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X-ray pulse preserving single-shot optical cross-correlation method for improved experimental temporal resolution

M. Beye,[1,2,a)] O. Krupin,[3,4] G. Hays,[3] A. H. Reid,[1,5] D. Rupp,[3,6] S. de Jong,[3] S. Lee,[3] W.-S. Lee,[1] Y.-D. Chuang,[7] R. Coffee,[3] J. P. Cryan,[3,8] J. M. Glownia,[3,8] A. Föhlisch,[2] M. R. Holmes,[3] A. R. Fry,[3] W. E. White,[3] C. Bostedt,[3] A. O. Scherz,[1] H. A. Durr,[1,8] and W. F. Schlotter[3,a)]

[1]*SIMES, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA*
[2]*Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 12489 Berlin, Germany*
[3]*LCLS, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA*
[4]*European XFEL GmbH, 22607 Hamburg, Germany*
[5]*Radboud University Nijmegen, Institute for Molecules and Materials, 6525 AJ Nijmegen, Netherlands*
[6]*Technische Universität Berlin, 10623 Berlin, Germany*
[7]*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*
[8]*PULSE, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA*

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We measured the relative arrival time between an optical pulse and a soft x-ray pulse from a free-electron laser. This femtosecond cross-correlation measurement was achieved by observing the change in optical reflectivity induced through the absorption of a fraction of the x-ray pulse. The main x-ray pulse energy remained available for an independent pump-probe experiment where the sample may be opaque to soft x-rays. The method was employed to correct the two-pulse delay data from a canonical pump-probe experiment and demonstrate 130 ± 20 fs (FWHM) temporal resolution. We further analyze possible timing jitter sources and point to future improvements. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3695164]

The x-ray free electron laser (FEL), the linac coherent light source (LCLS), has extended the study of femtosecond (fs) dynamics in nanoscale materials using ultrashort and intense x-ray pulses.[1] Such timescales are native to the electronic processes that mediate organization in correlated materials,[2] chemical reactivity,[3] and spin dynamics.[4]

Ultrafast evolution is typically investigated with a pump-probe scheme, whereby two ultrashort pulses of arbitrary delay impinge on a sample. The time resolution is limited both by the durations of the pump and probe pulses as well as their synchronization.[5] In most femtosecond pump-probe experiments, both pulses are derived from a common laser system near the experimental sample thus alleviating the need for actively stabilized synchronization.

Pump-probe experiments at the LCLS require active synchronization, because the primary laser system for the x-ray FEL is separated by one kilometer from the optical pump laser system and the experimental sample. To generate x-rays at the LCLS, the primary laser produces an ultraviolet laser pulse that strikes a photocathode to produce an electron bunch that is accelerated and injected into an undulator to produce an ultrashort x-ray pulse with of order 10^{13} photons. The intense x-rays originate from the amplification of spontaneous undulator radiation by the SASE (self-amplification of spontaneous emission) process.[6] This induces intrinsic variations in the arrival time in addition to variations in the long accelerator. Whereas, the optical pump laser system pulse originates from a stable oscillator before being amplified and transported 20 m to the experimental sample. A state-of-the-art synchronization system based on electron bunch time of arrival measurements is employed at LCLS to mitigate temporal drift and jitter between the optical and x-ray pulses to 280 fs (full width at half maximum, FWHM). This does not provide the certain path toward pulse duration limited time resolution that is possible with a direct cross correlation between the optical and x-ray pulses.[7]

In this letter, we present a way to measure the arrival time differences on a shot-to-shot basis. This enables the temporal resolution of data collected in a single shot mode to be enhanced by correcting each pump-probe delay after the measurement. The time resolution of pump-probe experiments at LCLS was improved at least by a factor of two to 130 fs (FWHM). The results achieved with this method present a clear path toward temporal resolutions below 100 fs.

Techniques for shot-to-shot measurements of the x-ray to optical cross correlation have been demonstrated in similar experiments.[8–11] The current experiment was implemented on the soft x-ray materials science instrument (SXR) at LCLS which is equipped with a monochromator.[12] Therefore, the pulse intensity on the sample is typically two to three orders of magnitude lower than in the full LCLS beam. In addition, the scientific scope of this beam line is centered around the study of solid state samples and liquids that absorb the full beam.[13] This prohibits timing measurements behind the interaction region as demonstrated on the atomic, molecular, and optical physics beamline at LCLS.[8]

We introduced the cross-correlation tool (a 200 nm Si_3N_4 film) into the x-ray beam in front of the monochromator to measure a signal that is independent from the monochromatization (Fig. 1). The film transmitted more than 80% of the x-rays for most photon energies available at SXR.[14]

[a)]Authors to whom correspondence should be addressed. Electronic addresses: martin.beye@helmholtz-berlin.de and wschlott@slac.stanford.edu.

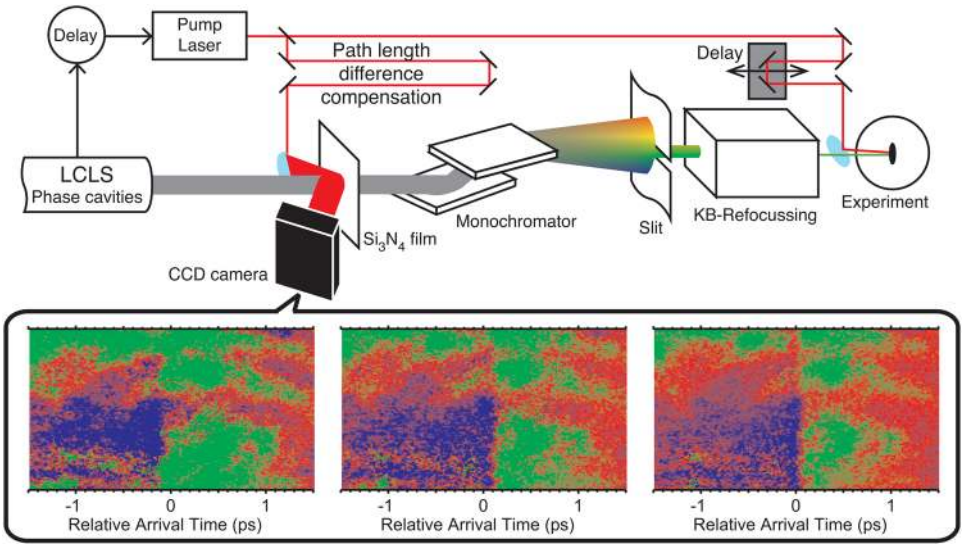

FIG. 1. (Color online) Experimental setup. The LCLS beam (grey) impinges on a thin Si_3N_4 film, prior to traveling through the monochromator. The rejected photon energies are blocked by a slit and the beam is refocussed into the experimental chamber. It is further illustrated how the electron bunch arrival time, as measured in a phase cavity, controls the timing of the optical laser. Part of this laser was split off to study the reflection from the x-ray irradiated Si_3N_4 film, while the main pulse traveled to the experiment. The optical laser path lengths were adjusted such that the split off laser pulse overlapped on the Si_3N_4 film with the same x-ray pulse as the main pulse in the optical x-ray pump-probe experiment. Typical camera images of the optical laser reflected from the film after background subtraction are shown at the bottom. The relative arrival time jitter is mapped onto a spatial coordinate on the camera (here already converted to time).

With an optical beam splitter, we divided the optical laser pulse after the amplifier and the weaker part was transported to the cross-correlator, whereas the main pulse traveled to the experiment. There was a 15 m spatial separation between the cross-correlator and the experiment. The transport of the optical beam over these paths introduces possible variations in the optical path that are not induced on the x-rays which propagate in vacuum.

Fig. 2 shows the geometry of the cross-correlator. A time-to-space mapping geometry was used to determine the ultrafast x-ray induced change in the optical reflectivity of the 200 nm Si_3N_4 film.[15,16] The x-ray pulses impinge normal to the surface of the film, quasi-instantaneously creating a lower optical reflectivity through the excitation of an electron-hole plasma in the irradiated millimeter-sized area.[17] The x-ray absorption events create core excitations that decay in 10 fs (Si 2p) and 6 fs (N 1s) mainly into energetic Auger electrons. The electron-hole plasma is generated through sub-femtosecond electron-electron scattering. The expected sub-15 fs reflectivity response has yet to be experimentally observed, because it is dominated by the x-ray and laser pulse durations. A laser pulse, relay imaged from a back illuminated iris impinges under 50° to the surface, effectively samples the reflectivity as a function of time. By imaging the reflected pulse onto a camera, the relative arrival times of the x-ray and the optical laser pulses are encoded into the position of the onset of the reflectivity change in the image.

After applying a background normalization, the images are corrected for possible tilts of the camera and projected onto the axis perpendicular to the reflectivity change. To find the position of the onset of the reflectivity change, a Gaussian is fitted to the first spatial derivative of the projection. After discriminating for unexpectedly large timing jumps and unphysical fit parameters, reliable timing information is obtained for 90% of the pulses, when an x-ray energy of 0.2 mJ is absorbed in the Si_3N_4 membrane.

To demonstrate the improvement of the time resolution, a simple pump-probe experiment was set up in the resonant soft x-ray scattering (RSXS) end station where the 800 nm laser pulses of around 55 fs (FWHM) duration were coupled into the experimental chamber collinearly with the unmonochromatized x-ray beam.[18] Both impinged on an 1000 nm Si_3N_4 film supported by a Si substrate. The optical laser reflectivity was monitored by measuring the reflected pulse energy relative to the incoming p-polarized pulse energy

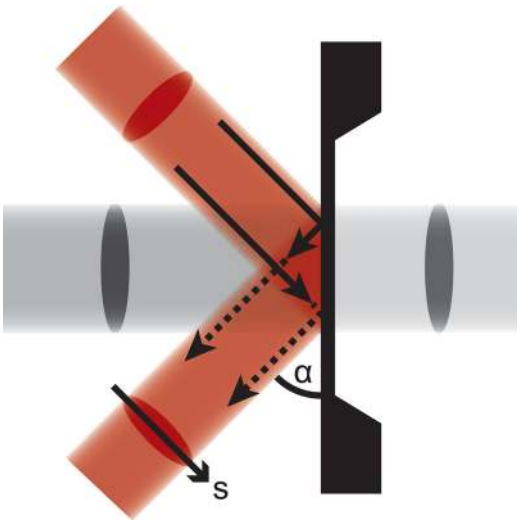

FIG. 2. (Color online) Geometry for the cross-correlator. The x-ray pulses impinge normal to the film. The optical laser pulses enter under an angle $\alpha = 50°$ to the film. This geometry effectively maps different relative arrival times Δt of both pulses onto different spatial coordinates Δs on the camera, following $\Delta t = \frac{\Delta s}{c \cdot \tan\alpha}$, with the speed of light c. The arrows depict the same arrival times in the optical laser time frame.

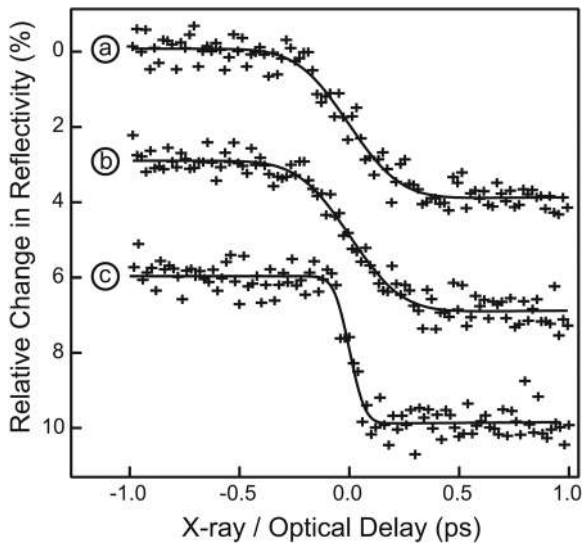

FIG. 3. Jitter corrected pump-probe data. The data shown are from the canonical measurement of the x-ray induced reflectivity change of a 1000 nm Si_3N_4 film on a Si substrate. The curves (b) and (c) are shifted downwards by 3% and 6% on the ordinate. Dataset (a) shows the uncorrected data that relies on the intrinsic timing synchronization at LCLS. The curve is fitted with a step function convolved with a Gaussian to model the pulse lengths and timing jitter broadening. The width in (a) is 420 ± 30 fs (FWHM). In dataset (b), the x-ray arrival time has been corrected by the phase cavity measurements of the electron timing relative to the master clock. The fitted width is 410 ± 30 fs (FWHM). In dataset (c), we correct the pump-probe delay with the relative arrival time information from the cross-correlator. The found residual width is 130 ± 20 fs (FWHM).

with photodiodes. Both lasers were operated at 60 Hz repetition rate. The electron bunch charge in the LCLS was 150 pC with a duration of 50 fs (FWHM), and the duration of the x-ray pulse is expected to be shorter.[8,10] The x-ray pulse photon energy was centered around 800 eV with pulse energies of 0.6 ± 0.3 mJ. To monitor the spatial and picosecond temporal overlap, similar methods were used at the cross-correlator interaction point as well as in the experiment.

The spatial overlap was checked on a frosted Ce:YAG crystal. Picosecond timing was achieved by temporally overlapping the rising edges of the photoemission current pulses that are either induced by the x-rays or the optical laser on a high bandwidth in-vacuum copper cathode at the interaction point.[11] Fine temporal overlap was then achieved in the cross-correlator by centering the reflectivity change onset on the camera. It is important to note that the timing of both pulses needs to be adjusted to independently achieve overlap in both the cross-correlator and in the experiment. For this purpose, the optical path length difference of the optical laser between both interaction points needs to be matched to the x-ray optical path length.

In the experiment, the delay stage for the optical laser was scanned back and forth over several picoseconds, changing the relative arrival time in the experiment, while the cross-correlator pulse was unaffected. The delay stage position was recorded for each shot, while the position of the onset of the reflectivity change in the cross-correlator was determined via analysis after the measurement. Both values were combined to yield the pump-probe delay in the experiment. The reflectivity data were then sorted into 20 fs wide bins. We show the results in Fig. 3, along with data where the timing has been deduced from the delay stage position only, as well as data where the electron bunch arrival time is measured and corrected with data from the electron bunch time-of-arrival diagnostic phase cavity.[7]

We find that the correction by the phase cavity data has only a minor influence on the time resolution in this pump-probe measurement. The correction with data from our cross-correlator improves the time resolution by more than a factor of three to 130 ± 20 fs (FWHM). This is on the time scale of electron dynamics and, therefore, enables measurements that disentangle electronic time scales (below 100 fs) from the timescales of typical phonon and spin dynamics in solid state systems which extend beyond several hundred femtoseconds.

We further estimate the performance of the different timing correction tools by correlating independent measurements of the same pulses. First, we compare cross-correlator data from independent regions on the Si_3N_4 film and find a cross-correlation width of only 22.7 ± 1 fs (FWHM), which is a performance measure of the cross-correlator. Relating this to the 130 ± 20 fs (FWHM) edge width in the data shown in Fig. 3, which contains the x-ray-laser cross correlation and additional timing jitter, we estimate that a residual jitter on the order of 100 fs is present.

The cross-correlation width of two independent measurements of the electron bunch arrival time in the phase cavities is 62.9 ± 1 fs (FWHM). Nevertheless, the phase cavity data and the cross-correlator values correlate only mildly with a Pearson coefficient of 0.34 and a correlation width of around 370 fs. This points to a dominant influence of the optical laser arrival time variations on the overall timing uncertainty. Already by looking at the raw arrival time data presented in Fig. 4, we observe a slow drift of the laser

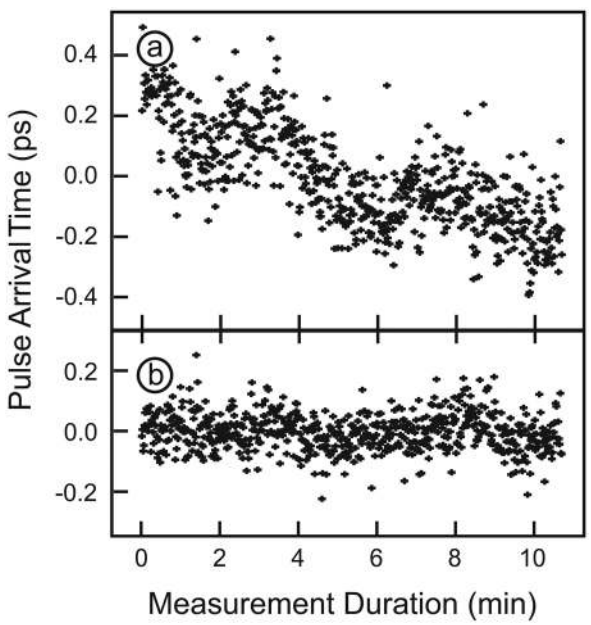

FIG. 4. Timing drifts during the measurement. The data shown are sampled with 1 Hz although we measured with 60 Hz repetition rate. (a) The relative arrival time between x-rays and optical pulses measured with the cross-correlator as a function of measurement time. (b) The arrival time of the electron bunch measured with the phase cavity relative to the master clock.

arrival time of several hundred femtoseconds over a period of 10 min.

We further analyze the origin and time scale of the different timing variations. By averaging the single-shot data over several shots, we can separate long term drifts from single-shot jitter. Furthermore, by comparing the cross-correlator data which contains the relative arrival times of both lasers with the x-ray only data from the phase cavity, we can disentangle the contributions of the different sources.

We find that the main contribution lowering the time resolution in the experiment stems from the hundreds of femtoseconds drifts of the optical laser arrival time on timescales that are longer than one second.[19] Correcting for this drift of the optical laser with the averaged cross-correlator data already improves the timing resolution from around 400 fs to around 200 fs (FWHM). We can further improve the time resolution by using the information on the electron bunch arrival time from the phase cavity, which brings the time resolution down to 180 fs (FWHM).

These findings lead us to conclude that the major source limiting the time resolution in pump-probe experiments are drifts of the optical laser arrival time on the time scale of seconds. Removing these drifts improves the time resolution by a factor of two. The electron bunch timing only influences the time resolution for shot-to-shot data through a jitter faster than a second which matches the bandwidth limit of the phase cavity timing correction loop.[7] Nevertheless, the best time resolution for SXR experiments can only be gained through the direct measurement of the relative arrival times with the demonstrated parasitic cross-correlator. The timing variations introduced through the independent transport of the optical laser to the different interaction points supposedly limit the time resolution, while the demonstrated 130 fs (FWHM) resolution is only a factor of two away from the ultimate pulse length limit of 70 fs for perfect synchronization between typical x-ray and optical pulses of 50 fs (FWHM) duration.

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