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Coherent Phase Control of Ultrashort Electron Pulses by Traveling Optical Waves and Whispering-gallery Modes

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The generation of high-coherence, ultrashort electron pulses and their manipulation by intense light fields promises new pathways in the active control of free-electron beams. Recent efforts that harness the temporal (longitudinal) coherence brought about a broad range of applications, including capabilities for imaging complex plasmonic fields [1,2], as well as efficient free-space acceleration [3] and attosecond bunching of electrons [4-6]. Relying on continuous electron beams and the ponderomotive potential in optical fields, a Zernicke phase plate was recently demonstrated [7] and elastic electron diffraction at standing light waves was observed in the Kapitza-Dirac effect [8]. However, precise optical phase control of free-electron wave packets in space and time remains challenging.

Here, we present two novel concepts for the coherent optical control of free-electron beams in an ultrafast transmission electron microscope (UTEM) [9,10], which rely on inelastic electron-light scattering [11,12].

The Göttingen UTEM instrument is based on a JEOL 2100F Schottky field emission TEM, which we modified to allow for optical sample excitation and the generation of photoelectron pulses (down to 0.8-nm focal spot size, 200-fs pulse duration, and 0.6-eV spectral bandwidth) from nanoscale field emitters [10]. The excellent coherence of the pulsed electron beam allows for investigating the optical manipulation of free-electron quantum states [4,13,14] by inelastic scattering in localized electromagnetic fields [1-2,4,11-14]. In this mechanism, the light field imprints a sinusoidal phase modulation onto the electron wavefunction, enabled by the broadened momentum spectrum of confined optical fields. As a consequence, the electron kinetic energy distribution evolves into a comb of spectral sidebands, separated by multiple quanta of the photon energy.

In the first experiment, we demonstrate the quantized transfer of optical energy and transverse momentum to a high-coherence electron beam [15]. Low-emittance ultrashort electron pulses are collimated to a micrometer-scale transverse coherence length and transmitted through a laser-illuminated graphite thin film (Fig. 1a). The imprinted three-dimensional sinusoidal phase modulation yields a coherent superposition of correlated energy-momentum ladder states, which is mapped by its far-field scattering distribution (Fig. 1b). Notably, this constitutes a coherent inelastic beam splitter for free-electron beams.

In a second study, we utilize optical whispering gallery modes (WGMs) traveling on the circumference of dielectric microsphere resonators to manipulate a traversing electron beam (Fig. 2a) [16]. The energy contained in femtosecond optical pulses is stored locally and we trace the associated cavity ringdown time (Fig. 2c). Moreover, the electron-light interaction is drastically enhanced by matching the phase velocity of the WGM to the velocity of 200-keV electrons, achieving a spectral bandwidth of up to 700 eV in a single interaction (Fig. 2d).

In conclusion, we demonstrate the coherent control of free-electron beams by spatially structured or cavity-enhanced optical phase modulation. We envision a new class of active electron-optical elements for TEM, that combine these capabilities with the flexibility of state-of-the-art integrated photonics, e.g. for laser-based electron beam splitters and programmable phase plates.

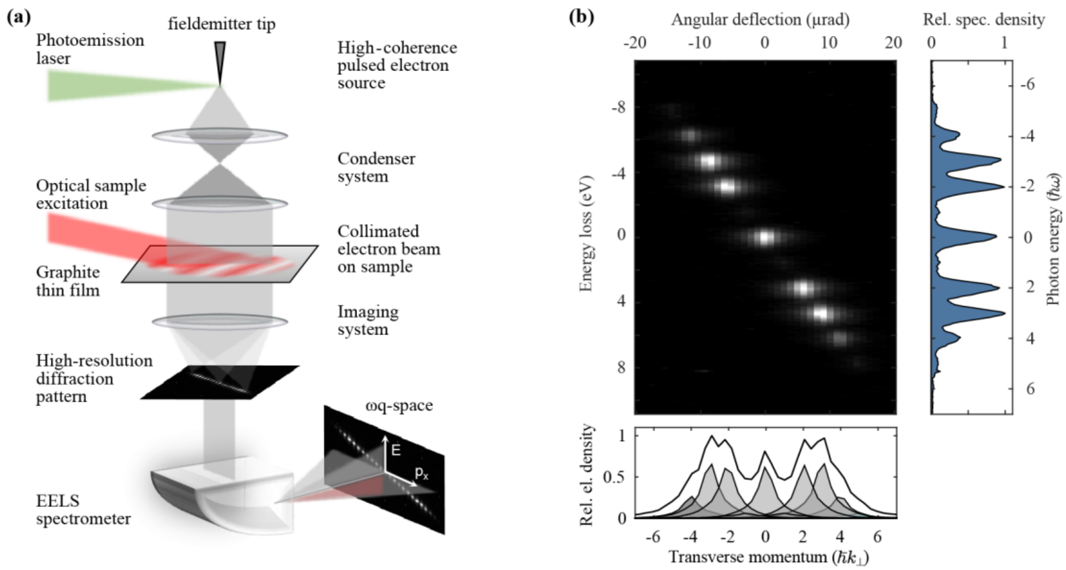

Figure 1. Transverse momentum resolved inelastic electron light scattering. (a) Inside of an ultrafast transmission electron microscope, photo-emitted ultrashort electron pulses are collimated and transmitted through a laser side-illuminated graphite thin film. The resulting optically phase-modulated electron beam is analyzed in terms of its distributions in reciprocal (transverse) and in energy (longitudinal) space. (b) Far-field electron scattering distribution resolved in energy and transverse momentum space (oriented along the optical plane of incidence) with normalized orthogonal projections.

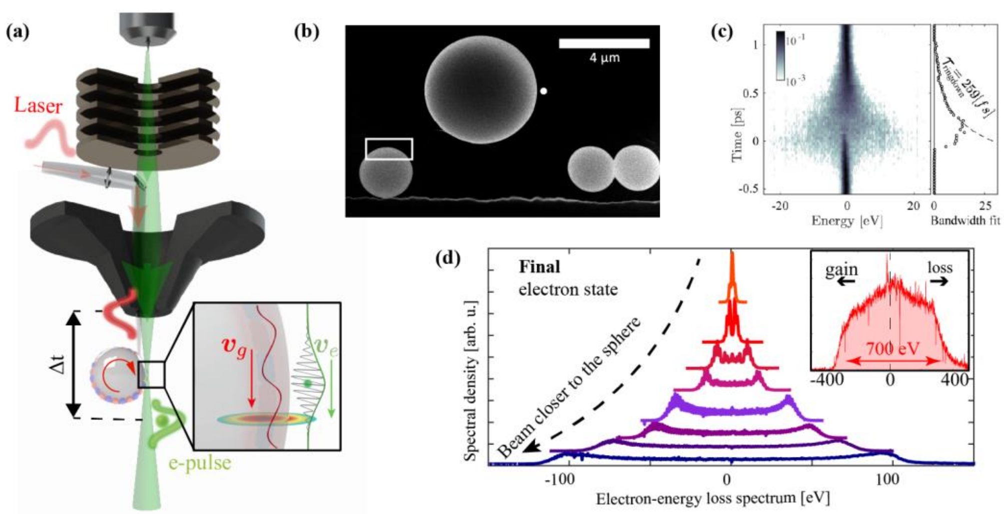

Figure 2. Enhanced electron-light interaction by whispering-gallery modes (WGMs) in an optical microsphere resonator. (a) An ultrashort electron pulse (green) is probing a femtosecond laser-excited (red) microsphere, with externally controllable time delay. After passing the nearfield of the microsphere, the electron beam is analyzed in an electron-energy loss spectrometer (EELS). The matching of the electron velocity to the WGM phase velocity, namely, the phase matching condition, allows the electron and the WGM to exchange multiple photon-energy quanta. (b) STEM image of dielectric microsphere resonators. (c) Colormap of the electron spectrum (log-scale) following a loading of WGMs by a 50-fs

optical pulse at time t = 0. At later times, the electron phase modulation is driven by light stored in the cavity, with a storage time of 260 fs, corresponding to a quality factor Q = 97. (d) For decreasing distance to the microsphere (white rectangle in (b)), the spectral width increases. We achieve up to 700 eV bandwidth for the most efficient coupling conditions, with the optical field reaching ~1.4 GV/m.

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