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Dynamics of electrons and holes measured by high-harmonic spectroscopy

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The dynamics of molecular valence-shell electrons offer a rich variety of phenomena ranging from simple two-level quantum beats to complex relaxation phenomena [1]. These dynamics are both of fundamental interest and relevant for understanding the properties and chemical reactivity of molecules. Molecular valence-shell dynamics take place on femto- to attosecond time scales and therefore require novel techniques that offer both an extreme temporal resolution and a high sensitivity. This lecture will present two novel schemes based on high-harmonic spectroscopy (HHS) that we have developed for measuring valence-shell electron dynamics of molecules. The first scheme [2] gives access to electronic dynamics in neutral molecules with unprecedented sensitivity: a 0.1% excitation fraction results in a 20% modulation of the high-harmonic signal, directly tracing the temporal evolution of the electronic density. This scheme exploits coherent cross-channels in high-harmonic generation that result from ionization and recombination in distinct quantum states. The intensity of the emission from these channels directly probes the electronic coherence and its evolution under coupling to nuclear degrees of freedom. In the second scheme we apply HHS as a unified pump-probe experiment to follow attosecond charge migration in a molecule following strong-field ionization. We impulsively orient the molecules using a recently developed all-optical technique [3] and measure both the amplitude and the phase of high-harmonic emission using multiple wavelengths and intensities of the driving laser pulse. We further introduce a new theoretical framework that accounts for correlation-driven electron-hole dynamics in high-harmonic spectroscopy. The comparison between theory and experiment reveals the signatures of the attosecond relaxation of the electronic valence shell following ionization.

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[2] P. M. Kraus, S. B. Zhang, A. Gijsbertsen, R. R. Lucchese, N. Rohringer, and H. J. Wörner, PRL 111, 243005 (2013).
[3] P. M. Kraus, A. Rupenyan and H. J. Wörner, PRL 109, 233903 (2012)