Project title: Coherent Diffractive Imaging of Quantum Dynamics in Isolated Nanosystems

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Current state of the art

With the advent of high-brightness short-wavelength radiation sources such as x-ray free electron lasers or intense lab-based XUV sources using high harmonic generation, single-shot coherent diffractive imaging (CDI) has developed into a key tool to decipher the structure and dynamics of individual particles in free flight. In the common application scenario, the diffracted light is detected as a single-shot image and then reconstructed via phase retrieval or advanced forward-fitting approaches. This methods has offered the analysis of clusters and nanoparticles [1, 2], aerosols [3], and viruses in free flight [4] as well as the identification of so far inaccessible quantum vortices in nanoscale quantum fluids such as Helium nanodroplets [5].

A general assumption underlying the conventional reconstruction approaches is that the diffraction image reflects the light-matter interaction in linear response or even in first Born approximation. The latter approximation is fully justified in the hard x-ray regime, where the diffraction image represents simply the Fourier transform of the projected density of the target [6]. However, the single scattering approximation underlying the first Born approximation becomes questionable already in the XUV domain, as substantial departures of the material's refractive index from unity require at least full account of the light propagation in linear response including substantial absorption and multiple scattering events. On the other hand, XUV CDI experiments offer access to wide-angle scattering and therefore enable the reconstruction of three-dimensional structural features from a single diffraction images, which opens so far inaccessible routes for the structural characterization, especially of irreproducible targets [2, 7].

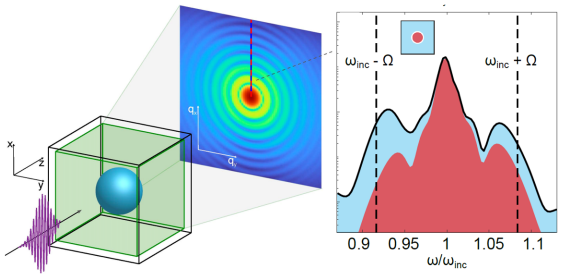

Figure 1: Simulated diffraction of a near resonant XUV pulse (21.6 eV) from a Helium droplet with $R = 300$ nm. At sufficiently high intensity the scattered light shows substantial spectra lwings due to local Rabi-cycling of the ground and excited state population in the Helium nanodroplet. From [8].

The information cast into the diffraction image is even richer as soon as nonlinear response features and quantum effects beyond liner response become relevant. This scenario has recently been explored in a first theoretical study [8] that predicts distinct new features due to the nonlinear quantum coherent response using a self-consistent description via a Maxwell-Solver coupled to a 4-state density-matrix approach. In particular, for the considered resonant excitation of Helium droplets the nonlinear response distorts the spatial structure of diffraction fringes and leads to spectral fragmentation of the scatterd light, see Fig. 1. This regime defines the core of this IRTG project.

Research goals and working program

We aim at exploiting so-far unresolved signatures in coherent diffractive imaging that will allows one to trace the spatial and temporal structure of the laser induced coherent quantum dynamics in wide-bandgap dielectric

materials or quantum fluids such as ^{4}He. Merging nonlinear and coherent local electronic dynamics with light propagation in such nanoscale quantum targets unfolds into a rich set of quantum processes ranging from coherent population transfer pattern, Rabi-cycling, spatial exciton migration, induced transparency, and optically controlled slow light.

A key fundamental prerequisite is that the imaging pulse duration is of the order of typical dephasing times of usually few fs, which is nowadays feasible as both XUV HHG sources as well as XFELs can deliver sub-fs pulses. In this case, the spatiotemporal visualization of the nontrivial coherent quantum dynamics by coherent diffractive imaging becomes possible. The analysis and clarification of the underlying physics and to-be-expected signatures for single or multi-color scenarios define the research goals of this project. One example would be the dynamic visualization of excitation waves resulting from local Rabi flopping, see Fig. 2.

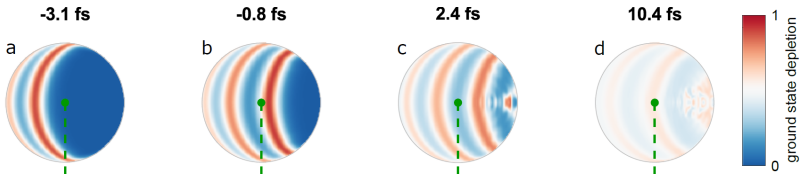

Figure 2: Dynamics of spatial excitation waves in the resonantly driven Helium nanodroplet. From [8].

As a first step we will explore theoretically the new opportunities that emerge from using waveform-controlled optical laser pulses from the near-infrared up to the XUV and x-ray spectral range as controllable drivers of quantum dynamics with the exquisite temporally and spatial resolution of single shot CDI. We will implement and test different approaches to model the excitation and control of sub-cycle population dynamics and will study the spatial mapping of the classical and quantum response via CDI.

Firm understanding of the underlying nonlinear photonic quantum dynamics will be key to harnessing a largely unexplored set of applications, ranging from quantum imaging including NIR pump XUV probe imaging scenarios to light-controlled quantum electronics in nanosystems.

While field propagation will be treated on a classical level by solving Maxwell's equations, various approaches will be considered for describing the nonlocal/local electronic response. Therefore both purely local models such as density matrix models as well as band structure-based descriptions via semiconductor Bloch equations (SBE) or fully fledged numerical solutions of the time-dependent Schrödinger equation (TDSE methods) will be explored.

Key work packages for PhD projects:

- self-consistent modelling of light propagation coupled to quantum few-level dynamics or SBE
- scattering simulations based on effective optical properties from TDSE simulations
- analysis of signatures from AC- and DC-Stark shifts in CDI
- spatial retrieval of excitation maps in resonantly driven few-level nanosystems

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