Attosecond Electron Microscopy

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Abstract: We report the advance of transmission electron microscopy to attosecond time resolution and show selected experimental results.

The primary step of almost any light-matter interaction is the electrodynamic response of the electrons in a material to the optical cycles of the impinging light wave on sub-wavelength and sub-cycle scales. To see such dynamics in space and time, we report here the advance of transmission electron microscopy to attosecond time resolution [1], combining the awesome spatial resolution of a transmission electron microscope with the awesome time resolution that is offered by the cycles of laser light. The idea is to use the oscillations of a laser wave to modulate the electron beam into a rapid sequence of electron pulses and then use an energy filter to resolve the electromagnetic near-fields in and around a specimen as a movie in space and time [1].

In the experiment, we use an electron microscope with Schottky field emitter at an electron energy of 183 keV (Fig. 1a). Continuous-wave laser light with a wavelength of $\lambda = 1064$ nm and a cycle period of 3.6 fs (red) is used to excite the material under investigation (black). The same laser light (red) also modulates the electron beam (blue) into a train of attosecond pulses by periodically accelerating and decelerating the freeelectron wave function with the optical electric fields [2]. Here, in order to conserve energy and momentum, an ultrathin dielectric modulation membrane (yellow) delivers the necessary optical asymmetry.

At the specimen, the compressed attosecond electron pulses at a speed of $v_e \approx 0.67$ c cover hundreds of nanometers in sub-femtosecond times, and electromagnetic near-fields in and around the specimen therefore appear approximately frozen in time. The longitudinal fields shift the energy spectrum of the attosecond electrons coherently towards a larger or smaller central energy and the positive or negative energy



Figure 1. Attosecond electron microscopy. (a) Concept and experiment to see optical phenomena as a movie in space and time. (b) Field-cycle-contrast results at the example of a tungsten needle tip. Scale bar, 500 nm.

sidebands therefore become more or less intense, depending on position and arrival time. An energy filter (90 $^{\circ}$ curve) then selects only such electrons that have gained energy and thereby produces a movie of the optical cycles as a function of space and time.

Figure 1b depicts one of our early results [1]. The specimen is a tungsten needle that is laser-excited from \sim 45°. The needle tip converts this excitation wave in part into a near-field surface wave that propagates along the tip and shaft. Indeed, the energy-filtered electron microscopy images reveal around the needle's surface a set of local regions with electron energy gain (white) and energy loss (black) as a function of time on attosecond dimensions. More results on chiral materials, inverse nanoantenna slits, metamaterials and chemically prepared mesocrystals show the general applicability of our novel attosecond electron microscopy to a large range of questions in contemporary research. These results establish attosecond transmission electron microscopy with field-cycle contrast as a versatile and sensitive method for visualizing the dynamics of light-matter interaction in complex materials on fundamental dimensions in space and time.

- [1] D. Nabben, J. Kuttruff, L. Stolz, A. Ryabov and P. Baum, Nature 619, 63-67 (2023).
- [2] A. Ryabov, J. W. Thurner, D. Nabben, M. V. Tsarev and P. Baum, Sci. Adv. 6, eabb1393 (2020).