

# TIMESTAMPING FOR RELATIVISTIC ELECTRON DIFFRACTION

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## Abstract

At the Pegasus photoinjector laboratory, sub-mm electron bunches interact with thin metal targets to produce diffraction patterns[1]. On account of the bunches' high brightness, suitable patterns can be obtained in only a single shot, making the bunches effective probes on the sub-ps scale. As detection improvements push the probes to the sub-100-fs regime, the ability to pump the sample with ultrafast laser pulses opens the door for relativistic ultrafast electron diffraction to study femtosecond dynamics with a variety of applications. In lieu of a viable fs synchronization method, we review and explore a sub-100-fs electro-optic sampling setup to obtain single shot timing information about the pump-probe event.

## INTRODUCTION

Thermal melting in metals has been studied extensively using xray as well as non-relativistic ultrafast electron sources. The time scale here of the electron-lattice equilibration spans several picoseconds and thus can be obtained by averaging over several shots using a mechanical delay of the pump laser pulse. This technique is especially useful for non-relativistic electron sources (e.g., Ref. [2]) that already must be limited to low bunch charge in order to maintain a sub-ps length pulse. The trade off for this resolution is that this technique requires multiple shots to build sufficient intensity to resolve the diffraction peaks. Using a relativistic gun as an electron source has the advantage that the high gun fields accelerate the bunch to the final energy quickly, mitigating the space charge forces that otherwise try to drive up the bunch length. Using this concept, Pegasus lab has shown the ability to do sub-100-fs diffraction with enough brightness to resolve patterns in a single shot.

Using a relativistic electron diffraction source thus opens a new regime of study inaccessible to conventional keV electron diffraction sources. Certain materials have recently been shown to have photo-induced responses that occur at much shorter time scales. These processes include surface phase transitions, non-thermal melting, and even chemical reactions. The several ps delay accuracy inherent to pump-delay-scanning is insufficiently short to probe these time scales.

Timestamping was achieved at third generation xray facilities using electro-optic (EO) time stamping methods[3]. It is the goal of our current research to replicate similar results using a small university photoinjector laboratory instead of an advanced light source facility. This paper will

focus on describing the implementation and timing properties of the electro-optic technique for timestamping sub-100-fs pump-probe diffraction experiments at Pegasus lab. For more information on our diffraction results to date, please see Ref. [1].

## ELECTRO-OPTIC TIMESTAMPING

The layout of the 90° spatially encoding electro-optic sampling (EOS) scheme used at Pegasus lab is described in Ref. [4]. The only recent update to the system is replacing the detector CCD with a larger version to increase the total single shot window to > 30 ps.

We have previously discussed the two-dimensional field profile obtained with this setup. However, an outstanding issue is related to the propagation of the bunch fields in the irregular (i.e., mixing cylindrical and rectangular) geometry of the EO crystal. By delaying the EOS-probe laser pulse with respect to the cathode drive laser pulse, we can take a sequence of shots as the bunch propagates from the crystal edge to the crystal center. Figure 1 shows the fields propagating through the crystal. The most peculiar feature is that the wavefronts appear to curve as they propagate farther into the crystal. This behavior is inexplicable using a 2-d particle-in-cell simulation. Current work using full 3-d particle-in-cell codes is being done to fully appreciate the complex boundary conditions of the crystal.

To make this setup effective at timestamping pump-probe diffraction, it is essential that the EOS-probe and the sample-pump laser pulses remain exactly synchronized. To accomplish this, the EOS-probe/sample-pump pulse propagates from the laser aperture and is only split just before it enters the vacuum chamber. The combined transport line includes a mechanical delay stage driven by a stepper motor. This allows the overall timing to be measured during long delay scans.

### *Mechanical delay stage scan*

A mechanical delay stage scan is the logistically simplest approach to measuring relative pump-probe timing. This method involves shortening or lengthening the path length of the sample pump laser relative to the laser path being used to drive the photoelectron gun. A laser retroreflector is mounted on a remotely controlled motorized platform. A stepper motor is finely calibrated to a linear distance measurement to discern the total relative path length change per step.

Due to rf amplitude jitter as well as phase lock jitter with respect to the cathode-drive/sample-pump laser, it is not possible to measure the timing to better than  $\sim 1$  ps[4]

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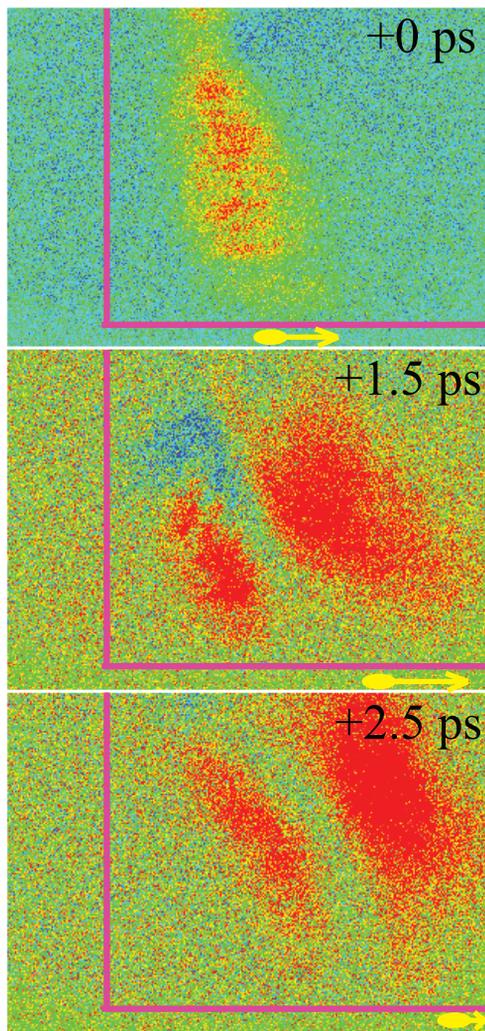


Figure 1: Scan of the mechanical delay stage as observed with the single-shot electro-optic time-of-arrival monitor. In this image, the electron bunch propagates from left to right. The laser has been delayed so that the IR probe arrives later in the lower frames. The drawn-in line indicates the boundary of the electro-optic crystal. The vertical direction represents the distance from the beam axis.

with this mechanical delay alone at Pegasus. This type of timing scheme is useful to look at processes such as the irreversible melting of metallic crystals which occur over tens of ps, but is not suitable for observing sub-ps changes.

### Analysis of the EO Signal for Time-of-Arrival

The Pegasus EOS time-of-arrival (TOA) monitor has the advantage of capturing single-shot time-of-arrival information non-destructively within a 30-ps timing window. Since our rf gun has a combined jitter of 1 ps, once the motorized pump/EOS delay is set, timing information is obtained for all subsequent shots until the delay is readjusted.

The method's inherent timing scale is set by the pulse duration of the EOS-probe laser, here about 35 fs. It is

well known that phase-velocity mismatch of the laser pulse and the electron bunch convolute the time profile of the bunch electric field. Additionally, THz-phonon resonances in ZnTe (the EO crystal medium) cause broadening of the bunch E-field in the crystal, which in the case of sub 100-fs bunches looks like a single-cycle THz pulse. It has been shown[4] that while these effects make longitudinal profiling of ultrashort bunches very difficult, they have little effect on the spatially-encoded EOS signal centroid, thus rendering EOS timestamping a promising technique for examining sub-100-fs processes.

## BENCHMARKING ELECTRO-OPTIC TIMING WITH RF DYNAMICS

Before moving on to the study of irreversible changes in pump-probe experiments, we find it necessary to benchmark the EO TOA apparatus. Since the pump-probe experiment is still being planned, it is necessary to use a well-understood source to verify the TOA information. In particular, we are currently examining using the dynamics of the rf gun in response to a change of initial conditions to imprint measurable changes in time-of-arrival and energy on the electron bunch. The concept is that there will be a clear and accurate correlation between the bunch energy centroid (as measured with a dipole spectrometer) and the relative TOA (equivalent to the gun output phase plus a drift, as measured at the electro-optic interaction point). These studies are used to establish confidence in the EO TOA measurement and to develop a better understanding of basic particle dynamics in our gun.

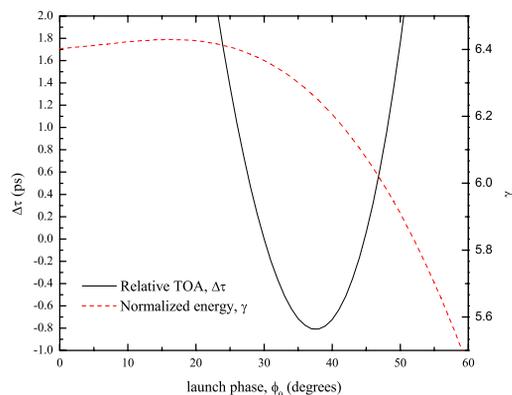


Figure 2: Predicted TOA and energy for the rf gun field gradient  $E_{0,\text{rf}} = 65$  MV/m. It is apparent by comparing this plot to Fig. 3 that the actual observed field gradient is lower (60 MV/m) resulting in a lower overall centroid bunch energy.

Our starting point is a very simple model[5] that couples the final energy of the bunch with the output phase (also known as the time-of-flight, or relative time-of-arrival). The model only deals with single-particle dynamics, and

ignores effects due to self-fields. Because we work with a high gun gradient and relatively low bunch charge (2 – 20 pC) we assume the single-particle model to be a faithful representation of the mean bunch behavior.

We determined the gun spatial field profile empirically by measuring the on-axis variation in the longitudinal direction during gun cold tests. To solve the coupled equations for the energy and gun exit phase we turn to a numerical integrator. The initial conditions include a initial launch phase term  $\phi_0$  that is sampled over a large range to obtain a final energy  $\gamma$  and gun output phase for each launch phase. The difference in launch and final phases gives the transit time through the gun, and a simple drift from the gun aperture added to this difference gives the time-of-arrival ( $\Delta\tau$ ) relative to the EOS probe laser pulse. Fig. 2 shows the model calculation of  $\gamma$  and  $\Delta\tau$  as a function of the launch phase  $\phi_0$ . These traces are obtained for a specific value of the mean rf electric field amplitude.  $E_{0,rf}$ .

This method of benchmarking relies on the fact that the rf amplitude jitter is much smaller than the rf phase jitter. Both effects contribute to the overall uncorrelated relative TOA jitter. This means that the energy measurement is very accurate since it is less sensitive to shot-to-shot fluctuations. It should be noted that the rf amplitude has a large (2 – 3%) slow drift that can be corrected with an active phase feedback circuit. The data in Fig. 3 was obtained over several minutes without the feedback correction. The typical measured fast amplitude fluctuation is on the order of 0.5%. Thus the spread in energies for this data set is higher than ideal for electron diffraction.

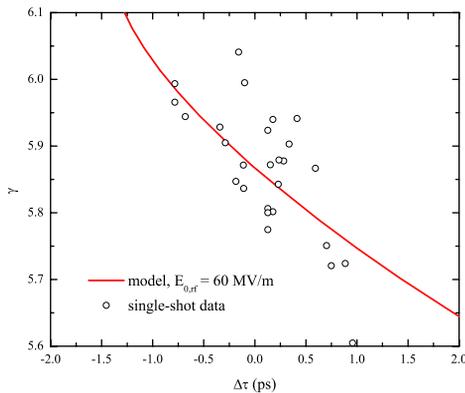


Figure 3: Measured correlation of the final energy with the relative time of arrival at the electro-optic detector. The red line represents the model calculation assuming the gun gradient is  $E_{0,rf} = 60$  MV/m. Phase jitter causes the points to move across the line, while rf amplitude jitter moves points onto different isoclines. The launch phase is  $\phi_0 = 25^\circ$  with respect to zero crossing. The mean energy is measured as 3 MeV with an rms fluctuation of  $\pm 100$  keV. The rms time of arrival is 0.5 ps.

It is apparent that the model predicts the correlation to some extent between  $\gamma$  and  $\Delta\tau$  as shown in Fig. 3. An effort will be made to reduced the uncorrelated jitter component from rf amplitude fluctuations by making future runs with the phase feedback loop enabled.

## CONCLUSION

Electro-optic based timestamping has been established at Pegasus photoinjector lab for sub-100-fs pump-probe experiments. New results from benchmarking the timestamping against predicted rf dynamics have reinforced confidence in this electro-optic timing method. Work will continue towards the goal of achieving relativistic femtosecond electron diffraction at Pegasus photoinjector laboratory.

## REFERENCES

- [1] P. Musumeci, J.T. Moody, and C.M. Scoby, Ultramicroscopy **108** (2008).
- [2] B.J. Siwick, et al., Science **302** (2003).
- [3] D.M. Fritz, et al., Science **315**, 5812 (2007).
- [4] C.M. Scoby, P. Musumeci, J.T. Moody, and M.S. Gutierrez, PR-ST Accel. and Beams **13** (2010).
- [5] Kwang-Je Kim, Nucl. Instr. and Methods in Phys. Res. (1989)