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# Defocus Phase Contrast in Photon-Induced Near-field Electron Microscopy

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Electron microscopy encompasses a variety of techniques to study the nanoscale optical properties of materials and devices. In particular, spontaneous inelastic electron-light scattering (IELS), resulting in cathodoluminescence and electron-energy loss, is frequently used to spectroscopically characterize the photonic density of states with high spatial resolution over a broad optical bandwidth [1]. In addition, stimulated scattering in the presence of optical illumination of the sample, allows for mode- and polarization-selective measurements of optical near fields in the form of Photon-Induced Near-field Electron Microscopy (PINEM) [2]. In this technique, inelastic scattering produces sidebands in the electron kinetic energy spectrum corresponding to energy gains and losses in multiples of the photon energy. The process has been used to quantitatively characterize nanoplasmonic and chiral near fields [3, 4], and to induce a longitudinal and transverse phase modulation of electron beams [5, 6], including the generation of attosecond electron pulse trains [7]. The number of populated sidebands is a direct measure of the near-field strength [1, 8, 9], and importantly, as the scattering process is coherent in nature, the spatial distribution of the optical near-field phase is imprinted onto the electron wave function. Here, we use Fresnel-mode defocus imaging to reveal electron phase contrast induced by stimulated IELS.

The experiments are carried out at the Göttingen Ultrafast Transmission Electron Microscope (UTEM) operated with a laser-triggered field emission electron gun delivering electron pulses of high spatial coherence. [10]. The instrument allows for the study of ultrafast nanoscale processes, offering a wide range of imaging techniques and contrast mechanisms. In the experiments, a gold nanotip is excited by femtosecond laser pulses (1.8 ps duration, central wavelength of 800 nm) shown in Fig. 1 a. Collimated electron pulses are used for PINEM, imaging individual sideband amplitudes of the electron wave function $\Psi_N$ ($N$ photon order). To this end, we employ a single-electron-sensitive hybrid pixel detector behind an imaging electron spectrometer with a controllable slit for energy selection. Figure 1 c presents an in-focus image collected from all electrons experiencing energy loss (selection of all loss orders $N$), exhibiting the shadow image of the nanotip and a standing wave pattern resulting from interference between surface plasmons excited on the conical tip and far-field radiation scattered at the tip-supporting shaft. A corresponding simulation of the region close to the tip apex is displayed in Fig. 1 d, showing general agreement. The in-focus image intensity $I(x, y)$ measures the inelastic scattering probability of electrons and is thus governed by the time-dependent electric field $E$ on the electron trajectory along the $z$-axis (ω: optical frequency, $v_e$: electron velocity) [8, 9]:

$$I(x,y) = \sum_{N<0} |\Psi_N^2|, \text{ where } \Psi_N = J_N(2|g|)e^{iN\arg(g)} \text{ and } g = \frac{e}{2\hbar\omega}\int_{-\infty}^{\infty} E(x,y,z)e^{-i\frac{\omega}{v_e}z}\mathrm{d}z.$$

It is evident that, in the absence of aberrations, the image intensity is independent of the phase of the coupling parameter $g$. However, gradients in the phase of $g$ result in electron beam deflections, which can be revealed using phase-contrast imaging. Specifically, we employ Fresnel-mode Lorentz imaging, a phase-contrast technique that is widely used to characterize in-plane magnetic fields. As illustrated in Fig. 1 b, defocus imaging translates locally varying beam deflections into amplitude modulations. In the specific geometry in study, strong phase gradients are induced near the apex of the tip (cf. simulation of phase gradients in Fig. 1 d, center). Working under overfocus conditions modifies the energy-filtered image, producing a bright spot of enhanced intensity near the tip apex, and caustics along the shaft in both experiment (Fig. 1 c, bottom) and simulation (Fig. 1 d, bottom, obtained from values of $g$ evaluated from Eq. 1 with the near field numerically calculated for a long tip using the boundary-element method).

In the presentation, we will compare differences in imaging contrast using energy gain and loss sidebands with $N > 0$ and $N < 0$, coupled by the relation $\Psi_{-N} = \overline{\Psi_N}$ and, thus, exhibiting opposite deflections and phase gradients (Fig. 2). Moreover, we will explore algorithmic near-field phase reconstruction and polarization-dependent imaging.

Studying PINEM with Fresnel-mode Lorentz contrast at a gold nanotip, we demonstrated a sensitivity to the spatial phase distribution of the optical near field. Our approach can be extended to other existing phase contrast methods in transmission electron microscopy, including differential phase contrast [11] or holography.

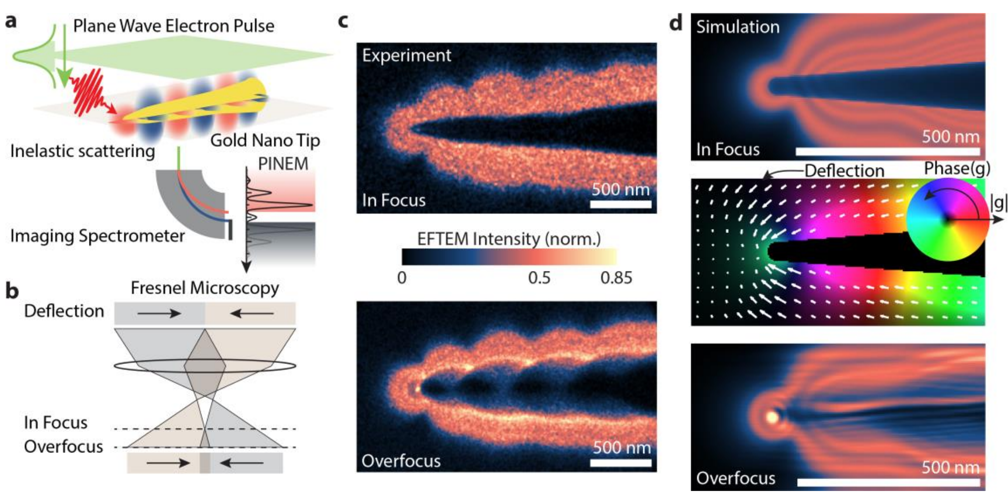


**Figure 1.** Defocus phase contrast in PINEM. (a) Sketch of the PINEM experiment with IELS and energy-filtered imaging. (b) Fresnel microscopy shows converging deflections as bright contrast at the boundary. (c) EFTEM micrographs reveal the amplitude and deflections of the optical near-field under in-focus and overfocus imaging conditions, respectively. (d) Simulated amplitude and phase of the nearfield interaction $g$ and resulting in focus and overfocus EFTEM images.

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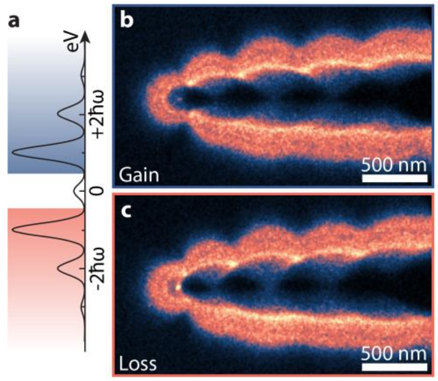


**Figure 2.** Gain and loss asymmetry in Fresnel PINEM. Filtering energy gain or loss sidebands of the PINEM spectrum (a) results in different phase contrast in the EFTEM micrographs (b) and (c), respectively.

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