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Enhanced Free-Electron Wavefunction Modulation via Photon-Induced Near-Field Electron Microscopy (PINEM) with Shaped Light Fields

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Background and aims

Photon Induced Near-Field Electron Microscopy (PINEM) is a novel imaging technique that relies on the inelastic interaction between a free-electron beam and laser light, mediated by a thin film or a nanostructure [1]. This technique is generally employed to study the ultrafast dynamics of local fields generated by light-induced polaritonic excitations in nanoscale systems. Recently, we further exploited the quantized nature of electron-light interaction, that is intrinsic to PINEM, to achieve coherent control of the electron wavefunction [2-4]. So far, light-induced shaping has relied on Gaussian laser beams illuminating plasmonic nanostructures. In contrast, on-demand electron modulation for dynamic and versatile control has not yet been fully explored. Here, we show that shaped light can be used to introduce an arbitrary modulation on the transverse electron beam profile [5]. In fact, it is already possible to arbitrarily control light fields through spatial light modulators (SLMs) and our goal is to exploit PINEM to transfer this tunability to electron beams, thereby broadening the capabilities of ultrafast TEMs (UTEMs).

Methods

Our experiments are conducted in a UTEM, specifically a JEOL 2100 TEM modified as shown in Fig. 1. A 200 keV electron beam interacts with a femtosecond IR laser beam within a Photonic-based ELectron Modulator (PELM), an additional module of our TEM with an extra sample holder. The laser profile is shaped using a HOLOEYE Phase-Only SLM. We probe the electron-light interaction either by energy filtering imaging (UTEM @ EPFL) or by imaging the momentum distribution of the electron beam in High Dispersion Diffraction (HDD) mode with a 100-m camera length on a direct electron detector (UTEM @ UNIMIB).

Results

We used the SLM to induce a Hermite-Gaussian (HG) shaping of the laser profile and, through energy filtering, we imaged at different focal planes only the electrons that interacted with light. By comparing the obtained data with simulations of an analytically modeled system (Fig. 2), we determined the coherence length δ of the electron beam to be larger than 50 nm. This is a crucial parameter that sets an upper limit to the laser phase profile that can be completely transferred to the electron beam for coherent electron modulation. Additionally, we have directly imaged the momentum distribution of the electron beam and probed the transverse-momentum exchanges at the PELM plane following the electron interactions with quanta of the IR photon beam. In these measurements, we have reached values of transverse coherence on the order of δ ~ 1 µm. By acquiring momentum images in HDD mode with a micrometric aperture at the image plane, we have obtained simultaneous momentum-and-space-resolved information at the PELM plane. Fig. 3 shows the light-modulated electron-beam profile. The inset illustrates the reconstruction of the SLM-modulated laser-beam profile at the PELM plane.

Conclusions

In conclusion, our findings demonstrate the potential of using shaped light fields in PINEM for enhanced tunability of electron modulation. The ability to control the electron wavefunction with such precision opens up new possibilities for the study of electron-light interactions. This could have far-reaching implications for various scientific domains, including life sciences, materials science, and nanotechnology. The rich dimensionality of light-electron interaction, spanning the transverse, longitudinal, temporal, and energy domains, harbors untapped potential and paves the way for future investigations in this dynamic field.

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Graphic:

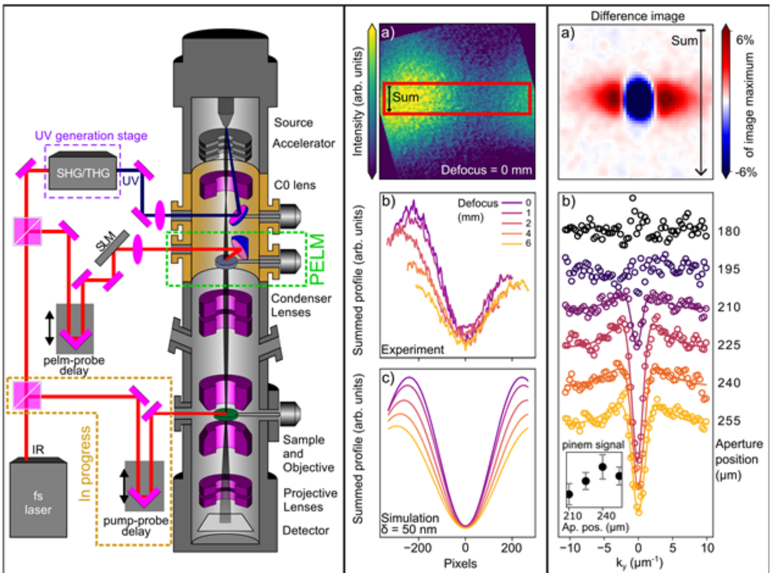

Fig. 1 | Ultrafast TEM setup at UniMiB. A pulse produced by a femtosecond IR laser is split in two. One pulse is converted into UV by 4th harmonic generation and it is directed to the TEM cathode to photoemit an electron pulse. The other pulse is syncronized with the electrons such that they reach the Photonic-based ELectron Modulator (PELM) at the same time and interact with each other. This light pulse is previously shaped with a Spatial Light Modulator (SLM) and it is used to dynamically shape the electron pulse. Currently, we are implementing a third laser branch to optically-excite transient states in a sample and to coherently control this excitations.

Fig. 2 | Energy-filtered PINEM with shaped light. a) Transverse profile of the HG-shaped electron-beam. b) Summed profiles obtained at different defoci. c) Simulations with an electron-beam transverse coherence patch δ of 50 nm. The minimum is preserved at different defoci, reproducing the experimental behaviour observed in panel b.

Fig. 3 | Momentum-resolved PINEM with shaped light. a) Diffence image of electron-beam momentum distribution at the PELM plane following light interaction. b) Summed profiles obtained with a 20-μm aperture placed at the sample plane at different positions (dots). Solid lines are least-square fits with two summed gaussians. Inset: variance of the positive fitted gaussian.

Keywords:

Photon-Induced Near-Field Electron Microscopy PINEM

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