

# Microscopy and diffraction with attosecond electron pulse trains

Yuya Morimoto<sup>1,2,\*</sup> and Peter Baum<sup>1,2,3,\*</sup>

<sup>1</sup>Ludwig-Maximilians-Universität München, Am Coulombwall 1, 85748, Garching, Germany

<sup>2</sup>Max-Planck-Institute of Quantum Optics, Hans-Kopfermann-Straße 1, 85748, Garching, Germany

<sup>3</sup>Universität Konstanz, Universitätsstraße 10, 78464 Konstanz, Germany

**Abstract.** Attosecond imaging with electron beams can access optical-field-driven electron dynamics in space and time. Here we report first diffraction and microscopy experiments with attosecond electron pulses. We study attosecond-level timing of Bragg-spot emission and visualize light-wave propagation in space and time.

## 1 Introduction

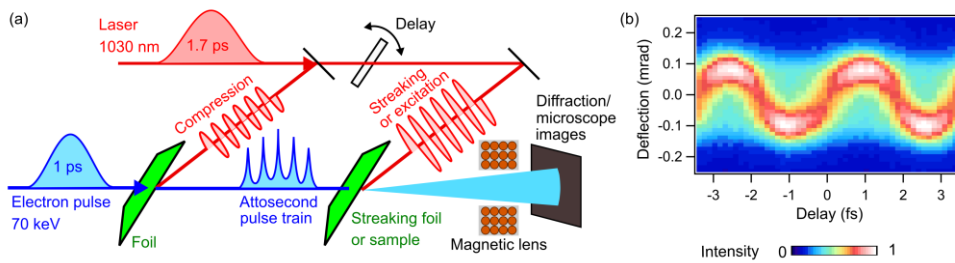
Attosecond optical spectroscopy allows to monitor electronic dynamics with unprecedented temporal resolution. However, it is still challenging to obtain a space-time visualization of fundamental charge/carrier dynamics, because the wavelength of attosecond optical pulses in the extreme-ultraviolet is about one-hundred times longer than atomic distances. In contrast, ultrafast electron microscopy and diffraction can provide nanometer and Angstrom spatial resolutions, but only with femtosecond temporal resolutions. In this work, we report the advanced ultrafast electron diffraction and microscopy with attosecond resolution [1]. We generate trains of attosecond electron pulses by interaction of an electron beam with the cycles of laser light. With these pulses, we report proof-of-principle attosecond diffraction and microscopy experiments that establish the feasibility and future potential of attosecond-Angstrom imaging, which allows to directly visualize fundamental light-matter interactions on their natural dimensions.

## 2 Concept and experiment

Figure 1(a) shows our concept for attosecond electron imaging [1,2]. A picosecond electron pulse (blue) at sub-relativistic energy (70 keV) passes through a free-standing dielectric foil (green) that is excited by a laser field (red). Although the dielectric foil is transparent to the laser, there is efficient momentum transfer to electrons because of a phase shift between the incoming and outgoing light waves due to the refractive index of the foil [1,2]. Longitudinal momentum gain/loss depends on the arrival time of an electron with respect to the optical cycles and therefore converts the incoming electron wavepacket into a train of

---

\* Corresponding authors: Yuya.Morimoto@lmu.de, peter.baum@uni-konstanz.de



**Fig. 1.** Attosecond electron pulse trains and applications. (a) Attosecond electron-beam control and microscopy/diffraction demonstrations. (b) Streaking characterization of attosecond electron pulses.

attosecond pulses [1-5]. Compared to metallic foils or nanostructures, dielectric membranes have higher laser damage thresholds [6] and thus can provide a higher degree of control. Also, it is possible to compress a collimated beam [1,2]. In a second stage (green), the attosecond electron pulses probe the dynamics of a sample that is driven by the cycles of another laser field (red). Here, for example, the temporal structure of the compressed attosecond electron pulses can be characterized by sideways deflection like in a cathode-ray oscilloscope [1,2,7]. Alternatively or simultaneously, we can also access sub-cycle dynamics of samples via Bragg diffraction or real-space microscopy (see below).

### 3 Attosecond electron pulses: generation and characterization

In a first experiment, we compressed a mono-energetic 70-keV electron beam by a near-infrared (1030 nm) picosecond laser field impinging on a 50-nm-thin  $\text{Si}_3\text{N}_4$  membrane. For characterization of the compressed electron pulses, we placed a second membrane, a 60-nm-thin silicon foil at 4 mm distance and excited it with another near-infrared laser field. The streaked electron beam profile was recorded by a single-electron detector. Figure 1(b) shows the observed streaking signals. Note that this streaking is not in the energy domain like in XUV spectroscopy but rather in real-space like in an oscilloscope [7]. The localized spot moves up and down with the sub-optical-cycle delay time between the electron pulse train and the streaking field. This oscillation directly demonstrates the successful compression of electron pulses to attosecond duration and the phase-locked nature of the sample excitation. Through fitting of entire streaking signals, we obtain a pulse duration of 350 as (rms) or 820 as (FWHM) above the  $\sim 30\%$  background [1,2].

### 4 Attosecond electron diffraction and microscopy

In two more experiments, we show that our attosecond electron pulses at sub-relativistic energies are suitable for atomic diffraction and sub-light-cycle real-space microscopy. First, in Fig 2(a), we show Bragg reflections from a single-crystal silicon membrane recorded with the attosecond electron pulses. We observed tens of Bragg spots up to (606) which directly demonstrate the Angstrom imaging capability with our attosecond pulses in diffraction mode. Attosecond streaking of each Bragg spot like in the pulse characterization experiment reported above reveals the potential delay of Bragg-diffracted electrons with respect to the direct beam (000). The results are summarized in Fig. 2(b), showing that the electron scattering process from the direct beam into the Bragg spots in the Si single crystal takes less than 40 as [1]. There is also a time-dependent intensity change of the Bragg spots related to charge motions in the unit cell [8] that could be revealed at better experimental conditions [1].

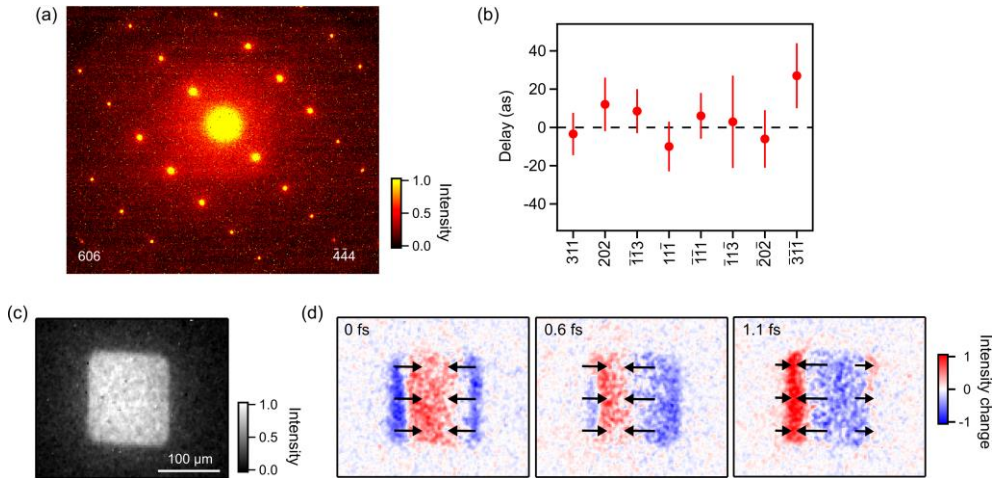


Fig. 2. Attosecond electron diffraction and microscopy. (a) Bragg diffraction of a single-crystal silicon membrane measured with attosecond electron pulses. (b) Attosecond delay of Bragg spots with respect to the direct beam. (c) Microscopic shadow image of the silicon sample. (d) Delay-dependent image changes. Black arrows represent direction of streaking fields.

We also report real-space electron microscopy with our attosecond pulses by advancing electromagnetic waveform electron microscopy [9] to attosecond resolution, measuring electromagnetic field vector dynamics at petahertz frequency. In the experiment, we record transmission images of a silicon foil (Fig. 2(c)) which is excited by the cycles of laser light at a slightly inclined angle of incidence. This configuration generated a travelling light wave at the silicon interface. Figure 2(d) shows the delay-dependent changes of the transmission electron microscopy images measured with the attosecond electron pulses. The spatial resolution was limited by our beam quality to the micrometer regime but still sufficient to see sub-cycle dynamics in space. We see pronounced intensity variations that are caused by time-frozen deflections, which are in turn directly related to the local and instantaneous optical waveform. Parts in the images where the electron counts are higher (red) indicate a spatially converging instantaneous optical force (see black arrows in Fig. 2(d)) while the less intense parts (blue) indicate a diverging deflection. The entire pattern rapidly circulates from right to left when varying the attosecond delay. This observation reveals the propagation of the optical wave on the nanometer foil, resolved with sub-wavelength resolution in space and with attosecond resolution in time.

## References

1. Y. Morimoto and P. Baum, *Nat. Phys.* **14**, 252 (2018)
2. Y. Morimoto and P. Baum, *Phys. Rev. A* **97**, 033815 (2018)
3. C. M. S. Sears et al., *Phys. Rev. ST Accel. Beams* **11**, 061301(2008)
4. K. E. Priebe et al., *Nat. Photon.* **11**, 793 (2017)
5. M. Kozák, N. Schöenberger, P. Hommelhoff, *Phys. Rev. Lett.* **120**, 103203 (2018)
6. Y. Morimoto et al., *J. Appl. Phys.* **122**, 215303 (2017)
7. C. Kealhofer et al., *Science* **352**, 429 (2016)
8. V. S. Yakovlev et al., *Sci. Rep.* **5**, 14581 (2015)
9. A. Ryabov and P. Baum, *Science* **353**, 374 (2016)