Attosecond Currents in Solids

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Achieving solid-state coherent electronics requires controlling light-induced carrier generation and motion on the one-femtosecond-scale to beat incoherence via equilibration. Because the durations of carrier and photon wavepackets are dictated by their spectral bandwidth, observing sub-femtosecond dynamics requires exploiting extreme ultraviolet laser pulses or nonlinear optical effects.

Carriers created *via* multi-photon absorption or strong-field injection are spectrally broad, and their energy spread in the conduction band is hard to predict. We avoid these complicated initial states by implementing extreme ultraviolet-inject and visible-drive photoconductive sampling (Fig. 1a) [1]: First, we single-photon-excite carriers from the valence to the conduction band of lithium fluoride (LiF, $E_{Gap} = 13.6$ eV) using a ∼1-fs long extreme ultraviolet light pulse generated via high-harmonic generation. Then, using the electric field of a delayed, carrier-envelope-stable, visible laser pulse, we drive the crystal momentum of the photocarriers and generate a macroscopic current that we detect using electrodes on the sample surface (Fig. 1b). At low drive intensities, we find that the current is linear to vector potential of the driving laser field at the instant of carrier injection.

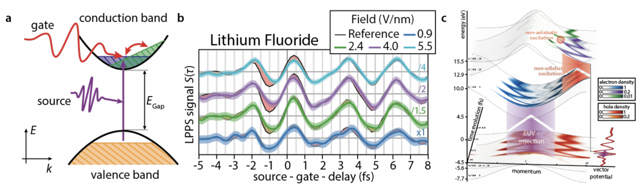

Figure 1: a) Experiment: an extreme-ultraviolet laser pulse excites carriers from the valence to the conduction bands of LiF. The electric field of a visible few-cycle laser pulse modulates their momentum. This accelerates the carriers and creates a macroscopic current. b) the measured current versus the delay between the extreme ultraviolet and the visible light pulses at different driving field strengths. c) semiconductor-Bloch-equation-modeled carrier dynamics in LiF. From [1]

At increased driving field, the delay-dependent current deviates from the vector potential (Fig. 1b). We identify two contributions to the deviations (Fig. 1c): at the maxima of the observed current, the driving laser field can move carriers into non-parabolic regions of the band structure. This effect crops such maxima locally and is fully reversible after the maximum. When further increasing the driving field, the carriers are pushed near the Brillouin zone edge, where the distance to the second conduction band decreases. Here, the drive laser can non-adiabatically excite them to the second conduction band, a non-reversible transition that inverts the group velocity of the involved carriers. Because these transitions only occur after carriers have been propelled to the Brillouin zone edge, they modify the current signal before the driving laser pulse maximum and can be recognized by their timing.

This observation highlights that populating multiple adjacent conduction bands diminishes light-driven currents, allowing us to formulate a Fourier-type upper limit for the speed of efficient optoelectronics: carriers in an optoelectronic device made from a material possessing a conduction band with bandwidth ΔE_{CB} can most efficiently generate light wave-driven currents if the duration of the carrier wavepacket exceeds $\Delta\tau_{FWHM}$: $\Delta\tau_{FWHM}\frac{\Delta E_{CB}}{2} > 3.66$eV fs.

References

[1] M Ossiander, K Golyari, K Scharl, L Lehnert, F Siegrist, J P Bürger, D Zimin, J A Gessner, M Weidman, I Floss, V Smejkal, S Donsa, C Lemell, F Libisch, N Karpowicz, J Burgdörfer, F Krausz and M Schultze, Nat. Commun. **13**, 1620 (2022)