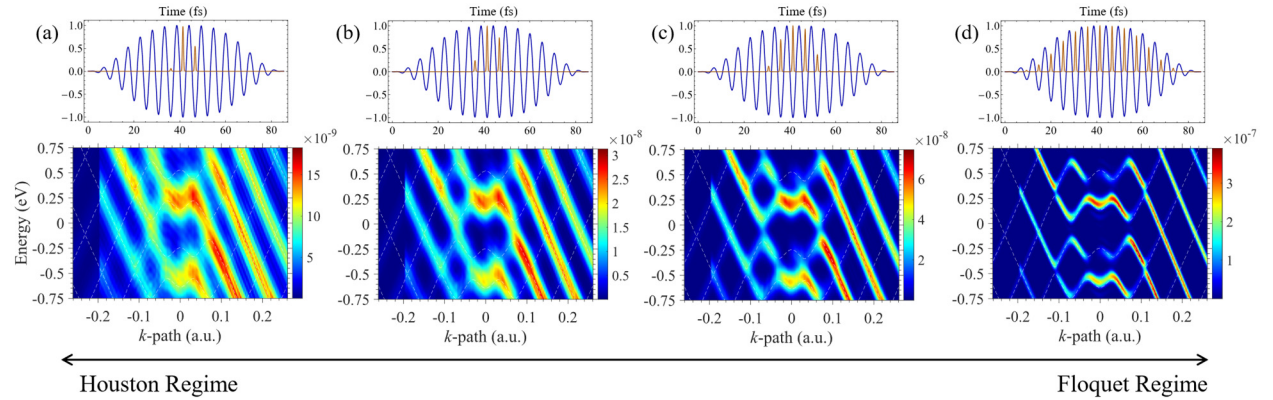


Figure. S4. Comparison of sub-laser-cycle resolved ARPES spectra with a circularly polarized quasi-monochromatic pulse probed with an attosecond pulse train (corresponding to Fig. 6(c) in the main text), but for a fixed pump-probe delay and while varying the probe pulse duration. The top row in each plot shows the relationship between employed pump and probe pulses. The figure corresponds to conditions in Fig. 6(c) in the main text for a few selected envelope durations.