Photoionization-Induced Harmonics in Solids: Amplitudes, Phases, and Mechanism Identification

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The formation of an electron–hole plasma during the interaction of intense femtosecond laser pulses with transparent solids lies at the heart of femtosecond laser processing. Advanced micro- and nanomachining applications require improved control over the excitation characteristics. We relate the emission of low-order harmonics to the strong laser-field-induced plasma formation. Together with a measurement of the total plasma density, we identify the contribution of two competing ionization mechanisms: strong-field and electron-impact ionization.

These mechanisms lead to the generation of high-order harmonics in bulk solids subjected to intense ultrashort laser pulses, which has opened up new avenues for research in extreme nonlinear optics and light–matter interaction on subcycle time scales. Despite significant advancement over the past decade, a complete understanding of the involved phenomena is still lacking. High-harmonic generation in solids is currently understood as arising from nonlinear intraband currents, interband recollision, and ionization-related phenomena. As all of these mechanisms involve or rely upon laser-driven excitation, we combine measurements of the angular dependence of nonlinear absorption and high-order harmonic generation in bulk crystals to demonstrate the relation between high-harmonic emission and nonlinear, laser-induced ionization in solids. An unambiguous correlation between the emission of harmonics and laser-induced ionization is found experimentally, which is supported by numerical solutions of the semiconductor Bloch equations and an analytical model of orientation-dependent ionization rates using maximally localized Wannier functions.

As a further characterization approach, we employed extreme ultraviolet (XUV) high-harmonic interferometry with phase-locked XUV pulse pairs to investigate excitation-induced bandgap dynamics in solids. Our experiments on amorphous fused silica and crystalline MgO, complemented by analytical modeling and semiconductor Bloch equation simulations, reveal a correlation between transient bandgap modifications and variations in the phase of harmonic emission. These findings suggest a potential pathway for sub-cycle, all-optical control of band structure modifications, advancing prospects for petahertz-scale electronic applications and attosecond diagnostics of carrier dynamics.