Contribution ID: **27** Type: **Invited talk**

Field-driven virtual charge dynamics in dielectrics

Friday, June 27, 2025 11:00 AM (30 minutes)

The interaction of intense, few-femtosecond infrared (IR) pulses with solids can induce light-field-driven phenomena, reversibly modifying their electro-optical properties on attosecond time scales. This opens new avenues for ultrafast optoelectronics and petahertz device applications. However, harnessing these coherent light–matter states requires a deeper understanding of the fundamental processes governing strong-field interactions in solids. In particular, the interplay between real and virtual carrier dynamics remains largely unexplored, despite its potential to constrain material response times, especially in wide-bandgap dielectrics.

We employ attosecond transient reflection spectroscopy to probe ultrafast virtual electron dynamics in monocrystalline diamond across a previously unexplored photon energy range (20-50 eV). Absolute pump-probe delay calibration enables a direct, parameter-free comparison with time-dependent density functional theory (TDDFT) simulations, revealing that virtual inter-band transitions (VITs) —often neglected in favour of intraband motion —play a crucial role in shaping both the timing and adiabaticity of the ultrafast nonlinear response. Since VITs emerge instantaneously as the light field dresses the material, they influence the available bandwidth and response time well before real vertical transitions occur. Further analysis using an independent-particle approximation and a simplified three-band model elucidates the origin and impact of VITs, paving the way for their exploitation in next-generation optoelectronic devices and enabling precise control of material properties on sub-femtosecond time scales.

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Session Classification: Session 16 - Materials III