Quantum Optics Meets Strong Field Physics: New Regimes of Coherent X-ray Generation with Strong Electron Correlation Dynamics and Attosecond Rabi Oscillations

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Abstract: Ultrafast imaging and spectroscopies using coherent EUV and X-ray light based on the extreme nonlinear process of high harmonic generation are already addressing grand challenges in complex molecular systems, advanced nanomaterials, and plasmas. The exquisite quantum control of the attosecond dynamics of the rescattering electrons in this upconversion process makes it possible to sculpt the classical and quantum properties of the light with unprecedented tunability of the spectral, spatial, temporal shape, and spin and orbital angular momentum state. The full spatial and temporal coherence of this unique light allows for multi-dimensional imaging at the spatio-temporal extreme with 4D resolution of nanometers and femtoseconds, including access to an effective 5th dimension – the periodic table of elements - due the X-ray absorption fingerprinting with elemental and chemical specificity.

In this talk, I will present two novel regimes of coherent X-ray generation using short wavelength UV and EUV driving lasers where the design of the light properties is dominated by the quantum dynamics of the entangled electrons in a simple He atomic system. Interestingly, the physics of these regimes extend beyond the three-step high harmonic model.

In the first regime, using intense UV lasers, the entangled electron dynamics in a He atom result in a secondary high harmonic plateau, extending well beyond the conventional EUV cutoff, up to the water window in the soft X-ray region. This is due to simultaneous double electron recombination where a single high-energy X-ray photon is emitted only in systems with strongly correlated electrons. This physical process paves a way to a sensitive attosecond-to-femtosecond spectroscopy as a probe of highly correlated interactions in atomic and molecular systems. Similar physics of EUV high harmonics from solids might be able to reveal correlated dynamics in phase transition materials and nanosystems of relevance to quantum computing and superconductivity.

In the second extreme regime, using shorter-wavelength EUV driving lasers operating at a resonance of He can result in very bright high harmonic generation in the X-ray regime. Here, macroscopic nonlinear optics favores phase and group velocity matching since the driving laser and the generated wavelengths have similar dispersion. Furthermore, on a single atom level, the emission is strongly enhanced, first, due to minimized quantum diffusion of the rescattering electrons when using EUV lasers. Second, fast attosecond Rabi oscillations lead to unique suppression of the ionization depletion of the ground state of the system, which otherwise would result in termination of the emission. Theoretically, this regime is scalable to produce bright coherent light at the technologically relevant EUV and soft X-ray wavelengths of interest for semiconductor metrology with near-wavelength resolution. These new advances of quantum control over the light properties enable new insights into complex entangled electron dynamics and new applications in nanomaterials and quantum technologies.

1. T. Popmintchev, et al., Bright coherent ultrahigh harmonics in the keV x-ray regime from mid-infrared femtosecond lasers, Science 336 (6086), 1287-1291 (2012)
2. D. Popmintchev, et. al., Ultraviolet surprise: Efficient soft x-ray high-harmonic generation in multiply ionized plasmas, Science 350 (6265), 1225-1231 (2015)
3. D. Popmintchev, at. al., Coherent EUV - Soft X-ray Light with Continuous Red and Blue Wavelength Tunability, CLEO: Fundamental Science, FW4M.4 (2023)
4. S. Wang, et. al., CLEO Europe EQEC: Postdeadline, PD-1.5 (2023)
5. A. de las Heras, et. al., Attosecond Rabi oscillations in bright high harmonic emission in the X-ray regime driven by XUV pulses, Frontiers in Optics, FTu1C.3 (2022)