Electron-photon Correlations for Probing Optical Excitations Nanometer-Scale Dynamics

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Background and state-of-the-art

Coherent interactions between particles and quasi-particles are fundamental to quantum technologies. In solid-state devices, the coherence time of these excitations is limited by interactions with the lattice, surfaces, interfaces, and defects, restricting their potential applications. These interactions inherently occur at the nanometer to atomic scale, as the objects themselves are spatially confined. Therefore, techniques capable of probing bulk materials with such high spatial resolution are essential.

Focused electron probes within electron microscopes (EM) enable diffraction, imaging, and spectroscopy with atomic precision. However, conventional experiments rely on Poissonian electron sources and lack temporal resolution, which is crucial for studying dynamic processes. The two groups proposing this doctoral project have developed innovative time-resolved electron microscopy and spectroscopy experiments that offer temporal resolution in the nanosecond to femtosecond range and ultimately sub-poissonian electron sources. These recent advancements are central to this project, enabling the successful candidate to explore the coherence and dynamics of optical excitations with nanometer spatial resolution.

Objectives

The core of the doctoral project is to investigate excitation dynamics at the nanometer scale in semiconductors and the photon generation process resulting from their decay. In a typical electron spectroscopy experiment, a fast electron (30-300 keV kinetic energy) with transverse dimensions ultimately smaller than an angström traverses a ~100 nm thin sample. With probabilities below 10^{-3}, the fast electron can lose energy, creating a primary excitation that propagates, interacts with the solid, and ultimately decays into photons. While the majority of excitations created in a solid are bulk plasmons (10-30 eV range), the precise energy loss per electron cannot be controlled, enabling the creation of phonons, excitons, and other excitations. Consequently, time-averaged experiments cannot disentangle the relationship between excitations and light emission.

The experimental approaches developed by the partners measure the energy loss of each scattered electron and the corresponding photon emission probability by unique time-correlated techniques. Both groups were pioneers in developing these methods in 2022.

By pushing these methods to ps resolutions, the project will address the following key questions:

- To what extent is the coherence of excitations maintained during their propagation in a solid?
- How does the excitation energy influence the probability of generating single and multiple photons from light emitters?
- Can controlling the excitation energy enable a solid-state electron-excited heralded single-photon source in semi-conducting nanodevices?

Specifically, the successful candidate will investigate excitations in nitrogen-vacancy centers in diamond, defects in h-BN flakes, and III-N heterostructures using time-resolved electron spectroscopy to answer the aforementioned questions.

Complementarity of the co-advisers

The partners have developed distinct time-resolved methods for electron microscopy and spectroscopy. The first method is based on a state-of-the-art ultrafast laser-driven electron source, enabling electron-photon experiments with femtosecond temporal resolution and moderate energy resolution (hundred of meV). The second method utilizes novel time-resolved electron detectors to implement synchronized electron-photon coincidence spectroscopy with moderate (nanosecond) temporal resolution and state-of-the-art spectral

resolution (few meV). Allying the two complementary approaches is key for understanding excitation dynamics in semiconductors.
The two groups also possess distinct scientific cultures which would prove fruitful to tackle the thesis subject. The group at U. Paris-Saclay has pioneered the application of electron spectroscopy for plasmonics, demonstrated the first detection of single-photon sources in an electron microscope, and led the implementation of event-based detectors for electron energy loss spectroscopy. The group at Max Planck, Göttingen, has performed pioneering experiments with ultrafast electron sources, including the observation of coherent control of the energy exchange between free-electrons and photons, the dynamics of matter under photo-excitation, and the generation of heralded photons by electron beams in photonics devices.
The successful candidate will benefit from the extensive knowledge and worldwide recognition of these two groups and will contribute to strengthening their collaboration.

Infrastructure available for the project

The two groups advising this project are world leaders in the field of spectrally- and/or time-resolved electron microscopy, having pioneered the use of electron spectroscopy for nano-optics and quantum nano-optics for more than 10 years. A unique aspect of their work is the development of spectroscopy methods through hardware development in their respective laboratories, which allowed the creation of two unique microscopes with some of the key functionalities described below.

Pulsed source electron microscope (Max Planck Institute for Multidisciplinary Sciences)

Nanometer scale femtosecond electron-photon spectroscopy, samples cooled to ~100 K, superconducting nanowire photon detectors, electron event-based detectors, Hanbury-Brown and Twiss (HBT) setup. Dedicated hard- and software for ultrafast imaging and spectroscopy, and simulations of ultrafast materials dynamics.

Electron microscope with synchronized electron-photon detection (U. Paris-Saclay)

Nanometer scale nanosecond electron-photon spectroscopy, 5 meV EELS resolution, samples cooled to 100 K or 5 K, HBT setup, electron event-based detectors, advanced knowledge on correlation electronics hardware and software.

In addition, the project will benefit from the infrastructure available at the two sites, including cryogenics, electronics, vacuum and micro/nano-fabrication technologies.

"3i" character of the project

The thesis will be based at U. Paris-Saclay, supervised by Luiz Tizei and Mathieu Kociak, and co-supervised by Armin Feist and Claus Ropers at the **Max Planck Institute for Multidisciplinary Sciences**. The successful candidate will start their PhD at U. Paris-Saclay for 18 months, will then spend 12 months at **Max Planck Institute for Multidisciplinary Sciences**, and will use the final 6 months at U. Paris-Saclay to write their thesis. During this period, the two groups will organize meetings to keep the project on track.

Advisors experience in thesis direction

Luiz Tizei and Mathieu Kociak have HDRs and are members of EDOM. They have co-supervised 5 theses. In total, they have supervised 17 theses. Co-affiliated with the University of Göttingen, Claus Ropers has supervised 18 completed PhD theses, and Armin Feist has supervised more than 10 Master's and Bachelor's theses. Most of the PhDs advised by the two groups currently hold research positions, either in the public or the private sectors.