# BUNCH LENGTH MEASUREMENTS IN LOW-α MODE AT SPEAR3 WITH FIRST TIME-RESOLVED PUMP/PROBE EXPERIMENTS*

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*Abstract*

By operating SPEAR3 in low-α mode the storage ring can generate synchrotron radiation (SR) pulses of order 1ps. Applications include pump-probe X-ray science and the production of THz radiation. Measurements of the bunch length are difficult because the light is low intensity and synchroscan streak cameras typically provide resolution of only a few ps. Laser/SR cross-correlation in a non-linear optical crystal, however, can resolve pulse lengths of order 1ps. In this paper we report on an improved experimental setup at SPEAR3 as well as results from time-resolved laser pump, x-ray probe studies of the orthorhombic-to-superionic conducting phase transformation in $Ag_2Se$.

## INTRODUCTION

Recent developments in ultrafast x-ray measurement techniques have enabled the detailed study of processes occurring at the fundamental timescale for molecular vibrations, chemical reactions, and phase transformations [1]. By operating in low-α mode, SPEAR3 can produce X-ray pulses on the order of the requisite 1ps time scale, with beam intensity limited by the vacuum chamber and coherent synchrotron radiation (CSR) impedance [2]. As a tool for laser pump, X-ray probe studies of material dynamics, this mode of operation has multiple advantages when compared to other available light sources. With a 1.28MHz repetition rate for a single bunch, data can be collected with high signal-to-noise when coupled with a high repetition rate laser. This allows SPEAR3 to serve as a powerful compliment to the low repetition rate, high time resolution LCLS facility with GW pulse power. Additionally, at ~$10^4$ photons/bunch, the SPEAR3 low-α beam intensity compares favorably with lower flux 'laser-sliced' sources in applications for which hundred-femtosecond resolution is not required [1].

Critical requirements for time-resolved pump/probe measurements include accurate knowledge of the X-ray bunch duration and its precise arrival time with respect to the laser pulse. There exist multiple electro-optic methods capable of measuring short bunch profiles [1,3,4]. Where the THz radiation component is not available, nonlinear optical mixing of visible SR with an ultrashort laser pulse holds multiple advantages [5,6]. Furthermore, using the same laser for both bunch length characterization and pump/probe measurements allows deconvolution of the real-time instrument response function from the data.

In this paper we report on improvements to laser/SR cross-correlation measurements at SPEAR3. This diagnostic tool clearly resolves the nominal 21ps rms bunch length as well as 1-5ps rms bunches with <10μA single bunch current in the low-α configuration. Using the same laser system, first measurements of temperature-driven structural phase transformations of orthorhombic $Ag_2Se$ nanocrystals were performed. A deeper understanding of this transformation may enable use in phase-change memory and high-density energy storage applications [7,8].

## EXPERIMENTAL CONFIGURATION

As first reported by Zolotorev, cross-correlation measurements can be performed by mixing a narrow-band visible component of power SR radiation with a short, high laser pulse in a non-linear optical medium [5]. The initial configuration at SPEAR3 used an 800nm mode-locked Ti:Sapp oscillator (Femtolasers, XL500) operating 5.1MHz repetition rate, pulse energy of 490nJ, and pulse width 60fs FWHM. The Ti:sapp laser was mixed with the 800nm SR component in a Type-I BBO crystal to yield a 400nm second harmonic product. The SHG photons were detected by an avalanche photodiode (APD) and passed through an RF lock-in amplifier to preferentially filter and record signal at 1.28MHz while rejecting any scattered laser component at 5.1MHz.

For these experiments the 500-nJ Ti:Sapp beam contained ~$10^{12}$ photons/pulse and the filtered 800nm SR beam ~$10^6$ photons/pulse/mA. Assuming the low-power BBO efficiency is linear with pump intensity, typical efficiencies for SHG production lead to an anticipated $10^{-3}$ cross correlation photons /pulse at low bunch intensities.

The main problems with the Ti:sapp system were obtaining precision spatial overlap between the laser and SR beams and isolation of the SHG photons. Nevertheless it was possible to resolve a 5.96ps rms SR pulse length with 86μA single bunch current [6]. The Ti:sapp oscillator was delicate, however, and therefore difficult to transport and tune for pump/probe experiments at other beam lines.

The second incarnation featured a 1030nm fiber laser operating at 1.28MHz, 2.4μJ pulse energy and pulse duration 500fs FWHM (Calmar Laser, Cazadero). A piezoelectric end mirror in the oscillator maintained the cavity length via phase lock to the 93rd or 371st harmonic, respectively. The overall jitter was nominally a few

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hundred fs. The arrival time of the narrow laser pulse relative to the SR beam was controlled by a precision RF delay unit with 250fs step size (Colby Instruments PDL-100A). For simulation purposes, the fiber laser could also be split, doubled to 515nm and directed collinear with the SR beam. This allowed testing of a relatively high power, short-pulse SR beam with excellent phase stability.

For the fiber laser configuration, the SR beam was filtered to 515nm and focused to a 50μm spot on a Type-II BBO crystal. The laser itself was focused to a 100μm diameter spot to ensure stable spatial overlap with the SR beam. To maintain reproducible overlap, a 25μm pinhole target was used to steer the SR and laser beams independently. Sum-frequency generation between the 1030nm fiber laser and the 515nm SR beam produced a 343nm mixing product, again at a rate of $\sim 10^{-3}$ photon/event or better. The 343nm product beam was separated from the input beams by means of multiple irises and bandpass-filters.

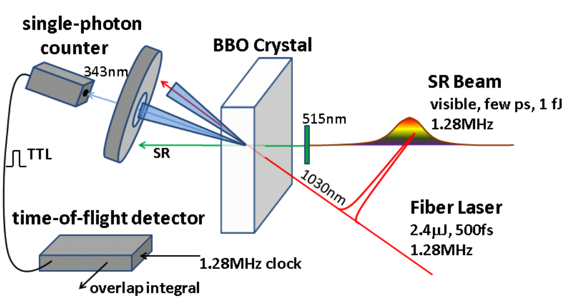


Figure 1: Cross-correlation with sum-frequency geometry.

Another upgrade to the fiber laser system was to replace the APD/lock-in amplifier photo-detection scheme with a single-photon counting module (SPCM, Hamamatsu) coupled to a fast time-of-flight digitizer card (ORTEC). The need for lock-in detection was eliminated by taking advantage of the clean wavelength separation between the 343nm signal photons and spurious scatter from the 1030nm laser and 515nm SR beams. With a Type-II sum-frequency BBO crystal the horizontal- and vertically polarized input beams were more co-linear leading to a higher non-linear conversion efficiency. Although SHG of the high-power laser light was not a problem (as in the Ti:sapp case), rejection of the THG component was a consideration.

## CROSS-CORRELATION RESULTS

The improved fiber laser cross-correlation configuration with Type-II sum-frequency BBO crystal and single-photon counting immediately yielded better results when compared to the Ti:Sapp system. As illustrated in Fig. 2, it was possible to clearly resolve a 3.6ps rms Gaussian pulse in low-α mode and only 10μA single bunch current. In this case the laser timing was adjusted in 250fs steps and each point integrated for 5s. For a reduction in momentum compaction by $\alpha=\alpha_0/60$, the theoretical bunch length is 2.6ps rms. Taking into account pulse-to-pulse laser jitter yields an estimate for

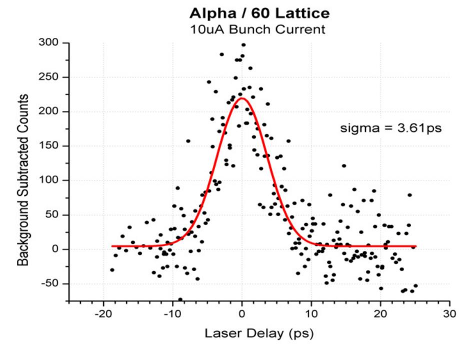


Figure 2: 3.6ps rms cross-correlation measurement in low-α mode with 10uA single bunch current.

2.5ps rms arrival time variations on the SR beam (synchrotron oscillations).

At even lower momentum compaction values ($\alpha \sim \alpha_0/400$) the bunch length enters the 1ps rms regime. As shown in Fig. 3a, the cross-correlation measurement can resolve the steep rising edges of the time-integrated data to about the 1.2ps level but in this case large synchrotron oscillations smeared the center of the measurement. Figure 3b illustrates direct measurement of synchrotron oscillations in terms of psec beam delay the same day of the cross-correlation measurements. The oscillations were unusually large. For the next set of measurements the RF system will be re-tuned to bring the oscillation amplitude to the 1psec range.

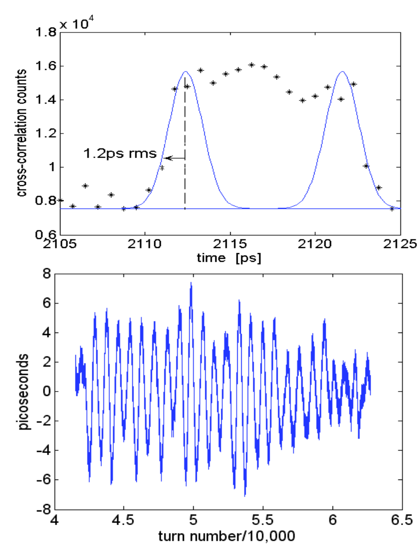


Figure 3: a) 1ps regime cross-correlation measurements taken with b) large phase oscillations.

## X-RAY PUMP/PROBE EXPERIMENTS

X-ray diffraction from 4nm diameter $Ag_2Se$ nanocrystals deposited on a $Si_3N_4$ window was collected at SPEAR3 beamline 10-2 in the nominal low emittance mode, at a photon energy of 12.5keV. Kirkpatrick-Baez mirrors were installed to focus the X-ray flux of $4\times10^{10}$ photons/sec to a ~50μm diameter spot, allowing the entire probe area to be illuminated by the ~100μm laser pump pulse. A powder diffraction pattern from the isotropically oriented nanocrystals was collected using a gated area detector (Dectris, PILATUS 100K) centered around the SPEAR3 timing bunch, while ignoring photons from other bunches. The laser fluence at the sample was 10mJ/cm$^2$, which is more than sufficient to raise the temperature of the nanoparticles to the 150°C orthorhombic-cubic phase-transformation temperature, assuming all of the energy absorbed by electronic excitation couples into the lattice, and using the bulk $Ag_2Se$ heat capacity to calculate the temperature jump.

The laser timing was configured such that the laser pump pulse arrival time at the sample was synchronized with the arrival of the X-ray timing bunch ($t_0$), and could be arbitrarily delayed about $t_0$ by means of either an optical delay stage (Newport, ILS) or the precision RF delay unit described above. High precision stages ensure that the limiting factor for experimental time resolution is limited to laser jitter and laser pulse duration (disregarding synchrotron oscillations).

For these experiments the powder diffraction rings were azimuthally averaged to produce a 1-D intensity profile vs. the scattering vector Q(Å$^{-1}$) at each time delay, as shown in Fig. 4.

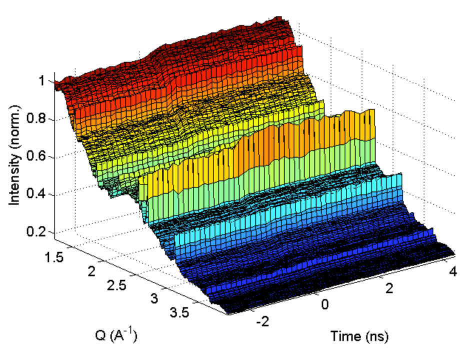


Figure 4: Powder diffraction pattern (Intensity vs. Q(Å$^{-1}$)) at multiple pump-probe time delays.

Changes to the diffraction pattern as a function of time delay after laser pump excitation are further elucidated by focusing on individual Q values of interest (Fig. 5). At scattering vector Q=1.535Å$^{-1}$, there was an increase in intensity at $t_0$, followed by a gradual recovery. Since an increase in intensity for all Q values has been observed (Fig. 4), it is difficult to definitively ascertain whether the observed transformation is precisely between the orthorhombic and superionic cubic phases.

Although these experiments did not take full advantage of the available X-ray time resolution, the data do provide early indications that structural transformations can be induced and observed using the high repetition rate laser pump/probe capability at SPEAR3.

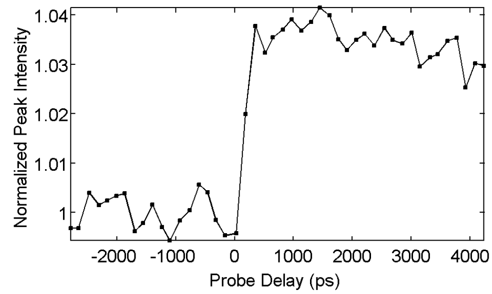


Figure 5: Diffracted intensity as a function of pump-probe time delay at Q=1.535Å$^{-1}$.

## SUMMARY AND FUTURE PLANS

SPEAR3 affords a unique opportunity to produce short-pulse, high-repetition rate x-ray beams in low-α mode. Without access to FIR or THz radiation, measurement of picosecond bunch lengths can be made with multi-turn laser/SR cross-correlation measurement. The initial Ti:Sapp laser system using a second harmonic BBO was replaced with a fiber laser with Type-II BBO in sum frequency configuration and single-photon counting. The new system can detect single bunches in low-α mode with <10μA in the 1-5ps range. Due to greater mechanical and thermal stability measurements are repeatable after a two week interval. In the future, we plan to investigate modulating the laser pulse arrival time to compensate for synchrotron oscillations and/or measure SR pulse arrival time directly and compensate in software.

Initial pump/probe experiments with the 1.28MHz fiber laser were carried out on several SPEAR3 beam lines. First tests indicate the ability to time-resolve structural transformations that are of fundamental interest to the materials science, chemistry, and physics communities. Future experiments will take full advantage of the short pulse X-ray time structure available in to further elucidate high speed dynamics of structural phase transformations such as the $Ag_2Se$ orthorhombic-cubic system.

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