Holographic imaging of electromagnetic fields using electron-light quantum interference

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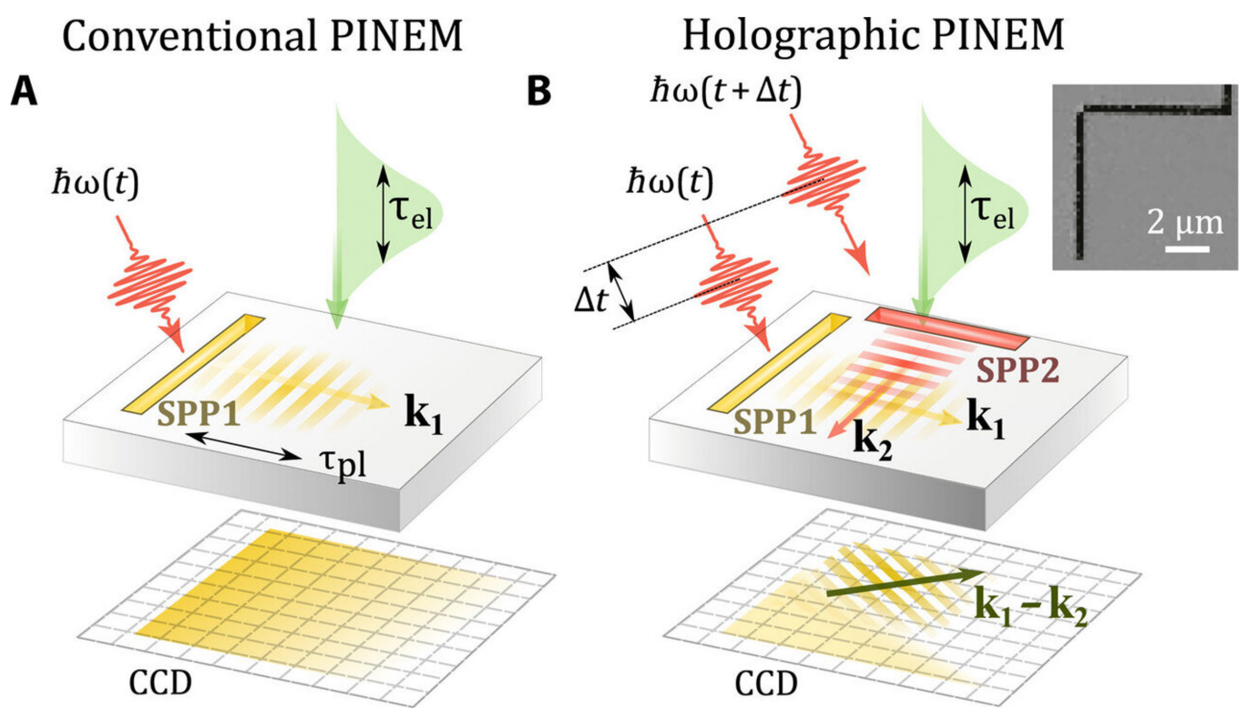
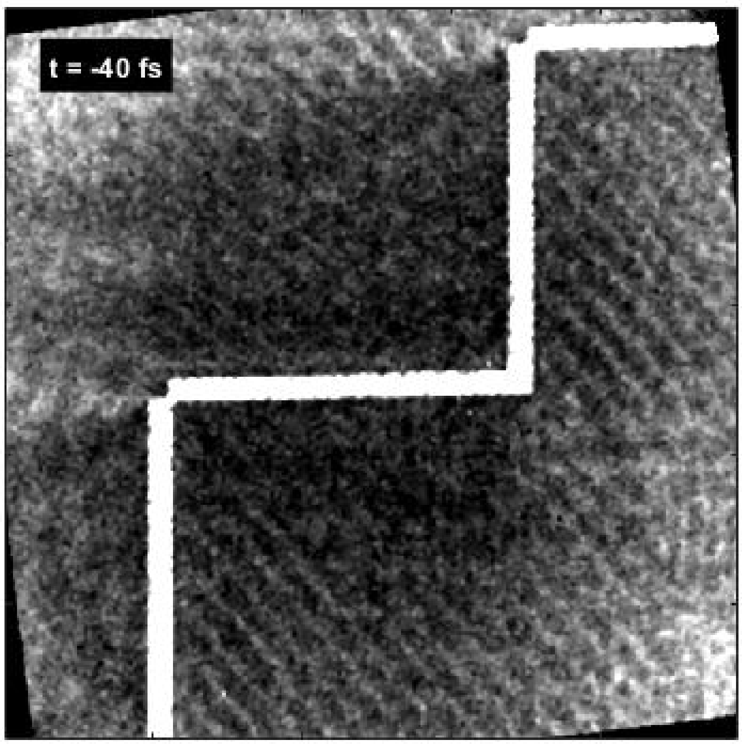


Conventional versus holographic PINEM imaging. (A) In conventional PINEM, propagating SPPs are imaged with long electron pulses, rendering only its time-averaged envelope with a spatial resolution Δx ~ τelvg. (B) In local holographic PINEM, two SPPs propagate with orthogonal wave vectors k1 and k2 forming a standing wave pattern along the direction k1 − k2, which is imaged as a periodic modulation in PINEM (the hologram). The interference contrast appears only when the two pulses overlap in space and time. Inset: SEM image of a fabricated structure. Black regions are grooves, which serve as plasmon sources. CCD, charge-coupled device. Credit: Science Advances, doi: 10.1126/sciadv.aav8358

In conventional holography a photographic film can record the interference pattern of monochromatic light scattered from the object to be imaged with a reference beam of un-scattered light. Scientists can then illuminate the developed image with a replica of the reference beam to create a virtual image of the original object. Holography was originally proposed by the physicist Dennis Gabor in 1948 to improve the resolution of an electron microscope, demonstrated using light optics. A hologram can be formed by capturing the phase and amplitude distribution of a signal by superimposing it with a known reference. The original concept was followed by holography with electrons, and after the invention of lasers optical holography became a popular technique for 3-D imaging macroscopic objects, information encryption and microscopy imaging.

However, extending holograms to the ultrafast domain currently remains a challenge with electrons, although developing the technique would allow the highest possible combined spatiotemporal resolution for advanced imaging applications in condensed matter physics. In a recent study now published in *Science Advances*, Ivan Madan and an interdisciplinary research team in the departments of Ultrafast Microscopy and Electron Scattering, Physics, Science and Technology in Switzerland, the U.K. and Spain, detailed the development of a hologram using local electromagnetic fields. The scientists obtained the electromagnetic holograms with combined attosecond/nanometer resolution in an ultrafast transmission electron microscope (UEM).

In the new method, the scientists relied on electromagnetic fields to split an electron wave function in a quantum coherent superposition of different energy states. The technique deviated from the conventional method, where the signal of interest and reference spatially separated and recombined to reconstruct the amplitude and phase of a signal of interest to subsequently form a hologram. The principle can be extended to any kind of detection configuration involving a periodic signal capable of undergoing interference, including sound waves, X-rays or femtosecond pulse waveforms.

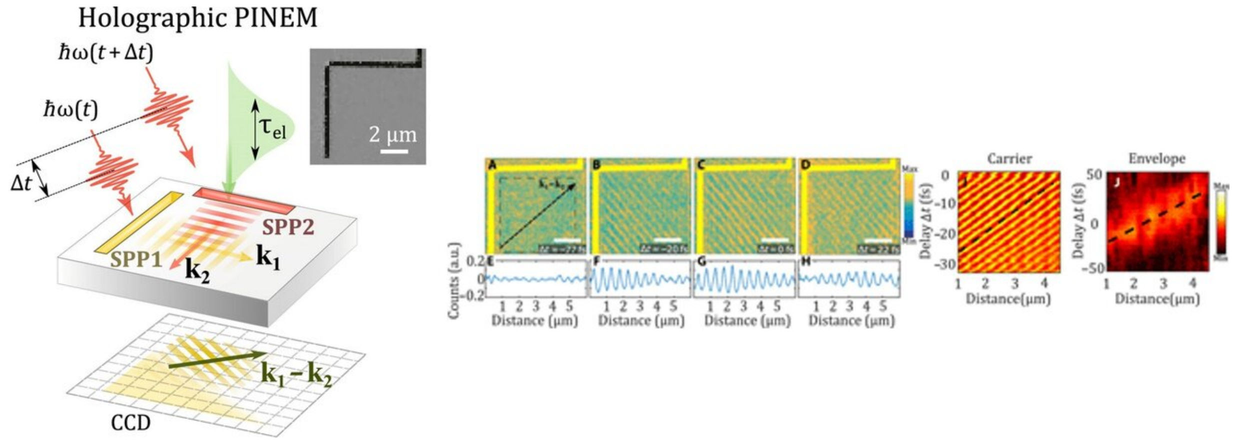


Plasmon hologram evolution with 0.33-fs time step. Credit: Science Advances, doi: 10.1126/sciadv.aav8358.

Further advances in the study of holography resulted in time-resolved optical holography, successfully realized in the femtosecond regime for enhanced spatial resolution in time-resolved photoemission electron spectroscopy (tr-PEEM). Reaching the ultrafast domain can also become a reality, due to recent developments in ultrafast transmission electron microscopy using femtosecond lasers to create ultrafast electron pulses. The developments have allowed real-time filming of collective electronic modes, strain fields and magnetic textures at a resolution of a few hundred femtoseconds.

In the new work, Madan et al. demonstrated a time-domain holography imaging technique in an ultrafast transmission electron microscope (UEM). They based the technique on quantum coherent interaction of electron wave packets with multiple optical fields. To illustrate the method, Madan et al. captured attosecond/nanometer-resolved phase-sensitive movies of rapidly evolving electromagnetic fields in plasmonic structures. The scientists implemented two key experimental methods in the study in an approach to parallelly access the quantum coherence of generic electronic states. The work will be relevant for further electron quantum optics applications.

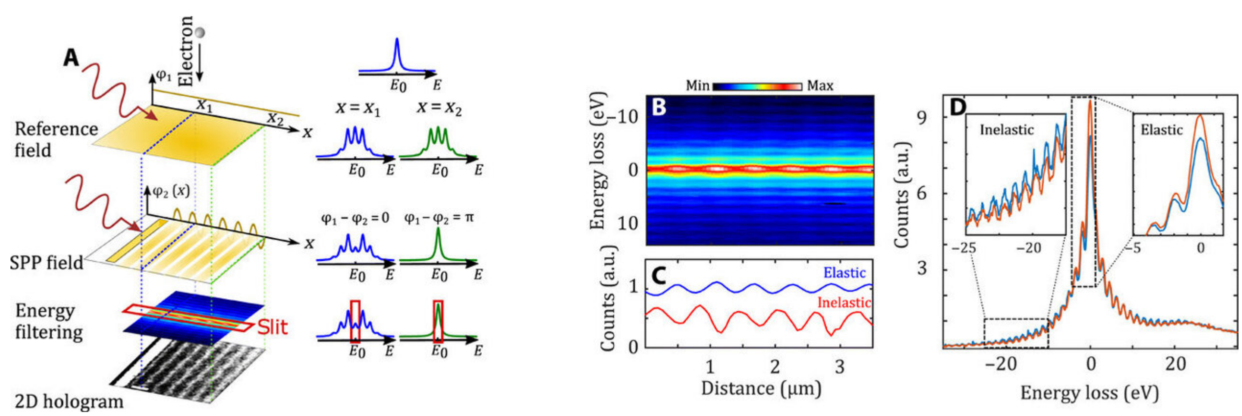
As a simple implementation of the holographic UEM, the scientists based the local interference of two fields on two propagating surface plasmon polaritons (SPPs) (i.e. a collective oscillation wave of free electrons along a metal). They described the interaction mechanism of the electron pulse with a single SPP using conventional photon-induced, near-field electron microscopy (PINEM) and then comparatively examined the holograms produced via the interference between two SPPs in a local holographic PINEM. During conventional PINEM, electrons can inelastically absorb or emit photon energy quanta and filter inelastically scattered electrons to allow the formation of real-space images of the plasmon fields.



LEFT: Schematic illustration of local holographic PINEM, where two SPPs propagate with orthogonal wave vectors k1 and k2 forming a standing wave pattern along the direction k1 − k2, which is imaged as a periodic modulation in PINEM (the hologram). The interference contrast appears only when the two pulses overlap in space and time. Inset: SEM image of a fabricated structure. RIGHT: Holographic images formed by two pulses of orthogonal polarization at different delays. (A to D) Micrographs of PINEM images for different values of the relative time delay Δt between the photo-exciting pulses, as indicated in each image. Scale bars, 2 μm. The SPP emitted from the vertical slit propagates from left to right. Correspondingly, the interference pattern moves from the bottom-left to the top-right corner. (E to H) Modulation of the electron counts along the k1 − k2 direction indicated in (A), calculated as the average of counts along the direction orthogonal to k1 − k2, taken within the dashed square indicated in (A). (I) Evolution of the profiles shown in (E) to (H) as a function of delay between the two pulses; because of the experimentally adopted sample orientation, retardation effects cause the slope of the fringes (see dashed line as a guide) to be decreased by a factor of 0.71 with respect to the SPP phase velocity. (J) Envelope of the interference pattern as a function of delay between the two pulses, with the slope of the peak (see dashed line as a guide) also decreased by a factor of 0.71 with respect to the SPP group velocity. Envelope data have been acquired in a separate measurement over a longer delay span and with larger time steps. a.u., arbitrary units. Credit: Science Advances, doi: 10.1126/sciadv.aav8358.

To implement the holographic PINEM concept, Madan et al. used an experimental nanostructure composed of two perpendicular slits, composed of silver (Ag) film fabricated by gallium (Ga) ion milling, deposited on a silicon nitride membrane (Si_3N_4). They conducted the experiments in a modified transmission electron microscope. In the work, the scientists used a second SPP wave as a reference and created an interference pattern with the SPP of interest to form a hologram when both waves overlapped in space and time. The scientists observed holograms formed by the 2 SPPs with relative pulse delays of -77, -20, 0 and 22 femtoseconds by energy filtering inelastically scattered electrons.

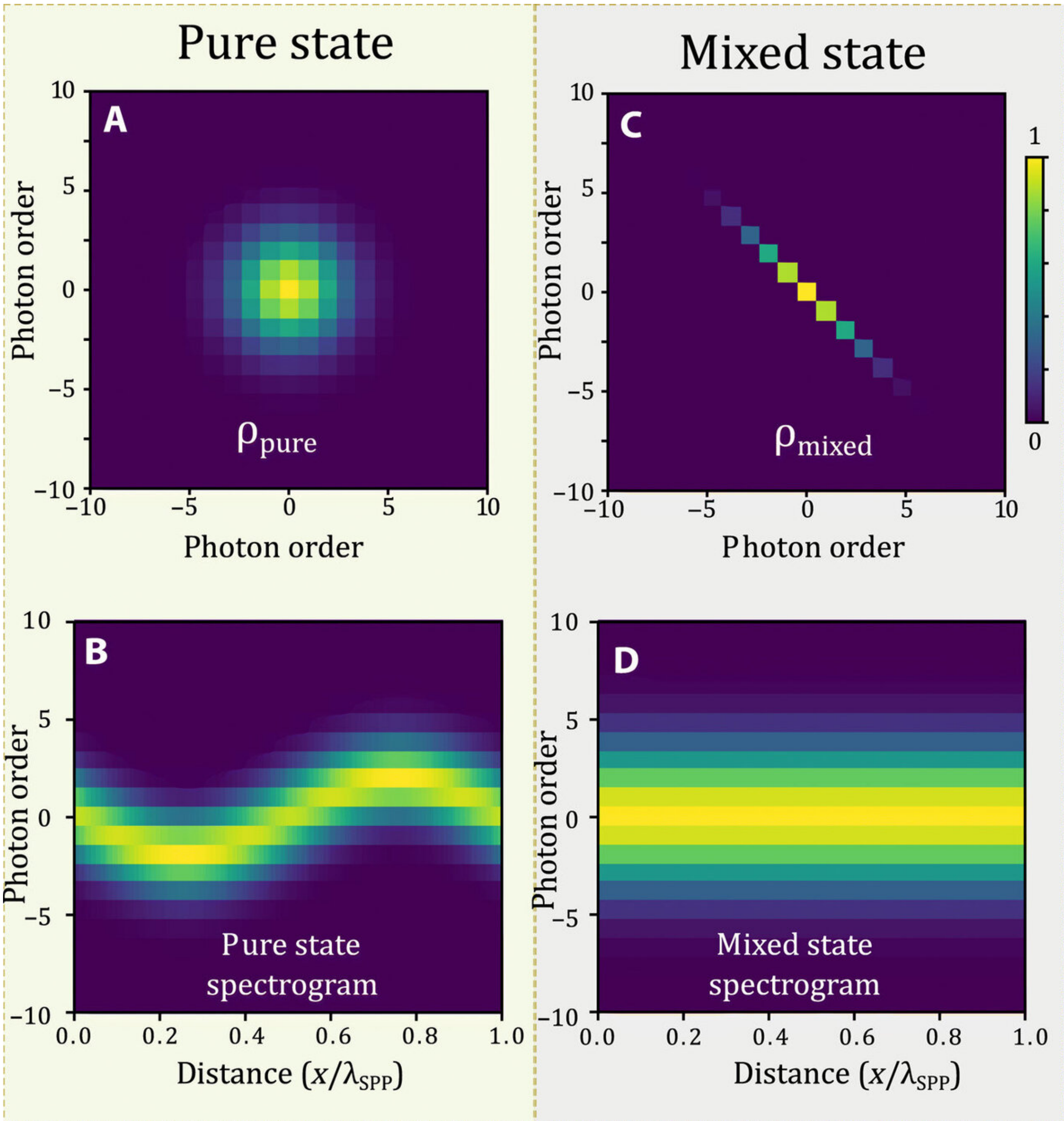
Madan et al. generalized the holographic approach using the coherence between different energy states of the quantum ladder, where the electron wave function split upon interacting with light. Since electrons carry information about the amplitude and phase of the optical field, even after completing the interaction, the scientists exploited this fact to enable quantum holography. In the experiments, they made use of a semi-infinite light field created by the reflection of the optical beam from an electron transparent optical mirror, to create a material-independent reference field. The setup allowed nearly constant spatial amplitude and phase to prepare an optimum reference field for holography in the study.



Principle of spatially separated electron holography. (A) The initial energy distribution of the electron beam is a function of energy that is singly peaked at E = E0 (right). Interaction with the reference field produces coherent superposition states with energies $E = E0 \pm n\hbar\omega$. The ensuing interaction with an SPP depends on the relative phase between SPP and reference fields, which results in a position-dependent electron energy distribution. The elastic part of the electron spectrum is then used to form the 2D hologram. The spectra on the right are simulations from an analytical model. (B) Hybrid energy-space map (spectrogram) of the electrons after interaction with the two fields, as schematized in (A). (C) Spatial profiles of the normalized intensity for elastic (blue curve) and inelastic (red curve) electrons, as obtained from (B) by energy-averaging from −1 to 1 eV for the elastic contribution and from −27 to −12 eV for the inelastic one. (D) Energy profiles at the maximum and minimum of the spatial modulation shown in (B), averaged over four periods. Credit: Science Advances, doi: 10.1126/sciadv.aav8358.

In the context of this study, quantum coherence of an electron state did not refer to the coherence between electrons, but to a measure of the monochromaticity (singularity) and phase stability of the electron plane wave. Madan et al. used the term to determine if an electron was in a pure state or entangled state in the environment. In the quantum sense, therefore, the phase between different energy states was determined by the time evolution operator and not at random.

The scientists then reconstructed the complex electric field distribution around 3-D particles or nanostructures. They showed that the mathematical equivalence of local plasmon holography and spatially separated quantum holography allowed the recorded holograms to be treated with the same formalism of propagating standing waves. Madan et al. thus presented an observation of this effect by recording holograms formed by the tilted wavefront of the light reflected from a silver mirror and a plasmon wave emitted from a hole carved in the silver layer. The resulting pattern exhibited a periodicity that was naturally absent from an untilted hologram.



Proposal for the determination of the coherence of photoemitted electrons. (A) Density matrix of a fully coherent (pure) state created by photoemission. (B) Spatially dependent spectrogram formed after interaction of the pure state with an SPP. (C) Density matrix of the completely mixed state. (D) Spectrogram formed after interaction of the mixed state with an SPP. Credit: Science Advances, doi: 10.1126/sciadv.aav8358.

Using model calculations, Madan et al. discriminated between a highly coherent (pure) and fully incoherent (completely mixed) electron distribution. For this, they modelled the density matrix of photoelectrons generated, for example, using UV illumination of a solid target. They then coordinated the electron states to interact with a traveling plasmon polariton in the experimental setup. By observing the electron energy distribution, the scientists were able to establish if there was partial coherence in the photoemitted electrons. Based on the observation, they proposed a further extension of the UEM holographic imaging to practically realize quantum holographic UEM. The scientists envision using the technique to study potential objects of interest such as atomic polarizabilities, excitons, phonons, Higgs and other collective and quasiparticle excitations in condensed matter systems in the future.

The present work provided sufficient information to reconstruct the complete density matrix of an unknown electronic state, similar to a previous approach on quantum state reconstruction with attosecond pulse trains. But unlike previous work, this method can also use well-controlled SPP fields to realize a number of projective measurements in parallel.

In this way Madan et al. demonstrated both local and spatially separated holographic approaches based on ultrafast transmission electron microscopy (UEM). The scientists showed that the nonlocal character of the technique allowed to completely decouple the reference and probe fields, which was not previously possible with near-field optical or photoemission microscopy techniques. The work offers a unique perspective to achieve atomic and sub-femtosecond combined resolution within a transmission microscope. The method will allow a spatially resolved detection method of coherences in electron quantum states with great potential for electron quantum holography and additional applications.

More information: I. Madan et al. Holographic imaging of electromagnetic fields via electron-light quantum interference, *Science Advances* (2019). DOI: 10.1126/sciadv.aav8358

Katharina E. Priebe et al. Attosecond electron pulse trains and quantum state reconstruction in ultrafast transmission electron microscopy, *Nature Photonics* (2017). DOI: 10.1038/s41566-017-0045-8

Xianglei Liu et al. Compressed ultrafast transmission electron microscopy: a simulation study, *Three-Dimensional and Multidimensional Microscopy: Image Acquisition and Processing XXVI* (2019). DOI: 10.1117/12.2510394

A new microscopic principle. www.ncbi.nlm.nih.gov/pubmed/18860291?dopt=Abstract Dennis Gabor et al. May 1948, *Nature*.