Development Of A Soft X-Ray Spectroscopy Beamline Based On Hhg For Studying Ultrafast Dynamics In Advanced Materials, With A Focus On Perovskite-Based Systems

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With conventional semiconductors facing a shortage, the need for alternative resources is evident. One potential candidate is 3D perovskites whose interesting properties – e.g., low exciton binding energy [1], defect tolerance [2], large polarons [3] - constitutes them perfect for light harvesting devices [4].

Condensed matter phenomena that may hinder their performance, like polaron formation, are observed at femtosecond timescales. Ultrafast soft X-ray spectroscopy allows studying this light-matter interaction with an unprecedented temporal and spatial resolution, with the further advantage of being element-selective, oxidation, and spin-state specific. As such, we plan to investigate large polaron formation dynamics in 3D perovskites with time-resolved Near Edge X-ray Absorption Fine Structure (NEXAFS). Pump-probe spectroscopy is one of the most common experimental schemes for these types of measurements. In summary, a first IR pump pulse excites the sample, then the evolution is probed by a second time-delayed XUV pulse. Hence, innovative experimental set-ups that focus on broadband detection, high dynamic range, and sensitivity have become necessary in ultrafast dynamics.

High-order harmonic generation of femtosecond laser pulses interacting with noble gases has become a popular table-top source of broadband coherent XUV radiation throughout the years. By implementing a microfluidic device instead of the trusted gas-jet configuration, we obtained an extended energy cut-off, and a higher generation yield [5, 6]. Furthermore, an integrated solution for filtering IR radiation under vacuum has been investigated. High vacuum systems were also realized to manipulate, detect, and characterize the generated XUV light.

However, all-optical techniques have huge untapped potential, given that conventional spectrometers are characterized by limited spectral ranges (either low or high-energy photons), and a lack of polarization characterization which excludes the studies of magnetic materials and biologically relevant molecules. Therefore, an innovative detection scheme was realized in our laboratory to surmount these limitations

References

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