Ultrafast electron diffractometer with Terahertz-driven pulse compression

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Abstract: We built an ultrafast electron diffractometer, which utilizes a Terahertz driven pulse compressor to probe the ultrafast dynamics of single-crystal silicon. We demonstrate high-quality diffraction with improved time resolution.

Introduction

Ultrafast electron sources have emerged as a powerful tool for revealing structural dynamics in molecules and materials at the atomic scale [1]. Such sources allow to capture the atomic structure of matter at an instant in time and provide structural information on nonequilibrium states of matter. Over the past years, there has been great interest in achieving sub-100 femtosecond (fs) time resolution with sufficient brightness, and high repetition rate to enable a direct observation of primary events governing physical/chemical processes [1].

Recently, it has been shown that laser-based Terahertz (THz) radiation powered electron acceleration and manipulation provides a promising solution to construct future ultrafast electron sources that support high energy, high repetition rate, short electron bunches while being compact [2–4]. THz radiation at millimeter wavelengths has been proven to enable GV/m field strength [5] which is well-suited for sub-picosecond electron beam manipulation. THz driven electron manipulation can be used to compress electron bunches to sub-100 fs duration. Combining the advantages of a compact conventional photo-triggered DC electron gun with the inherent synchronization provided by laser-based approaches makes it a promising hybrid solution for generating compact electron sources with high repetition rate and short bunch duration.

Here, we demonstrate the first application of a THz-enabled electron source in an ultrafast electron diffractometer. A multi-cycle THz-powered dielectrically lined waveguide (DLW) was used as electron rebuncher. 180 fs (FWHM) electron bunches were achieved with about 10000 electrons/pulse at 1 kHz repetition rate. This beam is used to probe the structural dynamics of single-crystal silicon demonstrating high-quality diffraction patterns at improved temporal resolution.

Result

In the experimental setup shown in Fig.1, the electron beam from a 53 keV photo-triggered DC gun is compressed by a multi-cycle THz powered DLW device. Its pulse duration is analyzed by a segmented terahertz electron accelerator and manipulator (STEAM), device not shown for simplicity (see ref. [3]). Ultraviolet (UV) pulses for photoemission in the DC gun, multi-cycle THz pulses to drive the DLW device, single-cycle THz pulses to drive the
STEAM device and optical pump laser pulses for sample excitation are all created using a single infrared Yb:KYW laser system producing 4-mJ, 650-fs, 1030-nm pulses at 1 kHz repetition rate. The linearly polarized multi-cycle THz beam is converted to a radially polarized beam via a segmented waveplate.

The DLW design consists of a cylindrical copper waveguide of diameter 790 µm and a dielectric layer of alumina with a wall thickness of 140 µm. Conical horns are used to couple THz energy into the waveguides. The DLW-structure supports a traveling transverse-magnetic waveguide mode (TM01 mode), which can be used for different THz-based electron manipulations [4]. In this work, we are mainly using its compression function. Compression of the electron bunch is based on “velocity bunching”, where the E field imparts a longitudinal temporally varying energy resulting in a velocity gradient that causes compression of the electron bunch as it propagates.

Compression of the bunch is shown in Fig. 2(a). At maximum compression the electron bunch duration was reduced by a factor of > 10 to around 180 fs (FWHM) measured by the STEAM streaker. Higher THz fields lead to much shorter pulse duration as well as a shorter focusing position [4]. However, limited by the mechanical design of the current vacuum chamber, we were not able to bring the sample closer to the buncher.

![Fig. 2](image)

Fig. 2 (a) Measured (red square) and simulated (black line) electron bunch length as a function of the applied THz field. (b) Electron diffraction images of single-crystalline silicon with a face-centered cubic structure. (c) The relative intensities change of the 400 diffraction spots as a function of time delay under incident laser fluence of around 5 mJ/cm² with compressed electron bunch (red) and uncompressed electron bunch (green).

To demonstrate the performance of the setup, we measured the ultrafast heating dynamics of single crystalline silicon. Figure 2(b) shows the diffraction signal collected with 1s exposure time. The sample is photo-excited with 515 nm laser pulses at a fluence of around 5 mJ/cm². The recovered structural dynamics are shown in Fig. 2(c). The dynamics measured with the compressed electron beam show a clear improvement in time resolution compared to the case measured with the uncompressed electron beam [Fig. 2 (a)]. The exponential fit of τ = 1±0.2 ps is slightly longer than previous measurements (0.88 ps) [6] which is mainly due to the pulse duration of the pump laser (~0.5 ps) that limits the system temporal resolution.

**Conclusion**

We have demonstrated ultrafast electron diffraction with electron bunches from a DC electron gun compressed by a THz-driven rebunching DLW. At present, 180 fs (FWHM) electron bunches with about 10000 electrons at 1 kHz repetition rate can be achieved, providing high quality diffraction patterns for ultrafast structural dynamics studies. Exploring the versatile functionality of the THz-powered devices paves the way for future compact high repetition rate, high energy ultrashort electron sources.

**References**

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