Attosecond electron microscopy by free-electron homodyne detection

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Abstract: We implement attosecond electron microscopy in a transmission electron microscope to measure the optical near-field of a plasmonic nanoprism with 23 as (rms) temporal resolution and a few-nm spatial resolution. This demonstrates the concept of free-electron homodyne detection, where the quantum state of the electron wavefunction is reconstructed by a phase-controlled second interaction.

Photon-induced near-field electron microscopy (PINEM) enables the imaging of near-field intensities [1,2]. However, access to the evolution of nanoscale fields and structures within the light cycle requires a sensitivity to the optical phase [3,4].

Here, we introduce Free-Electron Homodyne Detection (FREHD) as a universally applicable approach to electron microscopy of phase-resolved optical responses at high spatiotemporal resolution [5]. In this scheme, a phase-controlled reference interaction (Fig. 1a) serves as the local oscillator to extract arbitrary sample-induced modulations of a free-electron wave function. We demonstrate this principle through the phase-resolved imaging of plasmonic fields of a gold nanoprism (Fig. 1c) with few-nanometer spatial and sub-cycle temporal resolutions. The near field at a sample modulates the phase of the electron wavefunction, and this wavefunction modulation is amplified or attenuated for in-phase and anti-phase reference interactions, respectively, which allows for a coherent read-out of a position-dependent phase (Fig. 1b). The characterization of both the amplitude and phase of the near-field allows to plot the electric field at different phases in a time-resolved movie (Fig. 1d).

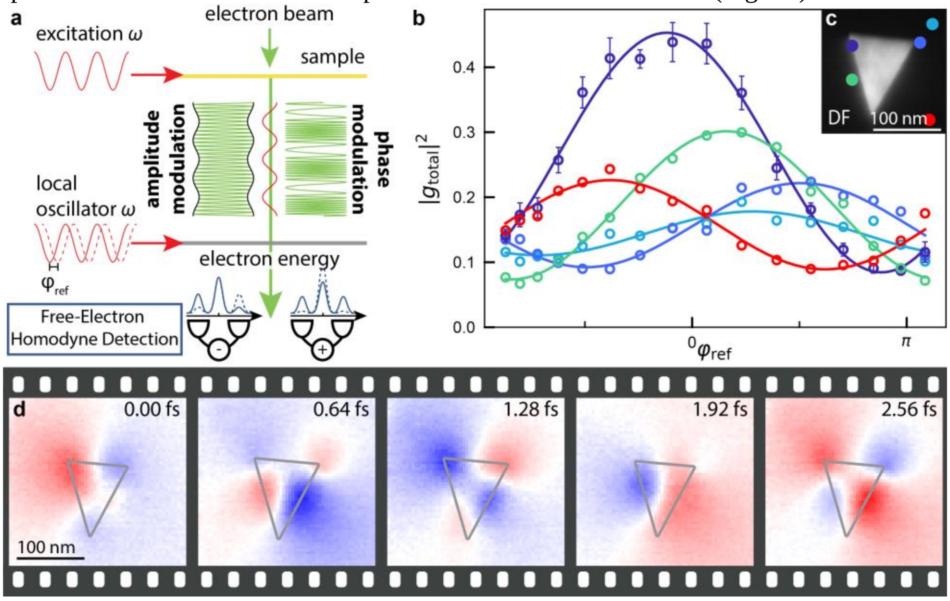

Fig. 1 **a** The principle of FREHD to resolve optically induced amplitude and phase modulation of the free-electron wavefunction by controlling the phase of a second reference interaction. **b** Interferograms of the total coupling coefficient at different positions of a gold nanoprism (dark-field image in **c**) allow to measure the amplitude and phase of the near-field at each pixel. **d** Time-dependent electric field (out-of-plane) at the gold nanoprism.

Free-Electron Homodyne Detection generalizes the high-resolution measurement of attosecond materials responses in electron microscopy, without a need for electron density bunching, and offers fascinating new possibilities to image local attosecond and phase-resolved responses on the nanometer scale.

References

[1] B. Barwick, D. J. Flannigan, and A. H. Zewail, "Photon-induced near-field electron microscopy," *Nature* **462**, 902–906 (2009).
[2] L. Piazza *et al.*, "Simultaneous observation of the quantization and the interference pattern of a plasmonic near-field," *Nat Commun* **6**, 6407 (2015).
[3] D. Nabben, J. Kuttruff, L. Stolz, A. Ryabov, and P. Baum, "Attosecond electron microscopy of sub-cycle optical dynamics," *Nature* **619**, 63-67 (2023).
[4] T. Bucher *et al.*, "Coherently amplified ultrafast imaging in a free-electron interferometer." arXiv:2305.04877 (2023).
[5] J. H. Gaida, H. Lourenço-Martins, M. Sivis, T. Rittmann, A. Feist, F. J. García de Abajo, and C. Ropers, "Attosecond Electron Microscopy by Free-Electron Homodyne Detection." *Nat Photon* (2024).