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Integrated Nanophotonic Electron Beam Modulators Enable Ultra-High Precise Method for Calibrating EELS Spectrometers

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Swift electrons and light do not interact directly due to their large momentum mismatch. However, free electrons can couple to the optical fields of nanostructures, e.g., surface plasmon polaritons, stimulated by intense light pulses. Such physical phenomena are at the heart of the Photon-induced Near Field Electron Microscopy (PINEM) technique [1]. The spatially confined optical excitations modify the wavefunction of free electrons that lose and gain discrete amounts of energy quantized by the electron-photon interactions. The PINEM interactions produce characteristic sidebands in the EELS spectra spaced at energies corresponding to the wavelength of the stimulating light pulses. Such interactions provide a quantum coherence in which the transverse and longitudinal electron wavefunctions of the electron pulses can be coherently manipulated [2-4] .

Recent research on the coherent quantum interaction of PINEM in Ultrafast TEMs has led to the development of new techniques, like Ramsey-type holography [2], which allows for the dynamical reconstruction of the quantum state of an electron beam interacting with a material of interest. These PINEM approaches demonstrate the potential for a combined attosecond-nanometer-millielectronvolt resolution in time-resolved spectroscopic imaging [4]. Beyond the imaging application, it also allows the implementation of optically-controlled and spatially-resolved quantum measurements in parallel, providing an efficient and versatile tool for electron quantum optics and coherent quantum spectroscopy.

Henke et al. [5] , in a recent study, used integrated photonics chips to modulate the phase of a continuous electron beam in conventional TEM mode. These nanophotonic chips enable continuous wave (CW) PINEM via a high-finesse ($Q\sim10^6$) microresonator. The resonator waveguide is designed for efficient phase matching of 120 kV electron–light scattering at extremely low, continuous-wave optical powers. Passing electrons in the vicinity of the microresonator cavity operating at coupled optical powers of ~10 mW can generate EELS spectra with several hundred sidebands spaced at the precise photon energy of narrowband laser light pumping the cavity. Such devices and the CWPINEM technique can enable EELS spectroscopy with μeV resolution in conventional TEMs.

Here, we describe a new method for the dispersion calibration of an electron energy loss spectrometer (EELS) in a transmission electron microscope (TEM) derived from the analysis CWPINEM produced by these nanophotonic microresonators (See Figure 1). The method relies on a strong coupling of the free electrons with the optical field produced by photons circulating in the microresonators, which produce a high number of precisely spaced sidebands in the EELS spectra. By counting the number of photons exchanged with the electrons in the EELS spectra and knowing the precise photon wavelength (emitted by a calibrated high-precision light source), we can deduce the electron energy with a precision

of few μeV – a precision as much as three orders of magnitude over conventional techniques, e.g, such as using BN and NiO samples (See Figure 2). Since the CWPINEW produced by strong coupling can generate spectra having sidebands that span the range of the detector, piece-wise (windowed) FFT analysis can be used to determine the non-linearity of the EELS system. Such calibrations combined with high-quality dark references reduce correlated noise [6] and have the potential to obtain high-energy resolution spectra on beam-sensitive materials.

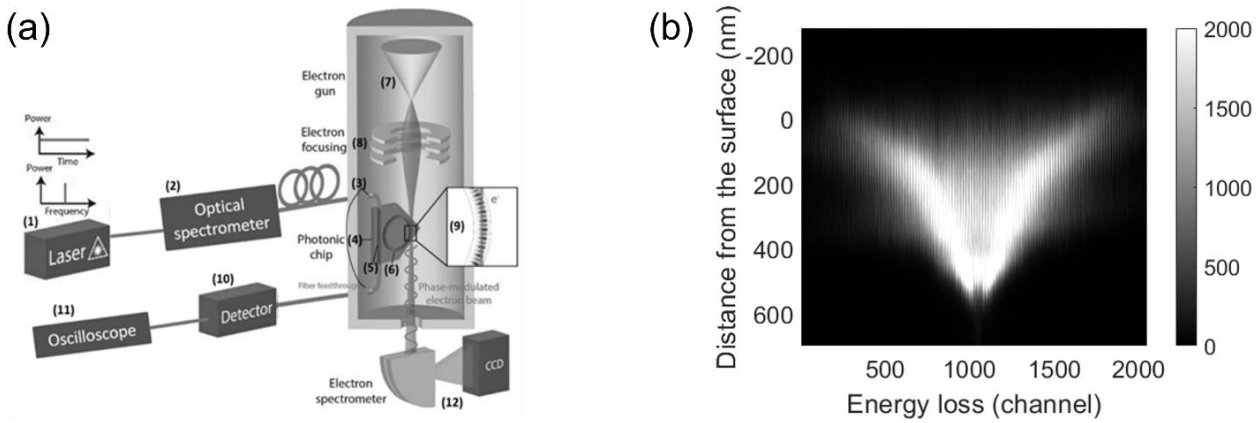

Figure 1. (a) The schematic of the experimental setup **(b)** EELS spectrum of the electrons after interacting with the electric field of the cavity pumped with a laser wavelength of 1533 nm. The vertical scale is the distance normal to the sample surface. The horizontal axis corresponds to the electron energy with the ZLP located at the 1100th pixel. The nominal dispersion is 50 meV/channel.

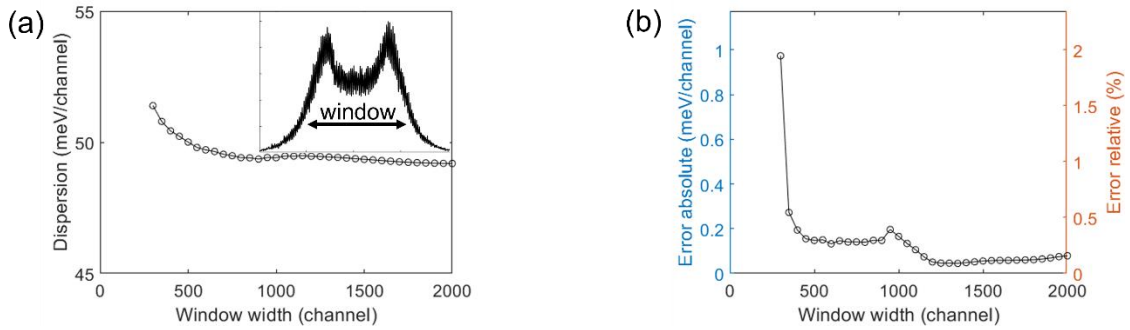

Figure 2. (a) The spectrometer dispersion obtained from the FFT of the spectrum shown in the inset utilizing the windows of different widths at the central part of the spectrum. The spectrum is the cross-section of the 2D EELS spectrum (Fig. 1(b)) corresponding to a distance of 100 nm from the sample surface. **(b)** The absolute and relative error of the determined dispersion depending on the width of the window.

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