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# ICFO International School on the Frontiers of Light

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Photonics with Free  
Electrons

5-7 July, 2023



# SHORT TALKS

5 July, 15:45-16:45

1

Anaswara Ramachandran

*University of Konstanz*

Pump-Probe bio-TEM with optically compressed pulses

2

Gloria Davidova

*Cornell University*

Understanding the Effect of Vibronic Interactions in Organic Materials for Polaritons

3

Valerio di Giulio

*ICFO*

Free electron beams as a source of quantum light

# SHORT TALKS

6 July, 12:00-12:40

4

Jing Li

*School of Physics, Peking University*

Topologically Protected Strong-Coupling of  
Photonics with Free Electron

5

Mohit Bera

*ICFO*

Steady-state Quantum Thermodynamics  
with Synthetic Negative Temperature

# SHORT TALKS

7 July, 15:45-16:45

6

Brittany (Ying-Ying) Lu

*University of California, Los Angeles*

X-Ray Generation with Shaped Ultrafast Electron Bunche

7

Dilan Perez

*ICFO*

Probing Superconductivity in Bernal Bilayer graphene devices

8

Leila Prelat

*ICFO*

Polariton Smith-Purcell emission

## **1. Pump-Probe bio-TEM with optically compressed pulse**

All matter transformations involve the motions of atoms and electrons. Many processes like atomic motion, molecular vibrations, photon emission, scattering etc. take place in very fast time scales. For understanding these processes, it is necessary to visualize the motion of atoms and electrons in space and time. For that, we need to achieve femtosecond and angstrom resolutions, therefore we are combining a fs-laser and a transmission electron microscope (TEM). However, the time resolution, given by the electron pulse length, is not good enough due to space charge and wave dispersion. Therefore, I will use terahertz pulses to compress the electron pulses in time. The planned THz field-controlled electron microscopy will consist of two THz-electron interactions: one is used for electron pulse compression and the other for temporal characterization by streaking. Compression using THz fields provides short electron pulses and provides stability in time. I will then apply this emerging, worldwide unique apparatus to study selected phenomena in ultrafast dynamics. One of my first applications will be the use of THz pulses in biomolecular imaging. We aim at putting a single-cycle THz field onto a phytochrome-A crystal and use time-resolved electron diffraction to look for potential structure changes in the strong excitation field.

## **2. Understanding the Effect of Vibronic Interactions in Organic Materials for Polaritons**

A molecular exciton polariton is a hybrid light and matter state which forms from the coupling of a photon and an exciton, the photoexcited state of a organic semiconductor or dye. Typically these states are studied by confining a layer of dye between closely spaced mirrors in a Fabry-Perot Microcavity. In the case of a simple polariton system, the interaction between one photon state and one exciton state in resonance can produce an upper and lower polariton band. This resonance is known to be collective across all dipoles available inside the cavity and the formed coherent states delocalize over the field volume. The newly formed hybrid states possess angular dispersion from the photonic mode of the cavity, which forces an anticrossing at the exciton-photon resonance. When more excitons are coupled to the photonic mode of the cavity, these interactions can give rise to a ladder of polariton states with varying degrees of photonic versus exciton character. In this study, we consider three different organic molecules with prominent vibronic structure, and thus multiple strong absorbing exciton transitions, in their steady-state absorption. The molecules DPP, PDI, and PTCDA were deposited via thermal evaporation within silver mirror microcavities. Strong coupling was attained in all structures, and we used angle-dependent reflectivity measurements to determine the dispersions of the polaritonic states for comparison to optical modelling. Studying the relation between the vibronic structure and the dispersion of the polariton branches is useful to aid in the development of devices based on exciton polaritons, like low-threshold lasers, and lays the groundwork for future studies with more sophisticated cavity architectures.

## **3. Free electron beams as a source of quantum light**

Electron beams in electron microscopes offer the possibility to measure material properties with nanometer resolution in several ways depending on the signal analyzed. In particular, electron energy-loss spectroscopy (EELS) is established as a rich source of information allowing for the visualization of optical surface modes as well as the bulk response of materials.

Recently, the synchronization ultra-short laser and electron pulses at the specimen triggered several studies aimed to record the near field generated by the ultrafast dynamics in nanostructured materials. This approach leverages on the evanescent nature of the field produced by the excitations, thus overcoming the energy-momentum mismatch between photons and massive particles in free space. Here, strong laser amplitudes ( $\sim 10^8$  V/m) make the electron interact with highly populated coherent states and thus exchange multiple light quanta. This technique, known as photon-induced near-field electron microscopy (PINEM), was exploited to longitudinally and transversally modulate electrons. Such possibility prompted a cascade of theoretical studies aimed to explore the effects of the interaction between so-shaped electrons and a secondary specimen, in particular trying to answer the question: “which state of light would a shaped electron produce in the interaction with a cavity?”.

In this work, we use macroscopic quantum electrodynamics to analyze the creation of quantum states of light by means of free electrons. Different kinematical regimes are studied, starting from a situation in which e-beams travel with a kinetic energy much higher than the resonant energy of the cavity mode and then going to more extreme cases of zero or few quanta transfer from the electron to the electromagnetic field. We conclude that free electrons under suitable conditions can behave as evanescent quantum sources of light, granting us access into the direct creation of coherent and squeezed states in cavity-supported dark modes.

#### **4. Topologically Protected Strong-Coupling of Photonics with Free Electron**

We propose a robust scheme of studying the strong interactions between free electrons and photons using topological photonics. Our study reveals that the topological corner state can be used to enhance the interaction between light and free electron significantly. The quality factor of topological cavity can exceed 20,000 and the corner state has very long lifetime even after the pump pulse is off. And thus, the platform enables to achieve strong interaction without the needs for zero-delay and phase-matching as in traditional photon-induced near-field electron microscopy (PINEM). We also show the topological photonic structures can be utilized to shape free electron wavepackets very flexibly, which facilitates the control of quantum electrodynamical (QED) processes and quantum optics with free electrons in the future.

#### **5. Steady-state Quantum Thermodynamics with Synthetic Negative Temperatures**

A bath with a negative temperature is a subject of intense debate in recent times. It raises fundamental questions not only on our understanding of negative temperature of a bath in connection with thermodynamics but also on the possibilities of constructing devices using such baths. In this work, we study steady-state quantum thermodynamics involving baths with negative temperatures. A bath with a negative temperature is created synthetically using two baths of positive temperatures and weakly coupling these with a qutrit system. These baths are then coupled to each other via a working system. At steady-state, the laws of thermodynamics are analyzed. We find that whenever the temperatures of these synthetic baths are identical, there is no heat flow, which reaffirms the zeroth law. There is always a spontaneous heat flow for different temperatures. In particular, heat flows from a bath with a negative temperature to a bath with a positive temperature which, in turn, implies that a bath with a negative temperature is 'hotter' than a bath with a positive temperature. This warrants an amendment in the Kelvin-Planck statement of the second law, as suggested in earlier studies. In all these processes, the overall entropy production is positive, as required by the Clausius statement of the second law. We construct continuous heat engines operating between positive and negative temperature baths. These engines yield maximum possible heat-to-work conversion efficiency, that is, unity. We also study the thermodynamic nature of heat from a bath with a negative temperature and find that it is thermodynamic work but with negative entropy.



## **6. X-Ray Generation with Shaped Ultrafast Electron Bunche**

We explore the generation of parametric x-ray generation through the interaction of ultrashort non-relativistic electron bunches traveling through thin-film multi-stacked layers and triggering photon emission through cathodoluminescence. We present an ultracompact instrumental design of an ultrafast free electron beamline for performing temporally and spatially shaped electron bunches down to the attosecond scale, as well as for the generation of coherent X-ray radiation in the quantum electrodynamical regime.

## **7. Probing Superconductivity in Bernal Bilayer graphene devices**

*TBC*

## **8. Polariton Smith-Purcell emission**

We theoretically study the coupling between free electrons and polaritons mediated by a single small scatterer, as well as polariton Smith-Purcell emission assisted by scatterer arrays, obtaining the conditions to maximize electron--polariton coupling.



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