Ultrafast Electron Spectroscopy with Slow and Fast Electrons

Talebi, N.[1]

[1] Max Planck Institue for Solid State Research, Germany

The local density of states of nanostructures can be characterized using either laser radiation or controlled electron beams. Whereas these methods are assumed to be complementary to each other in the sense of acquired information, interesting phenomena appear by combing laser and electron guns in an electron microscopes. This method, which is called ultrafast electron diffraction or photon-induced near-field electron microscopy (PINEM), where for the former diffraction patterns and for the latter spectra are acquired, has been recently further developed by several groups around the world, into a form of time-resolved pump-probe characterization methodology.[1]

The idea behind PINEM is the coherent control of photoemission electrons from an electron gun using laser light, whereas a second path of the laser light excites the sample at a controlled delay with respect to the photoemission electrons. An inverse approach, i.e. the coherent control of cathodoluminescence (CL) radiation in interaction of the electron beams with nanostructure can be used as well (Fig. 1a).[2, 3] This latter method have advantages over the former, in offering a mutual coherence between the phase of the CL emission and the evanescent field adjacent to the electron. This mutual coherence can be facilitated by proposing particular designs for an electron-driven photon source (EDPHS), which is composed of precisely fabricated metallic metamaterials (Fig. 1b). The sample is located along the electron path at certain distance (L) with the EDPHS, and hence interacts with both the electron and the EDPHS light. The light emitted from the sample will interfere with the EDPHS light at the far-field, and this in particular causes interference maps at the CL detectors (Fig. 1c). Here, we further discuss the progress in design, fabrication, and characterization of an EDPHS, using the concepts from transformation optics and metamaterials, hence describing in more details our steps towards the realization of spectral interferometry with electron microscopes.

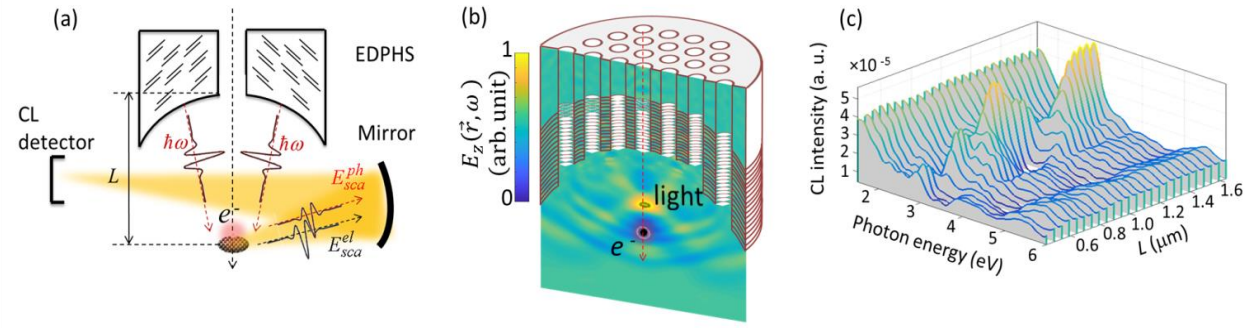

Fig. 1. (a) an electron interacting with the EDPHS and the sample. (b) Interaction of the electron with a precisely engineered EDPHS will result in a focused and coherent transition radiation. The emitted light which is detected then by a CL setup will demonstrate interferences within the energy-distance map.

Although PINEM has been only discussed within the framework of transmission electron microscopes, fundamentally there is no reason to restrict the technique to the area of relativistic beams. Low energy electrons at the kinetic energies of less than keV ranges are even more sensitive to the electromagnetic interactions, and hence can be more sensitive probes of electronic excitations of the samples. Using electron beams at only few tens of electron-volt energies, there exists certainly benefits in combing PINEM with point-projection electron microscopy (PPEM).[4] Here, we will discuss the theory underlying the interaction of slow electron beams with the electron gas and optical excitations within a carbon nanorod, for the purpose of understating such interactions beyond Wolkow approximations. For this, we use a

recently developed self-consistent numerical toolbox within the Maxwell-Schrödinger semi-classical framework.[5] We notice both transversal and longitudinal interferences (Fig. 2), and formation of several Ewald spheres displaced at the momentum domain at steps of harmonics of laser energy.

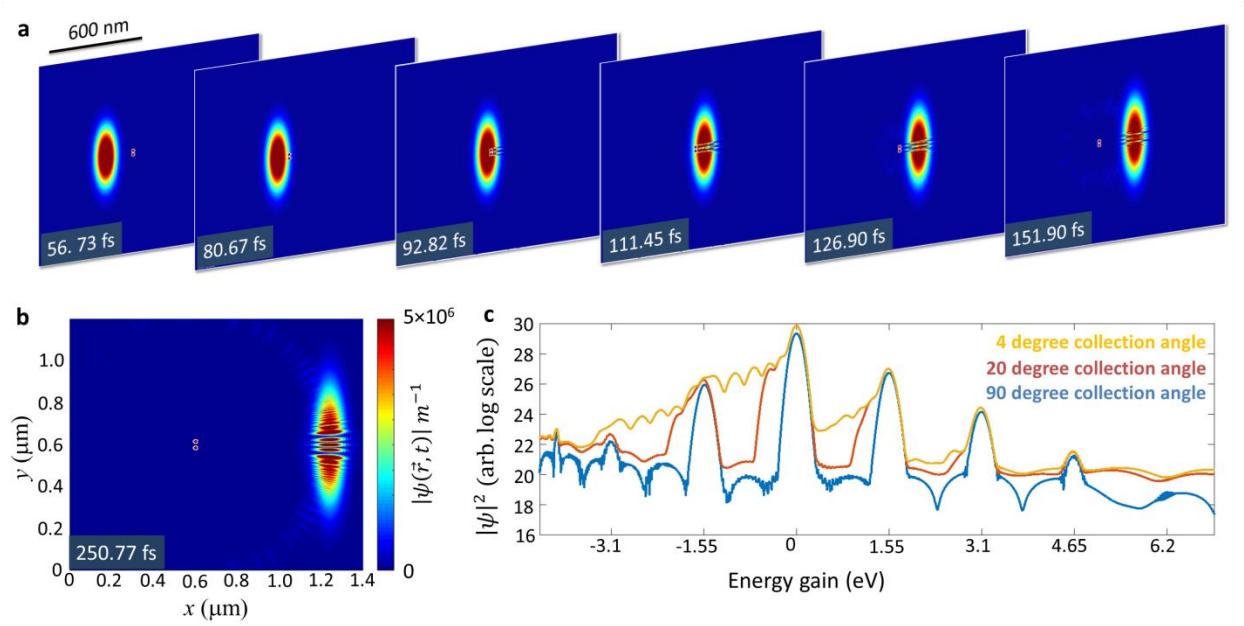

Fig. 2. (a, b) Snapshots of the spatial representation of the magnitude of the electron wave function during its interaction with two carbon nanorods with the diameter of 10 nm. The initial kinetic energy of the electron wave function is 50 eV. The lateral and longitudinal broadening of the initial wave packet is 1.8 nm and 20 nm respectively. The carbon rods are additionally excited by laser light at the carrier wavelength of 800 nm (1.55 eV), and the peak amplitude of 1×10^8 V m^{-1}, and the temporal duration of 20 fs. (c) Energy distribution of the electron wave function after the interaction, which is clearly modulated at energies equal to the harmonics of the laser energy, both at the energy loss and energy gain sides, though not symmetrical, hinting at the presence of a non-Hermitian Hamiltonian.

References

1. Barwick B, Flannigan DJ, Zewail AH. Photon-induced near-field electron microscopy. *Nature* 2009, **462**(7275)**:** 902-906.

2. Talebi N. Spectral Interferometry with Electron Microscopes. *Sci. Rep.-Uk* 2016, **6**.

3. Talebi N. Interaction of electron beams with optical nanostructures and metamaterials: from coherent photon sources towards shaping the wave function. *J. Opt. (Bristol, U.K.)* 2017, **19**(10)**:** 103001.

4. Quinonez E, Handali J, Barwick B. Femtosecond photoelectron point projection microscope. *Rev. Sci. Instrum.* 2013, **84**(10).

5. Talebi N. Schrödinger electrons interacting with optical gratings: quantum mechanical study of the inverse Smith-Purcell effect. *New J. Phys.* 2016, **18**.