Investigations on Ultrafast Atomic and Molecular Dynamics with Harmonic Sources

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Abstracts: *High harmonics have been applied for probing ultrafast atomic processes of He and also for revealing multi-orbital structure of CO_2 molecules.*

High harmonic light sources with attosecond duration in the EUV and soft x-ray spectral range can be applied to probe ultrafast dynamics of atoms and molecules. By tuning harmonic wavelength a specific state of an atom or molecule can be excited, which can be further excited or ionized by applying a time-delayed femtosecond laser pulse. This process of combining harmonic pulses and time-delayed femtosecond laser pulses was applied to investigate the dynamics of photoexcitation and photoionization processes of He. In addition high-harmonic radiation generated from molecules contains the information on the structure of molecules. When multiple molecular orbitals are exposed to a strong laser field, the highest-occupied molecular orbital (HOMO) is mostly ionized and thus emits strong high-harmonic radiation containing the characteristics of HOMO. In order to resolve multiple orbitals of CO_2 molecules we employed two-dimensional high-harmonic spectroscopy (HHS) by applying an orthogonally polarized two-color laser field consisting of the fundamental frequency and its second harmonic. In this case odd and even harmonics carried the characteristics of the HOMO and HOMO-1, respectively. The multi-orbital characteristics were thus revealed in the two-dimensional spectroscopy employing the two-color laser field.