

# TERAHERTZ DRIVEN COMPRESSION AND TIME-STAMPING TECHNIQUE FOR SINGLE-SHOT ULTRAFAST ELECTRON DIFFRACTION

M. A. K. Othman, A. E. Gabriel, M. C. Hoffmann, F. Ji, E. A. Nanni, X. Shen, E. C. Snively, X. Wang, SLAC National Accelerator Laboratory, Menlo Park, CA, USA

## Abstract

We demonstrate a new technique for MeV-UED beamlines that enables unprecedented temporal resolution in pump-probe measurements. This technique utilizes two counter-propagating quasi-single-cycle THz pulses generated from two OH-1 organic crystals, coupled into an optimized THz structure to produce compressed electron bunches with suppressed jitter. We show that the time-stamping technique can improve the temporal resolution of single-shot time-resolved diffraction measurements in single-crystal samples by 3 fold.

## INTRODUCTION

Ultrafast structural dynamics are well understood through pump-probe characterization using ultra-fast electron diffraction (UED) and X-ray free electron laser (XFEL) instruments. Advancements in electron diffraction and spectroscopy techniques open new frontiers for scientific discovery through interrogation of ultrafast phenomena [1-4]. UED technology has been a very active area of innovation bolstered by pivotal research in rf accelerators that can achieve an ever-increasing brightness and unprecedented spatiotemporal resolution. Furthermore, laser-generated THz radiation has seen a surge of interest as an efficient approach for manipulating ultrafast photoelectrons with high temporal precision and efficiency. Indeed THz technology [5-10] offers potential improvements in accelerator performance and brightness. Previously, we have demonstrated that strong-field THz radiation can be utilized to efficiently manipulate and compress ultrafast electron probes and also offer temporal diagnostics with sub-femtosecond resolution enabled by the inherent phase locking of THz radiation to the photoemission optical drive [11-14]. In this work, we demonstrate a novel THz compression and time-stamping technique to probe solid-state materials at time scales previously inaccessible with standard UED [15].

## MEV-UED SETUP WITH THz-INDUCED TIME-STAMPING

A simplified schematic of the dual-fed THz compression and time stamp setup at the SLAC MeV-UED beamline is shown in Fig. 1 (see Table 1 for beam parameters). Laser pulses from a Ti:Sapphire laser source at 800 nm with 25 fs r.m.s and up to 13 mJ of pulse energy are used to generate UV for the rf photoinjector, THz pulses for electron temporal streaking diagnostics, and near infrared ( $\lambda = 1300$  nm) source for separate THz pulses used for compression and time-stamping. Two OH-1 crystals were pumped with two 1300 nm laser beam obtained from an optical parametric amplifier (OPA) and transported into the

vacuum chamber for the compression stage. The laser pulses are controlled in delay and amplitude. The realized THz energy efficiency is about 0.4%. Both THz pulses are then guided to feed a compressor structure using 2" diameter off-axis parabolic (OAP) mirrors with 2" focal length. An example of the measured THz pulses inside the compressor structure using electro-optic sampling (EOS) is shown in Fig. 2.

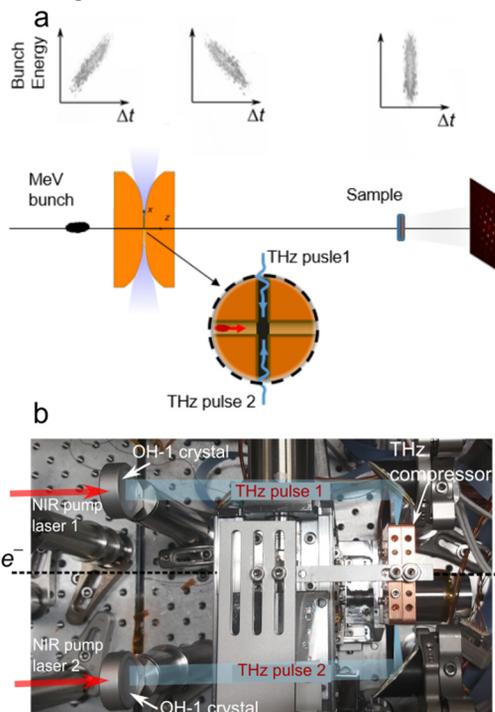


Figure 1: (a) Layout of the dual fed THz compressor and (b) photo of the setup inside the MeV-UED beamline.

Table 1: Summary of SLAC MeV-UED Beamline Parameters with THz Induced Time-Stamping

Parameter	Value
Beam Charge	10 fC
Beam Energy	3 MeV
Repetition Rate	360 Hz
Normalized Emittance	8 nm·rad (THz ON) 48 nm·rad (THz OFF)
Bunch Length	154 fs r.m.s. (THz ON) 41 fs r.m.s. (THz OFF)
Time of arrival jitter	70 fs r.m.s. (THz OFF) 23 fs r.m.s. (THz ON)

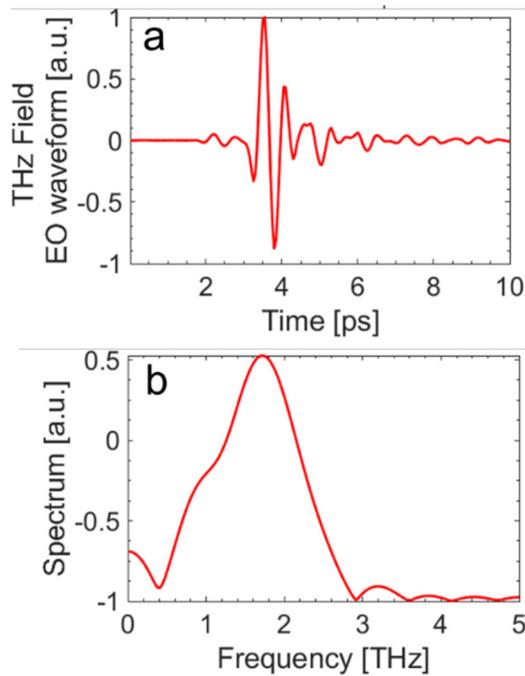


Figure 2: Measured EOS trace of the electric field inside the THz compressor, only from one THz pulse.

### Bunch Compression and Jitter Suppression

In the optimal compression phase, the electron probe centroid exhibits no transverse deflection. The two THz pulses were relatively delayed by about 200 fs to achieve the peak energy chirp required to achieve a compression by a factor of 3.8. The corresponding peak field gradient is about 130 MV/m. The minimum electron bunch length is measured to be 40 fs r.m.s. down from an average of 154 fs r.m.s. in the delay scan in Fig. 3.

A simultaneous improvement of the beam's shot-to-shot TOA is achieved, with a minimum TOA jitter of 23 fs r.m.s., down from 69 fs r.m.s.

### Time-Stamping for UED Time-Resolved Measurements

We use the time-stamping generated from the THz compression interaction to correct for TOA jitter using the transverse time-position correlation on a single-shot basis.

In Fig. 4 we show the ultrafast dynamics of THz-pumped single-crystal Au sample of thickness 11 nm as well as an integrated diffraction pattern. The Bragg peak intensity variations show faster dynamics in the case with THz compression and time stamping that leads to observing a resonance feature at  $\sim 1$  THz. This corresponds to a plasmonic resonance in the gold film that is excited by the THz pulse, which in turn cannot be resolved in the absence of compression. The overall temporal resolution is found to be around 56 fs r.m.s. compared to 181 fs r.m.s. without THz compression. Within individual single-shot time-stamped probes, a 5 fs temporal resolution was observed on the detector [15]. Measurements in polycrystalline samples were also demonstrated in [15].

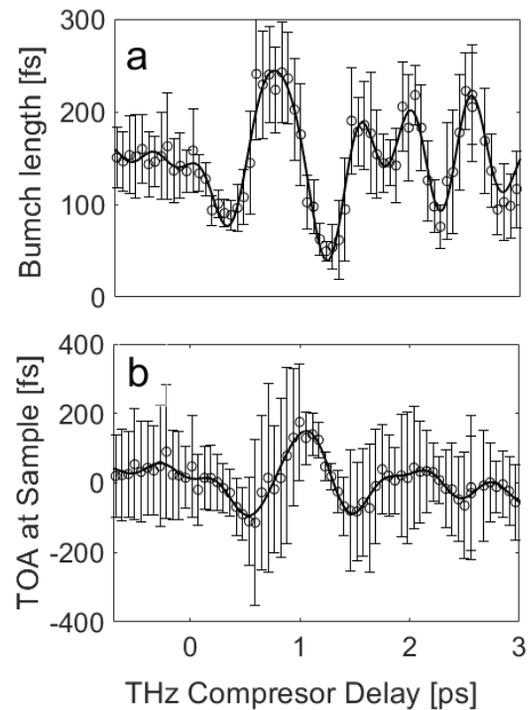


Figure 3: Single shot delay scan of the bunch length and time of arrival at the sample; measured using THz streaking, and showing a minimum bunch length  $\sim 40$  fs and jitter suppression at around 1.3 ps.

## CONCLUSION

We demonstrated a novel THz dual-fed compression and time-stamping technology leading to an unprecedented temporal resolution in optically-pumped thin film samples with at least 3 fold improvement over standard UED. The time-stamping technology offers new routes toward unveiling new features in the dynamics of quantum materials previously obscured by timing jitter of the rf accelerator systems.

## ACKNOWLEDGEMENTS

This research has been supported by the U.S. Department of Energy (DOE) under Contract No. DE-AC02-76SF00515. The SLAC MeV-UED program is supported in part by the DOE Basic Energy Sciences (BES) Scientific User Facilities Division Accelerator & Detector R&D program, the Linac Coherent Light Source (LCLS) Facility, and SLAC under Contracts No. DE-AC02-05-CH11231 and No. DE-AC02-76SF00515. M. C. H. is supported by the DOE Office of Science, BES, Award No. 2015-SLAC100238-Funding. This work was also supported by NSF grant PHY-1734015.

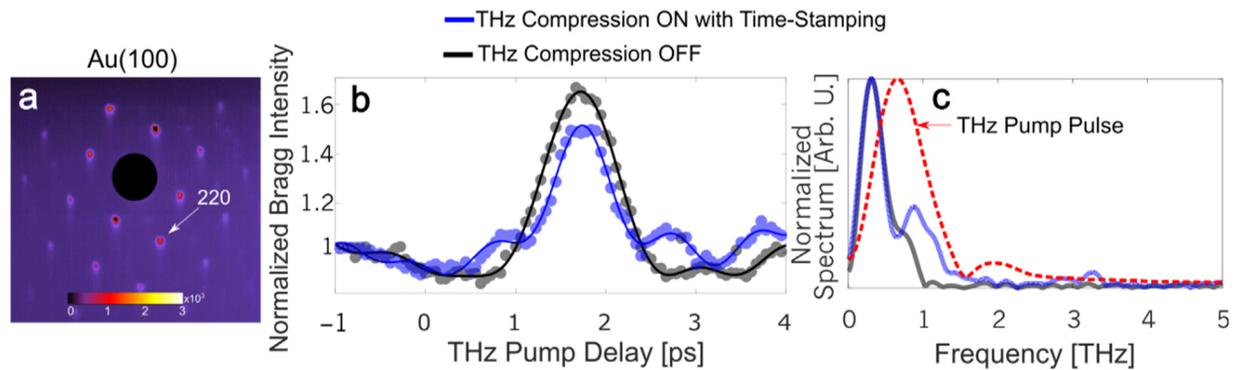


Figure 4: Temporal resolution of the THz time-stamped electron probes using a single crystal Au sample. (a) Diffraction pattern a single crystal Au(100) probed by a time-stamped electron probe taken before the arrival of the THz pump acquired from averaging 20 single shots. (b) Intensity of a Bragg peak (220), for THz compression and time-stamping and without, and (c) the corresponding frequency spectrum of the Bragg peak variations as well as the pump THz pulse in free space.

## REFERENCES

- [1] S. P. Weathersby *et al.*, “Mega-electron-volt ultrafast electron diffraction at SLAC National Accelerator Laboratory”, *Rev. Sci. Instrum.*, vol. 86, no. 7, p. 073702, 2015. doi:10.1063/1.4926994
- [2] E. J. Sie *et al.*, “An ultrafast symmetry switch in a Weyl semimetal”, *Nature*, vol. 565, no. 7737, pp. 61–66, Jan. 2019. doi:10.1038/s41586-018-0809-4
- [3] X. Shi *et al.*, “Ultrafast electron calorimetry uncovers a new long-lived metastable state in 1T-TaSe2 mediated by mode-selective electron-phonon coupling”, *Sci. Adv.*, vol. 5, no. 3, p. eaav4449, Mar. 2019. doi:10.1126/sciadv.aav4449
- [4] Y. Liu *et al.*, “Spectroscopic and Structural Probing of Excited-State Molecular Dynamics with Time-Resolved Photoelectron Spectroscopy and Ultrafast Electron Diffraction”, *Phys. Rev. X*, vol. 10, no. 2, Apr. 2020, doi:10.1103/PhysRevX.10.021016
- [5] M. Dal Forno *et al.*, “rf breakdown tests of mm-wave metallic accelerating structures”, *Phys. Rev. Accel. Beams*, vol. 19, no. 1, p. 011301, Jan. 2016. doi:10.1103/PhysRevAccelBeams.19.011301
- [6] M. A. K. Othman, B. J. Angier, A. A. Haase, E. A. Nanni, M. R. Roux, and A. V. Sy, “Prototyping of Brazed mm-Wave Accelerating Structures”, in *Proc. 10th Int. Particle Accelerator Conf. (IPAC'19)*, Melbourne, Australia, May 2019, pp. 3764-3765.  
doi:10.18429/JACoW-IPAC2019-THPGW078
- [7] M. A. K. Othman, M. C. Hoffmann, M. K. Kozina, X. J. Wang, R. K. Li, and E. A. Nanni, “Parallel-Plate Waveguides for Terahertz-Driven MeV Electron Bunch Compression”, *Opt. Express*, vol. 27, no. 17, pp. 23791–23800, 2019. doi:10.1364/OE.27.023791
- [8] M. A. K. Othman *et al.*, “Experimental demonstration of externally driven millimeter-wave particle accelerator structure,” *Appl. Phys. Lett.*, vol. 117, no. 7, p. 073502, Aug. 2020. doi:10.1063/5.0011397
- [9] S. M. Lewis *et al.*, “Design of a High Gradient THz-Driven Electron Gun”, in *Proc. 10th Int. Particle Accelerator Conf. (IPAC'19)*, Melbourne, Australia, May 2019, pp. 2098-2100. doi:10.18429/JACoW-IPAC2019-TUPTS077
- [10] H. Xu *et al.*, “Cascaded high-gradient terahertz-driven acceleration of relativistic electron beams”, *Nat. Photonics*, pp. 1–5, 2021. doi:10.1038/s41566-021-00779-x
- [11] E. C. Snively *et al.*, “Femtosecond compression dynamics and timing jitter suppression in a terahertz-driven electron bunch compressor”, *Phys. Rev. Lett.*, vol. 124, no. 6, p. 054801, 2020. doi:10.1103/PhysRevLett.124.054801
- [12] R. K. Li *et al.*, “Terahertz-based subfemtosecond metrology of relativistic electron beams”, *Phys. Rev. Accel. Beams*, vol. 22, no. 1, p. 012803, Jan. 2019. doi:10.1103/PhysRevAccelBeams.22.012803
- [13] L. Zhao *et al.*, “Few-femtosecond electron beam with terahertz-frequency wakefield-driven compression”, *Phys. Rev. Accel. Beams*, vol. 21, no. 8, p. 082801, Aug. 2018. doi:10.1103/PhysRevAccelBeams.21.082801
- [14] L. Zhao *et al.*, “Terahertz Oscilloscope for Recording Time Information of Ultrashort Electron Beams”, *Phys. Rev. Lett.*, vol. 122, no. 14, p. 144801, Apr. 2019. doi:10.1103/PhysRevLett.122.144801
- [15] M. A. Othman *et al.*, “Visualizing femtosecond dynamics with ultrafast electron probes through terahertz compression and time-stamping”. arXiv:2104.05691