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Attosecond dynamics in polycrystalline diamond

The possibility to manipulate the electrical properties of matter with very short optical pulses is a fascinating field of research with possible far reaching applications in many relevant technological fields. The first step towards the realization of this goal is to understand the ultrafast dynamics at the basis of light-matter interaction. Short and intense pulses allowed us to investigate a very interesting regime where the photon energy becomes comparable to the cycle-averaged kinetic energy of the electrons in the field. As the optical response of the material transitions from a classical to quantum-mechanical description many intriguing effects co-exist in this regime and the importance of inter- versus intra-band transitions is still debated. We used attosecond transient absorption spectroscopy (ATAS) to study the optical response of polycrystalline diamond driven by few-femtosecond, intense (IIR ~10^12 W/cm^2) infrared (IR) pulses. We monitored the system response by looking at the induced change in the absorbance with a 250-as pulse centred around 40 eV. We observed the appearance of oscillating features which modulate at twice the IR frequency, ωIR, and fully recover after the interaction. Simultaneous photoelectron acquisition from a gas nozzle placed in front of the diamond target allowed us to study the phase relation of the oscillating features and the pumping IR field. We found that the timing of the diamond response changes significantly with the probing energy and does not always follow the IR field adiabatically. Ab initio calculations performed by coupling time-dependent density functional theory (TDDFT) in real time with Maxwell's equations reproduced the experimental observations. Further comparison with a numerical two-band model allowed us to conclude that intra-band motion dominates over inter-band transitions, thus identifying the dynamical Franz-Keldysh effect as the dominant mechanism in this regime. Our analysis constitutes an important step towards a full understanding of the optical properties of dielectrics in the Petahertz regime.