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Attosecond Electron Pulse Trains and Applications to Time-Resolved Diffraction and Microscopy

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ABSTRACT

Attosecond spectroscopy with laser-generated photons can in principle resolve electronic processes in real time, but a movie-like space–time imaging is impeded by the wavelength, which is ~100 times longer than atomic distances. Here we advance attosecond science to sub-atomic spatial resolution by using sub-relativistic electron beams instead of the high-harmonic photons. A beam of 70-keV electrons at 4.5-pm de Broglie wavelength is temporally modulated by the electric field of laser cycles into a train of attosecond pulses with the help of a dielectric modulation element. The pulses in the train have 820-as duration and maintain the degree of coherence of the original electron beam. We demonstrate the feasibility of analytic attosecond–Angstrom imaging by recording time-resolved Bragg diffraction from a single-crystalline silicon. Real-space electron microscopy with the attosecond electron pulses visualizes the propagation of optical waves at a dielectric membrane with sub-wavelength and sub-optical-cycle resolution. This unification of attosecond science with electron diffraction/microscopy will enable the direct visualization of fundamental and complex light-matter interaction in space and time.

Keywords: Attosecond physics, attosecond electron pulse, attosecond electron microscopy, attosecond electron diffraction, electron pulse compression, attosecond streaking, sub-cycle electron microscopy, light-matter interaction

1. INTRODUCTION

Almost any light-matter interaction begins with light-driven electronic motions on sub-cycle time scales and on length scales of nanometers to angstroms. Visualizing such electronic motions in space and time therefore requires attosecond and nanometer/Angstrom resolutions. Attosecond spectroscopy using the high harmonic sources continues to reveal the electronic dynamics with unprecedented temporal resolutions. However, it is still challenging to obtain a movie-like space-time visualization because the wavelength of attosecond photon pulses is typically one-hundred times longer than atomic distances. On the other hand, electron microscopy and diffraction with keV-electron beams can easily provide nanometer and picometer resolutions, but the temporal resolution is only tens of femtoseconds or above, limited by space charge and dispersion effects. In this work, we advance electron microscopy and diffraction to attosecond resolution by generating attosecond electron pulses[1-5] with the help of a dielectric free-standing membrane.

2. ATTOSECOND CONTROL OF ELECTRON BEAMS AT DIELECTRIC MEMBRANES

Figure 1 illustrates the concept of attosecond electron-beam control for the generation and characterization of attosecond electron pulses. If electron and laser beams would cross in free space (i.e. without the membrane shown in green), the periodic oscillations of electron momentum driven by the laser field cycles cancel out after the laser has passed (black dotted curve in Fig. 1(b)). Therefore, in free space, the electron does not obtain any net momentum from the laser field, with an exception of ponderomotive effects, which are weak. In contrast, if the laser is impinged on a dielectric foil (Fig. 1(a)), which is transparent to both electron and the laser beams, there occurs a phase shift between the incoming and outgoing electromagnetic waves due to the foil's refractive index in combination with the thin-film interferences. This phase shift breaks the periodicity of the cycle-driven momentum oscillations and therefore causes an overall net momentum change $\Delta \boldsymbol{p}$ after the interaction (red curve in Fig. 1(b)). The final amount of momentum change is periodic with the laser frequency[1,2]. The change along the forward direction, i.e. acceleration and deceleration, can compress incoming electrons into ultrashort pulses after subsequent propagation in free space[6,7]. The sideways momentum

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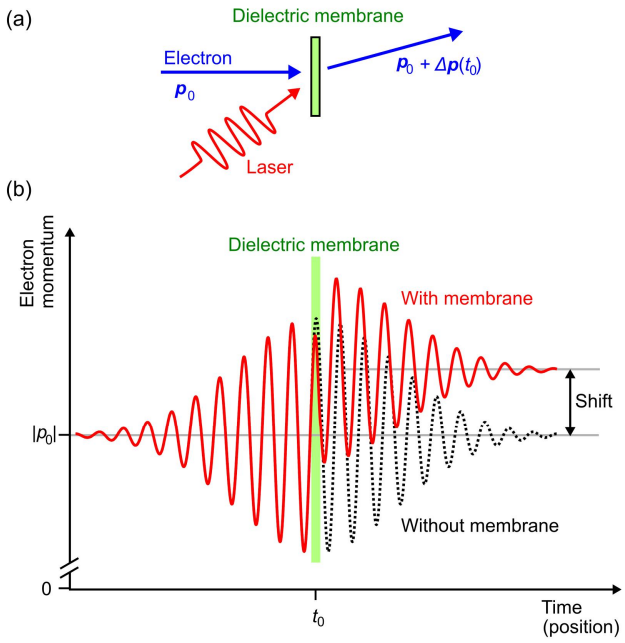

Figure 1. Attosecond control of electron beams at dielectric free-standing membranes. (a) Schematic of attosecond control of electron beams (blue) with dielectric membranes (green). (b) Mechanism of the momentum exchange mediated by the dielectric foil. Without a foil, electron does not gain momentum (black dotted curve). In the presence of a foil, electron momentum can be shifted after interaction (red curve). $\boldsymbol{p}_0$ represents the initial momentum. $\Delta\boldsymbol{p}$ is the momentum shift which depends on the laser-electron timing t_0.

contribution causes time-dependent streaking like in a cathode-ray oscilloscope but now at the ultrahigh laser frequency (petahertz). Unlike absorbing foils[4], metallic foils[7] or nanostructures, dielectric membranes have negligible linear absorption at optical frequencies and can therefore sustain at an extreme level of field strengths[8], which enables a high degree of electron pulse compression and efficient streaking metrology[2].

3. ATTOSECOND ELECTRON PULSE TRAINS

Figure 2(a) depicts our experimental setup for attosecond electron pulse generation and characterization. A mono-energetic 70-keV (4.5-pm wavelength) electron pulse is produced by the photoemission and subsequent acceleration by a dc electric field. The electron pulse passes through a 50-nm Si_3N_4 foil which is excited by a laser beam of 1030-nm wavelength. At the foil, the electron is periodically accelerated and decelerated by the laser cycles and then forms a train of attosecond electron pulses after the propagation of ~4 mm. The temporal structure of the attosecond electron pulses is characterized by the sideways deflection at the second stage (60-nm Si foil).

Figure 2(b) left panel shows the observed streaking signals. The localized spot moves up and down with the sub-femtosecond delay between the streaking field cycles and the compressed electron pulses. This oscillation directly demonstrates the successful compression of electron pulses to attosecond durations. We obtain a pulse duration through a global fitting of the entire 2D signal[1,2] (see Fig. 2(b) right panel). Figure 2(c) plots the measured pulse durations as a function of the compression field strength. At 60 MV/m, we obtain the shortest pulse duration of 350 as (rms) or 820 as (FWHM) above a ~30% background, as shown in Fig. 2(d).

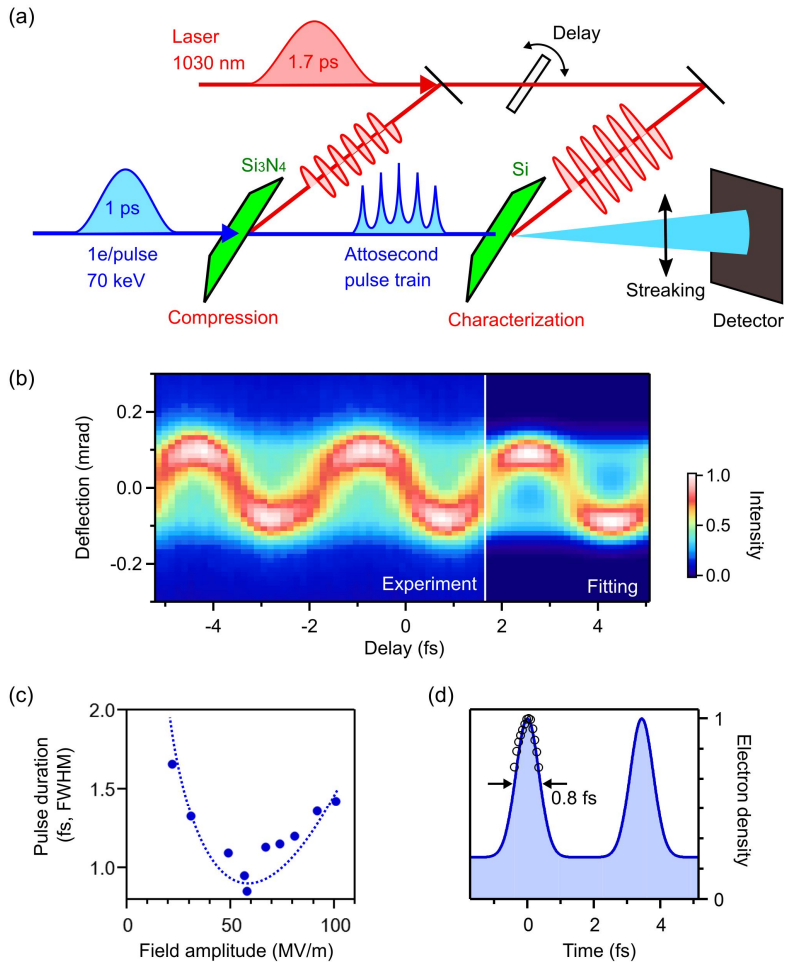

Figure 2. Attosecond electron pulses. (a) Concept and experiment of attosecond electron pulse generation and characterization. (b) Observed streaking signal (left panel) with a result of the global fitting (right panel). (c) Pulse duration of the compressed pulses as a function of the compression field strength. Dotted curve is for eye guiding. (d) Obtained electron pulse shape. Circles plot a vertical cut of the streaking signal in the left panel of (b) at 0 fs delay time[1,2].

4. ATTOSECOND BRAGG DIFFRACTION

In the following we report two proof-of-principle experiments which show that attosecond electron pulses at sub-relativistic energies[1,2,4,5] are indeed suitable for time-resolved microscopy and diffractive imaging with attosecond resolution. First, we report ultrafast Bragg diffraction with the attosecond electron pulses. Figure 3(a) shows the measured Bragg spots from a single-crystal silicon foil. We observe a large set of diffraction spots up to Miller indices of (606) and (444), demonstrating the atomic resolution. With an integration of a few minutes, we obtain a signal-to-noise ratio of 100 which is high enough to perform diffractive imaging of atomic-scale charge carrier dynamics that is driven by light cycles[9-12].

When the crystalline silicon sample is excited by the laser field, Bragg diffraction and sideways deflection occur in synchrony. Each Bragg spot is streaked by laser cycles in a similar way as the direct beam (000-spot). Figure 3(b) shows an example of such a streaking signal. A center-of-mass analysis of each streaking trace reveals the potential

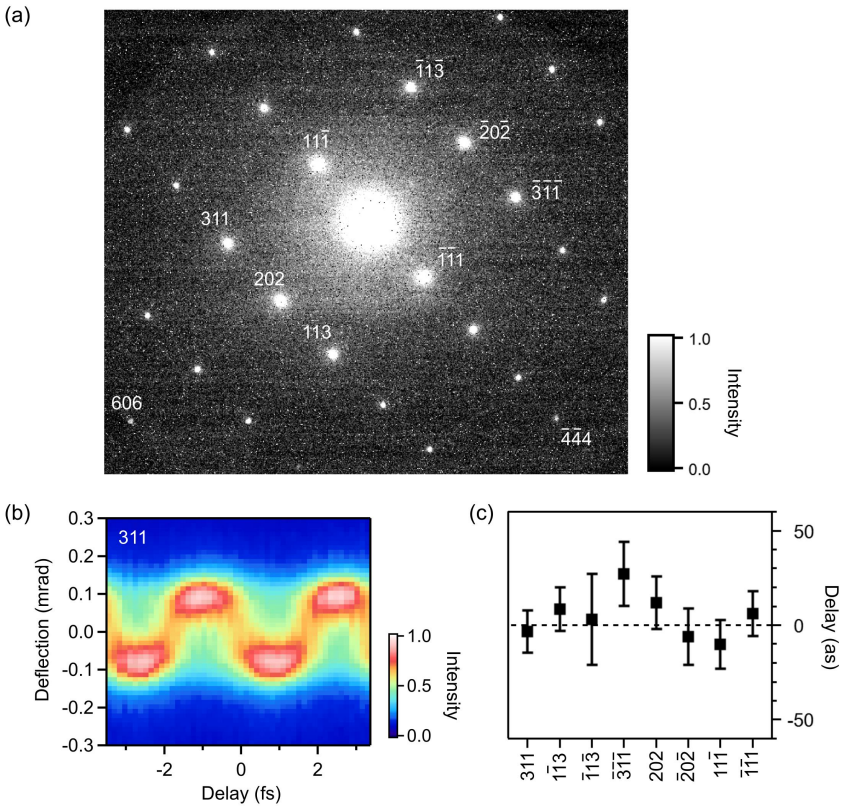

Figure 3. Attosecond electron diffraction. (a) Bragg diffraction of a single-crystal silicon foil measured with the attosecond electron pulses. (b) Observed attosecond streaking signal of the (311) spot. (c) Measured attosecond-level delays between Bragg diffracted electrons and the direct beam (000 spot).

delay between a Bragg spot and the direct beam. Figure 3(c) summarizes the results. We find that the scatterings of sub-relativistic electrons in nanometer-thick solids take less than tens of attoseconds.

5. SUB-CYCLLE ELECTRON MICROSCOPY

The second proof-of-principle experiment is about attosecond electron microscopy. In principle, waveform electron microscopy[13] can provide electromagnetic field vectors in space and time, but the demonstrated resolutions have been tens of femtoseconds. Here we advance waveform microscopy to attosecond resolution. Figure 4(a) illustrates the imaging geometry. We record transmission images of a nanometer-thick foil excited by laser cycles that impinge from a slightly off-angle direction. This configuration generates a travelling optical wave at the foil interface. The attosecond electron pulses are instantaneously deflected at each sample location because their passage through the sample is shorter than an optical period[13]. Figure 4(b) shows a transmission image of the silicon foil measured with the attosecond electron pulses without laser excitation. Figure 4(c) shows the delay-dependent changes of that image. The entire pattern moves rapidly along the vertical direction when varying the attosecond-level delay between the excitation and the electron pulse train, hence revealing the space-time propagation of the optical wave. The intensity variations at each delay time are directly related to the local and instantaneous optical fields. The parts with higher signal intensity (red) indicate such locations where the instantaneous electromagnetic deflection angles are convergent. In contrast, the less intense parts (blue) represent a diverging deflection. These observations directly reveal the propagating optical waves, resolved on attosecond time scales.

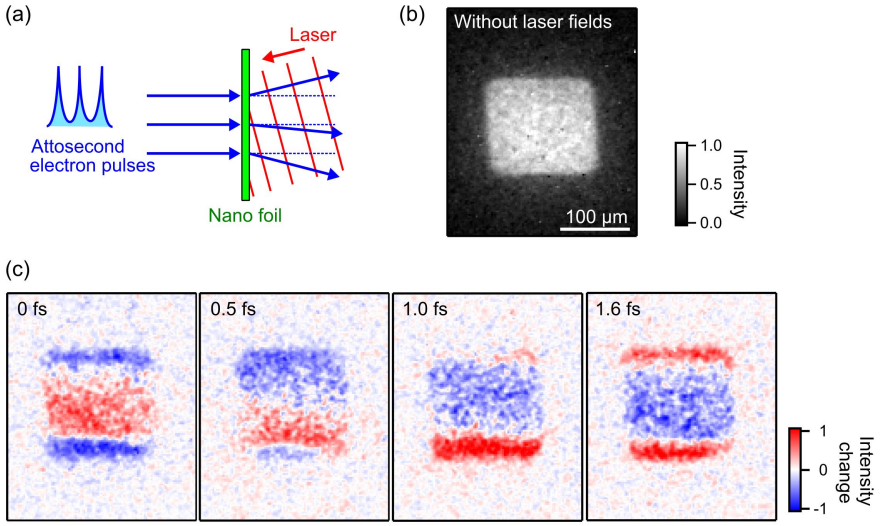

Figure 4. Attosecond electron microscope of electromagnetic waveforms. (a) Sketch of the experiment. (b) Transmission electron microscope image of the silicon foil without laser excitations. (c) Delay-dependent image changes due to the local and instantaneous deflecting fields at the foil.

6. SUMMARY AND OUTLOOK

In summary, we unified attosecond science with the atomic-resolution capabilities of keV electron beams such as used in table-top diffraction experiments or in commercial electron microscopy devices. Attosecond electron pulses are simple to generate, versatile to apply, and therefore should become a valuable alternative to high-harmonic photon sources for understanding basic light-matter interaction. The first attosecond electron diffraction and microscopy concept demonstrated in this work[1] allows to investigate fundamental light-field-induced dynamics such as field-driven charge dispacements, tunnel/multiphoton/Auger photoionization or linear/nonlinear plasmonics on their natural space-time dimensions.

7. ACKNOWLEDGEMENTS

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