

Quantum Light and Free Electron Beams

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Fast electrons in electron microscopes offer the possibility to measure material properties with nanometer resolution in several different fashions depending on the signal analyzed. For instance, inelastically scattered electrons can be collected and energetically separated in order to obtain spectra as a function of their energy loss or, with the same purpose, the radiated light can be collected in the far field and filtered in polarization and frequency. In particular, electron energy loss spectroscopy (EELS) can be considered the richest source of information as it carries the influence from the surface-sustained modes as well as from the bulk response of the system. Additionally, low-loss EELS provides insight into the spatial and spectral distributions of plasmons in metallic nanostructures and in more recently experiments used also to detect phonon resonances in polaritonic materials thanks to remarkable improvements in the instrument resolution. Additionally, the synchronization of femtosecond laser illumination with electron pulses at the sample allows to study the ultrafast dynamics of nanostructured materials and their influence on optical near-fields. The non-monochromatic field, produced by the excitations in the material, efficiently overcomes the severe energy-momentum mismatch between free photons and free electrons yielding appreciable interaction probabilities. This strong light-electron coupling leads to multiple exchanges of quanta between the electron and the evanescent component of the optical field. Based on this principle, photon-induced near-field electron microscopy [1] (PINEM) is performed by analyzing the resulting multiple gain and loss features in the electron spectra. PINEM experiments have so far relied on coherent light sources such as lasers, for which the measured spectra are well reproduced by assuming sample bosonic excitations that are coherently populated with a large number of quanta. The probability of each electron spectral peak associated with a net exchange of l quanta is then simply given by squared Bessel function. Additionally, this technique, performed with coherent light, has been combined with an electron free propagation of few millimeters to yield a temporal compression of the electron density of the order of attoseconds [2]. Despite the increasing number of results obtained in this field, different aspects of the interaction between free electrons and confined light remain unexplored. For instance, the effects induced by the replacement of the coherent laser pulses with a quantum light source on electron spectra, are expected to show interesting physics significantly departing from the one previously studied [3].

In this work [4], we use macroscopic electrodynamics to describe the quantum nature of the evanescent optical field and to understand how the resulting electron spectra strongly depend on the statistics of the sample excitations and their population. In particular, we show that the light autocorrelation functions can be directly retrieved from the ratios of electron gain intensities. In particular, in the limit of highly populated mode, we recover the usual results of classical PINEM for Fock and coherent states but we find a significant difference in the distribution of the gain and loss peaks of electron spectra in the case

of thermal state (see Fig. 1). Furthermore, we study the role of quantum states of light, e.g. squeezed states, in shaping the longitudinal part of the electron wave function after a macroscopic propagation distance observing a faster compression of the electron density for light states with low phase uncertainty (see Fig. 2). Finally, we analyze the possibility of directly creating coherent squeezed states in a cavity-supported dark mode by letting a free electron beam interacting with it. We believe that this work represents a step of fundamental importance in the understanding of quantum many bodies excitations, their statistics, in improving the ability of tailoring the electron wave function via the use of light as well as a fundamental step to merge the fields of electron beams and quantum optics [5].

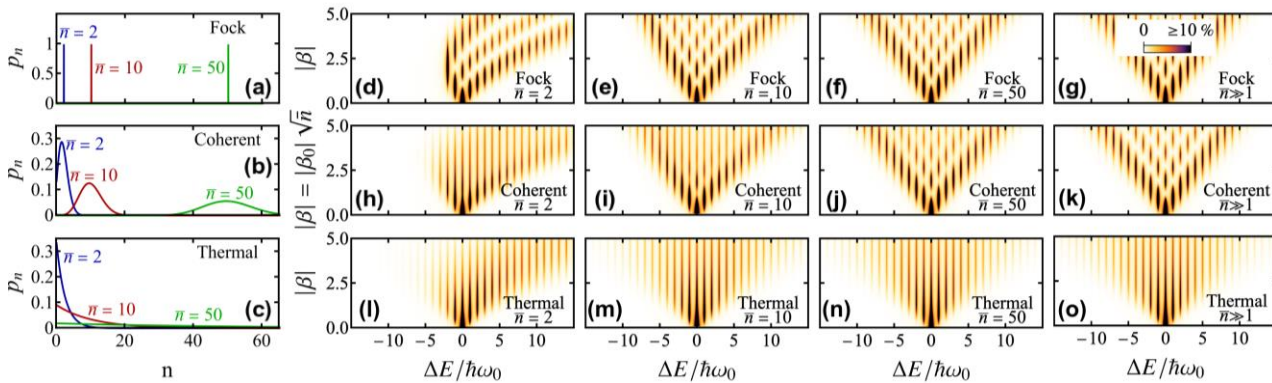


Figure 1. Dependence on boson population distribution in the interaction with an electron beam. (a-c) Distribution of the probability p_n for occupation of each state $|n\rangle$ in the three types of population statistics considered at the moment of electron interaction: Fock (a), coherent (b), and thermal (c), with average values $\bar{n} = 2, 10, \text{ and } 50$. (d-o) Electron spectra after interaction with a dipolar mode with the initial populations of (a-c) as a function of the electron-mode coupling parameter $|\beta_0|\sqrt{\bar{n}}$.

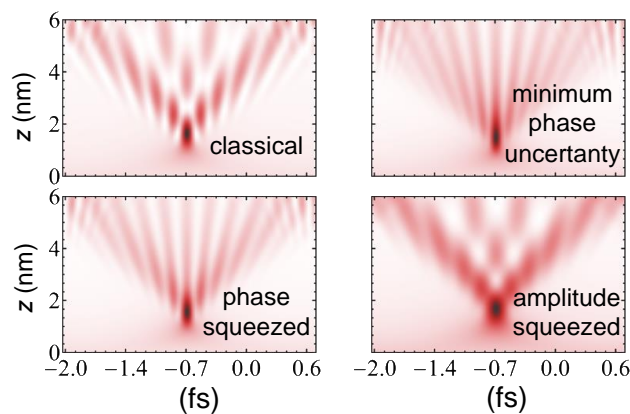


Figure 2. Electron density after the interaction with a mode for different quantum light states: classical (coherent state), minimum phase uncertainty state, phase and amplitude squeezed states.

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 [3] Valerio Di Giulio, Mathieu Kociak, and F. Javier García de Abajo, *Optica* **6** (2019), 1524-1534.

[4] V. Di Giulio and F. J. García de Abajo, "Free-electron shaping using quantum light", *Optica* **7**, 1820 (2020).

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