optica

Femtosecond timing synchronization at megahertz repetition rates for an x-ray free-electron laser

Tokushi Sato,[1,2,†,*] Romain Letrun,[2,†] Henry J. Kirkwood,[2,†] Jia Liu,[2,8,†] Patrik Vagovič,[1,2,3] Grant Mills,[2] Yoonhee Kim,[2] Cedric M. S. Takem,[2] Marc Planas,[2] Moritz Emons,[2] Tomasz Jezynski,[2] Guido Palmer,[2] Max Lederer,[2] Sebastian Schulz,[4] Jost Mueller,[4] Holger Schlarb,[4] Alessandro Silenzi,[2] Gabriele Giovanetti,[2] Andrea Parenti,[2] Martin Bergemann,[2] Thomas Michelat,[2] Janusz Szuba,[2] Jan Grünert,[2] Henry N. Chapman,[1,5,6] and Adrian P. Mancuso[2,7]

[1]Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22607 Hamburg, Germany
[2]European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany
[3]Institute of Physics, Academy of Sciences of the Czech Republic v.v.i., Na Slovance 2, 182 21, Praha 8, Czech Republic
[4]Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22607 Hamburg, Germany
[5]Department of Physics, Universität Hamburg, Luruper Chaussee 149, Hamburg, Germany
[6]The Hamburg Center for Ultrafast Imaging, Universität Hamburg, Luruper Chaussee 149, Hamburg, Germany
[7]Department of Chemistry and Physics, La Trobe Institute for Molecular Science, La Trobe University, Melbourne, Victoria 3086, Australia
[8]e-mail: jia.liu@xfel.eu
*Corresponding author: tokushi.sato@xfel.eu

Received 5 May 2020; accepted 7 May 2020 (Doc. ID 396728); published 17 June 2020

A critical challenge of pump-probe experiments with x-ray free-electron lasers (XFELs) is accurate synchronization of x-ray and optical pulses. At the European XFEL we observed megahertz rate timing jitter of 24.0 ± 12.4 fs.

https://doi.org/10.1364/OPTICA.396728

The emergence of x-ray free-electron lasers (XFELs) operating at photon energies of around 10 keV (hard x-rays) over the past decade has opened up new possibilities for the physical, chemical, and biological sciences due to their high brightness and femtosecond (fs) pulses [1]. These intense and ultrashort x-ray pulses are used to record so-called molecular movies on fs time scales using a pump-probe (PP) measurement to achieve both high temporal and spatial resolution—a necessary step to truly understand the dynamics of matter. One of the greatest difficulties to achieve fs time resolution in PP experiments is the shot-to-shot timing jitter, which can be hundreds of fs in XFELs [2]. Therefore, the temporal resolution of an experiment may be severely limited by the timing jitter if the relative PP delay time is not monitored on a single-shot basis, prohibiting studies of fs dynamics. Unlike other hard x-ray FELs so far, which have been operating at ≤120 Hz, the European XFEL (EuXFEL) has been designed to deliver up to 27,000 pulses per second at megahertz (MHz) repetition rates with burst operation [1]. In order to fully benefit from the increased number of x-ray pulses that the EuXFEL can provide for PP experiments, a unique optical PP laser system was developed to deliver pulses as short as 15 fs with a burst structure matching that of the x-rays while maintaining minimal jitter between the XFEL and the optical laser using an optical synchronization system [3].

Characterization of the PP jitter at MHz repetition rate in the hard x-ray regime, which is presented here, was made possible with the installation of the photon arrival time monitor (PAM) at the Single Particles, Clusters, and Biomolecules and Serial Femtosecond Crystallography (SPB/SFX) instrument [4]. The PAM uses the spectral encoding technique that has already been successfully applied at other hard x-ray FELs [5] at up to 120 Hz. A 100 μm thick Ce:YAG was utilized as a target sample. Two GOTTHARD (Gain Optimizing microsTrip sysTem witH Analog ReaDout) detectors [6] operating at 564 kHz recorded alternating optical pulses in each MHz pulse train. Using radiofrequency (RF) pre-lock synchronization [7], the inter-train RMS jitter was measured to be 279 ± 32 fs. Figure 1 shows the relative time of arrival of x-ray and optical pulses using the RF and optical synchronization (OS) systems. An extremely low inter-train RMS jitter of 24.0 fs with an uncertainty of 12.4 fs was observed over a period of 10 min using OS [Fig. 1]. Additionally, measurements over a period of 2 h show no significant slow drift while using OS. These results demonstrate a significant advance towards fs PP experiments at MHz repetition rates.

In conclusion, the spectral encoding technique was developed for operation in the hard x-ray regime at a MHz repetition rate. The PP inter- and intra-train timing jitter was characterized at the EuXFEL with optical synchronization. Since the previous report [7], the inter-train RMS jitter has been reduced by 1 order of magnitude, down to 24.0 fs with 12.4 fs uncertainty, which

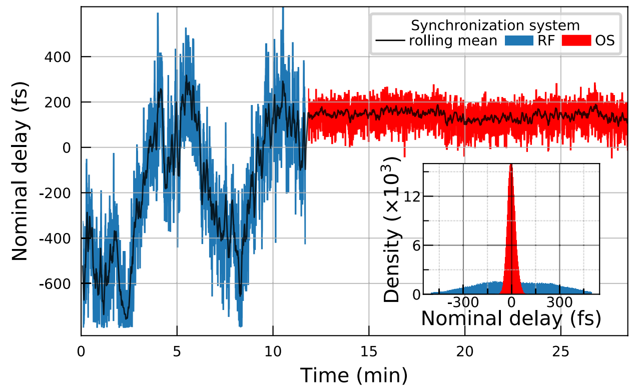

Fig. 1. The black line shows the rolling mean over a 5 s window. The inset shows a histogram of the relative arrival times using 100 x-ray pulses per train (10 Hz inter-train and 1.128 MHz intra-train repetition rates) with RF synchronization (blue, 854,430 pulses) measured using a 3.1 ps temporal window and optical synchronization (red, 431,300 x-ray pulses) measured using a 1.2 ps temporal window.

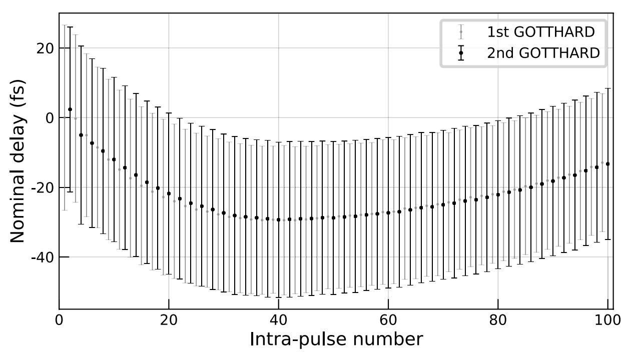

Fig. 2. Arrival time for each pulse with respect to the first pulse in its train when averaging over 5000 x-ray trains. The error bars show the standard deviation for each pulse in the train over a period of about 500 s.

can be attributed to the OS system and improved optical laser beam transport. Furthermore, the intra-train jitter was measured at 1.128 MHz repetition rate using a device that is available for users of the SPB/SFX instrument [Fig. 2]. Our work highlights the importance of the OS for future PP studies at high repetition rate and ultrashort x-ray sources. Owing to the excellent synchronization between the XFEL and the PP laser at EuXFEL, experiments requiring a temporal resolution above 100 fs will not require the conventional "measure and sort" approach and can thus be performed much more readily.

Funding. Helmholtz Association Strategic Investment funds; BioXFEL Science and Technology Center (STC-1231306); Max Planck Society for Medical Research; Australian Nuclear Science and Technology Organisation; Australian Research Council Center of Excellence in Advanced Molecular Imaging (CE140100011); Stiftelsen för Strategisk Forskning; Röntgen-Ångström Cluster; Knut och Alice Wallenbergs Stiftelse; Vetenskapsrådet (822-2013-2014); Ministerstvo školstva, vedy, výskumu a športu Slovenskej republiky; Bundesministerium für Bildung und Forschung (05E13GU1, 05K13GU7); Wellcome Trust.

Acknowledgment. We acknowledge European XFEL GmbH in Schenefeld, Germany, for provision of x-ray free-electron laser beamtime at the SPB/SFX instrument, and we thank the instrument group and facility staff for their great assistance. We thank T. Katayama and S. Owada for fruitful discussions.

Disclosures. The authors declare no conflicts of interest.

[†]These authors contributed equally to this work.

REFERENCES

1. B. W. J. McNeil and N. R. Thompson, Nat. Photonics **4**, 814 (2010).
2. J. M. Glownia, K. Gumerlock, H. T. Lemke, T. Sato, D. Zhu, and M. Chollet, J. Synchrotron Radiat. **26**, 685 (2019).
3. G. Palmer, M. Kellert, J. Wang, M. Emons, U. Wegner, D. Kane, F. Pallas, T. Jezynski, S. Venkatesan, D. Rompotis, E. Brambrink, B. Monoszlai, M. Jiang, J. Meier, K. Kruse, M. Pergament, and M. J. Lederer, J. Synch. Radiat. **26**, 328 (2019).
4. A. P. Mancuso, A. Aquila, L. Batchelor, R. J. Bean, J. Bielecki, G. Borchers, K. Doerner, K. Giewekemeyer, R. Graceffa, O. D. Kelsey, Y. Kim, H. J. Kirkwood, A. Legrand, R. Letrun, B. Manning, L. Lopez Morillo, M. Messerschmidt, G. Mills, S. Raabe, N. Reimers, A. Round, T. Sato, J. Schulz, C. Signe Takem, M. Sikorski, S. Stern, P. Thute, P. Vagovič, B. Weinhausen, and T. Tschentscher, J. Synch. Radiat. **26**, 660 (2019).
5. M. R. Bionta, H. T. Lemke, J. P. Cryan, J. M. Glownia, C. Bostedt, M. Cammarata, J.-C. Castagna, Y. Ding, D. M. Fritz, A. R. Fry, J. Krzywinski, M. Messerschmidt, S. Schorb, M. L. Swiggers, and R. N. Coffee, Opt. Express **19**, 21855 (2011).
6. A. Mozzanica, A. Bergamaschi, R. Dinapoli, H. Graafsma, D. Greiffenberg, B. Henrich, I. Johnson, M. Lohmann, R. Valeria, B. Schmitt, and S. Xintian, J. Instrum. **7**, C01019 (2012).
7. H. J. Kirkwood, R. Letrun, T. Tanikawa, J. Liu, M. Nakatsutsumi, M. Emons, T. Jezynski, G. Palmer, M. Lederer, R. Bean, J. Buck, S. D. D. Cafisio, R. Graceffa, J. Grünert, S. Göde, H. Höppner, Y. Kim, Z. Konopkova, G. Mills, M. Makita, A. Pelka, T. R. Preston, M. Sikorski, C. M. S. Takem, K. Giewekemeyer, M. Chollet, P. Vagovič, H. N. Chapman, A. P. Mancuso, and T. Sato, Opt. Lett. **44**, 1650 (2019).