Final Project Report

1. DE-SC0004078 , Purdue University (West Lafayette)
2. "Optoelectronic Picosecond Detection of Synchrotron X-rays,"
   PI: Stephen M. Durbin.
3. Report date: Aug 4, 2017.
   Period covered: 05/15/2015-05/14/2017

4. **Accomplishments**

Executive Summary

- The original optoelectronic design used a semiconductor sensor to convert an x-ray pulse time profile into an electrical profile that would then be sensed as it traversed a biased coplanar stripline. This proved to be a nominally workable approach, but the sensitivity and time resolution were limited, such that this appears to require much more development to become a practical detector.

- An all-optical design was discovered and developed that is much more promising for producing a practical picosecond x-ray detector. An x-ray pulse absorbed by a lithium tantalate sensor induces changes in the optical refractive properties that track the x-ray time profile and can be directly sensed with laser optics. This has immediate application at x-ray free electron laser sources and at certain synchrotron beamlines, and has potential for wider applicability.

The goal of this research program was to develop a detector that would measure x-ray time profiles with picosecond resolution. This was specifically aimed for use at x-ray synchrotrons, where x-ray pulse profiles have Gaussian time spreads of 50-100 ps (FWHM), so the successful development of such a detector with picosecond resolution would permit x-ray synchrotron studies to break through the pulse width barrier. That is, synchrotron time-resolved studies are currently limited to pump-probe studies that cannot reveal dynamics faster than ~50 ps, whereas the proposed detector would push this into the physically important 1 ps domain. The results of this research effort, described in detail below, are twofold: 1) the original plan to rely on converting electronic signals from a semiconductor sensor into an optical signal proved to be insufficient for generating signals with the necessary time resolution and sensitivity to be widely applicable; and 2) an all-optical method was discovered whereby the x-rays are directly absorbed in an optoelectronic material, lithium tantalate, which can then be probed by laser pulses with the desired picosecond sensitivity for detection of synchrotron x-rays. This research program has also produced new fundamental understanding of the interaction of x-rays and optical lasers in materials that has now created a viable path for true picosecond detection of synchrotron x-rays.

A. The GaAs Detector Program

The starting point of this research program was to try to emulate the successful sub-picosecond measurement of ultrafast laser pulses using a coplanar stripline geometry. This initially utilized a coplanar stripline on a photoconductive semiconductor substrate. The laser pulse would be absorbed at one end of the stripline, create an electrical pulse that propagates and is then detected using a second optical pulse and a pick-off electrode. This worked quite well for sub-picosecond laser pulses, but ultimately it did not translate well to 100 ps x-ray pulses. None of the various configurations we tested ever had true picosecond resolution, and sensitivity was also not sufficient.

The explanation for this difference between laser and x-ray pulses apparently is due to the difference between 1 ps and 100 ps pulses. The shorter pulse can be associated with an electromagnetic travelling

wave excited by the acceleration of charges during the turn-on phase of the induced photocurrent. The longer (100 ps) pulse also excited such a wave, but the acceleration is a hundred times smaller. This allows other current flow processes to mix with and to dominate the electromagnetic pulse, it seems, so that the overall time response was never as short as predicted by the laser results.

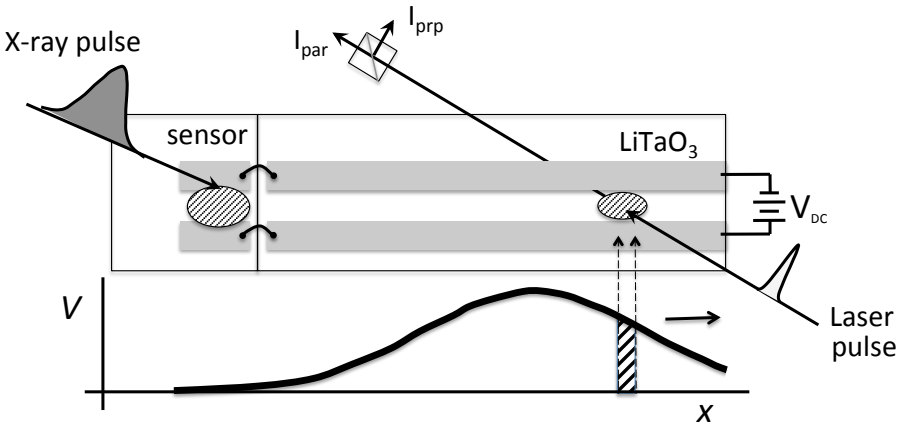

Figure 1: Conceptual design of the semiconductor-based optoelectronic detector. The sensor is photoconductive GaAs with coplanar stripline electrodes, electrically connected to coplanar striplines on a transparent $LiTaO_3$ substrate with a constant DC voltage bias. The absorbed x-ray pulse creates a current with a time profile based on the x-ray time profile, causing a pulse to travel down the $LiTaO_3$. A laser pulse transmitted through the substrate will have a perturbed polarization that is proportional to the instantaneous electric field. Measuring the polarization ratios versus x-ray – laser time delay gives the pulse time structure, to be compared to the ideal x-ray pulse profile.

The final version of the sample, the result of many iterations and design improvements in micro-fabrication of GaAs devices, provided an excellent test of the capabilities of this concept. The photo-conducting sensor was based on semi-insulating gallium arsenide (SI-GaAs), a commercially available material of excellent quality with high resistance but strongly photo-conducting under x-ray and laser illumination. The details of the final design are shown in Figure 2. One of the concerns with earlier designs was that the electric field from true "coplanar" stripline electrodes is a strong function of depth. This matters with x-rays, because the absorption depth at 12 keV in GaAs is about 12 microns, much longer than optical penetrations depths. Having highly variable field strengths versus depth could be responsible for poor sensitivity as well as temporal broadening of the response. The new "mesa" design solved this problem by using a controlled acid etch to remove GaAs in such a way as to produce straight sidewalls along the long mesa. Au contacts were then evaporated onto the sides (but not the top) of the mesa. This design was conceived and implemented by Dr. Yi Xuan, a collaborator and scientist at the Birck Nanotechnology Center at Purdue University. The internal electric field created by an applied DC bias produces a constant, strong electric field in the volume of the mesa that absorbs the x-rays. Implementation of this design improved signal strengths by at least an order of magnitude, and these sensors were also more robust at higher applied voltages. This design finally provided a good test structure for the optoelectronic detection of synchrotron x-ray pulses.

When an x-ray pulse is absorbed, many electron-hole pairs are created that produce a photocurrent in this otherwise highly resistive material. The turn-on of this pulse reflects the creation of these carriers; the decay of this pulse is generally limited by the electron-hole recombination time, unless they are otherwise collected before recombination occurs. When using laser pulses with this coplanar stripline detection system, it is essential to reduce the carrier lifetimes using extrinsic defects to get the fastest pulse decays. The most effective approach has been to use "low temperature" GaAs on the top surface

of the detector, which is grown by MBE on SI-GaAs substrates at lower growth temperatures than normal. This leads to a high density of trapping sites (As precipitates) and sub-picosecond decay times. Normally this is done with 1-2 micron layers, but we were able to convince our Purdue Physics Dept. MBE expert, Professor Michael Manfra, to grow LT-GaAs up to a thickness of 11 microns, meaning that well over half of the x-rays absorbed by these samples would do so within LT-GaAs.

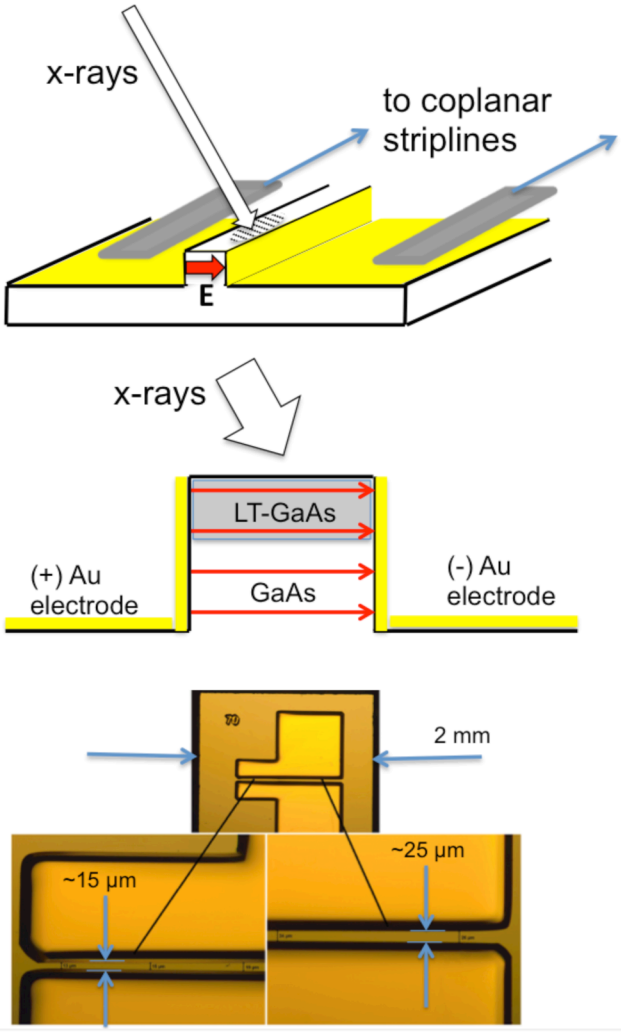

Figure 2: GaAs sensor configuration. Top: X-rays strike the bare top of the central "mesa," whose side walls are deposited with an Au thin film, forming the sensor stripline. Wires are bonded to the electrodes to connect with the $LiTaO_3$ coplanar striplines (not shown), and have a constant DV voltage bias. The mesa height is ~ 30 μm; various widths were tested, with some as small as 15 μm.

Middle: Close-up view of the mesa. This geometry ensures that the entire mesa has the same large electric field (red arrows), so that x-ray generated electrons and holes experience the same driving force at all absorption depths. One specimen had an 11 μm thick layer of low temperature gallium arsenide (LT-GaAs) grown by MBE, indicated by the shaded region.

Bottom: Optical micrograph of a semi-insulating GaAs sensor. Bright contact regions in upper image are the Au electrodes; large square pads on right are where wires are bonded. Lower images show the thickness of the mesa, which is only 15 microns on the left end where x-rays are absorbed.

The results of measurements on both plain GaAs and LT-GaAs are shown in Figure 3. The Advanced Photon Source running in 324 bunch mode has the shortest bunch durations, only 52 ps FWHM (σ=22 ps rms), which is shown as the black dashed curves in both plots. The ideal detector would yield data that closely tracked these curves, perhaps broadened slightly by x-ray-laser jitter. What is seen, however, is that the data for both detectors is significantly broader than the APS pulses. We assume that the measured curves are Gaussian functions that arise from convolution of the APS pulse Gaussian plus an unknown broadening Gaussian, which means the widths add in quadrature:

$$\sigma_{measured} = \left(\sigma^2_{x-ray} + \sigma^2_{broadening}\right)^{1/2}.$$

The "plain" GaAs detector was tested with applied biases ranging from 10 to 60 VDC. The average observed width was σ ~ 50 ps rms; the best case was σ=37 ps, using 50 volt bias. Note that even with the best case of 37 ps, the deduced $\sigma_{broadening}$ was still nearly 30 ps.

The other data shown in Figure 3 is for the LT-GaAs detector. When this type of material is utilized for ultrafast optical pulses, sub-picosecond resolution can be obtained, indicating sub-picosecond lifetimes for trapping of photo-excited carriers. With x-ray excitation, however, the total width is σ=48 ps, which yields en excess width of $\sigma_{broadening}$ = 42 ps. That is, the presence of LT-GaAs did not improve the time performance of this detecor. In fact it nominally increased the broadening; however this increase is within the range expected for variations in actual mesa width, applied voltage, etc. It seems fair to conclude that using LT-GaAs has little if any effect on the photoconductive response to 50 ps x-ray pulses, as opposed to its significant enhancement for sub-picosecond optical pulses. Further studies would be required to establish the physical origins of this difference, but for the present research project it appears that LT-GaAs is unlikely to improve performance.

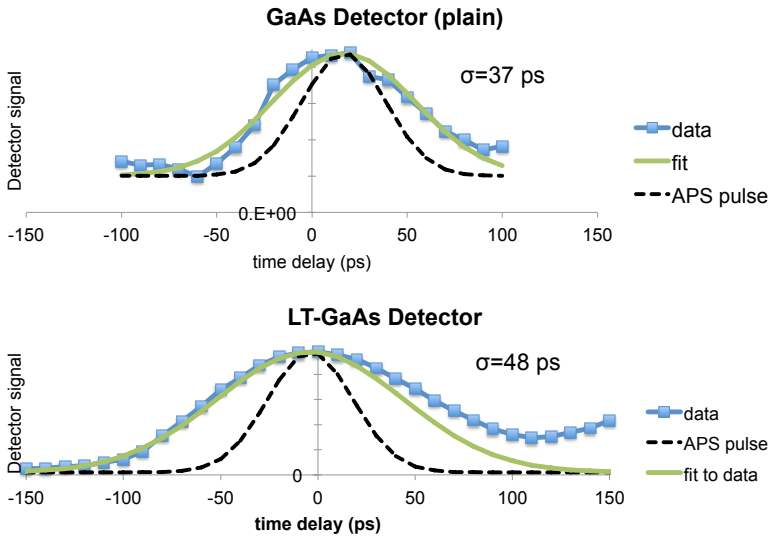

Figure 3: Final results for "plain" GaAs and LT-GaAs detectors. X-ray pulses at the Advanced Photon Source were used to test both a plain GaAs detector and one where 11 microns of LT-GaAs was grown on top. Blue curves are the data points, green is a best fit Gaussian, and the dashed black curves show the width of APS x-ray pulses in 324 bunch mode (22 ps RMS, which corresponds to 52 ps FWHM). The LT-GaAs had a slower time response, contrary to expectations. In general such measurements were very sensitive to applied bias and local width of the mesa. The top curve in fact is the fastest time response of any sample tested.

The following observations can be made from these results:

- The mesa geometry provides the strongest and most uniform electric fields for driving the x-ray induced photocurrent; there really is no room for design improvement here.

- Temporal broadening depends strongly on mesa width, applied voltage, and probably upon local imperfections in the mesa, but it was not possible to reduce the broadening below ~30 ps.

- An attempt to improve the time resolution by using LT-GaAs failed. The reasons for this are not clear.

While these results have advanced the state of the art in optoelectronic detectors for x-rays, the path towards true 1 ps resolution is not clear. It appears that the signal is very dependent on the actual transport of charge carriers across the mesa gap, whereas for femtosecond optical pulses the role of electromagnetic radiation is more important. An explanation for this difference is not readily available. The clear conclusion, however, is that a clear path towards the goal of 1 ps resolution is not seen. This makes the purely optical detection success described in the next section all the more important.

B. The Lithium Tantalate Detector Program

One of the major advances in the GaAs detector program described above arose when the scheme for detecting the electrical pulse traversing the coplanar stripline switched from laser interrogation of a small photoconducting pick-up electrode, to optical polarization detection through ferroelectric lithium tantalate ($LiTaO_3$) as illustrated in Figure 1. This led to a 100-fold increase in signal-to-noise, and was utilized for collecting the data shown in Figure 3. While pursuing this new detection scheme, an intriguing question arose: what happens of the x-ray beam hits the coplanar stripline gap on the lithium tantalate, instead of on gallium arsenide? What signals can be measured?

It was quickly noted that no electrical response on the striplines themselves are seen. That is because lithium tantalate is not a photoconductor, so no current is generated. However, it was readily seen that using the laser to probe the spot where x-rays are absorbed produced enormous changes in the polarization of the laser pulses. That is, x-ray absorption produced creates a localized charge distribution that strongly perturbs the dielectric constants, allowing easy detection with polarization analysis. We argue below that this presents a new and simpler route to optical (instead of optoelectronic) picosecond detection of x-rays.

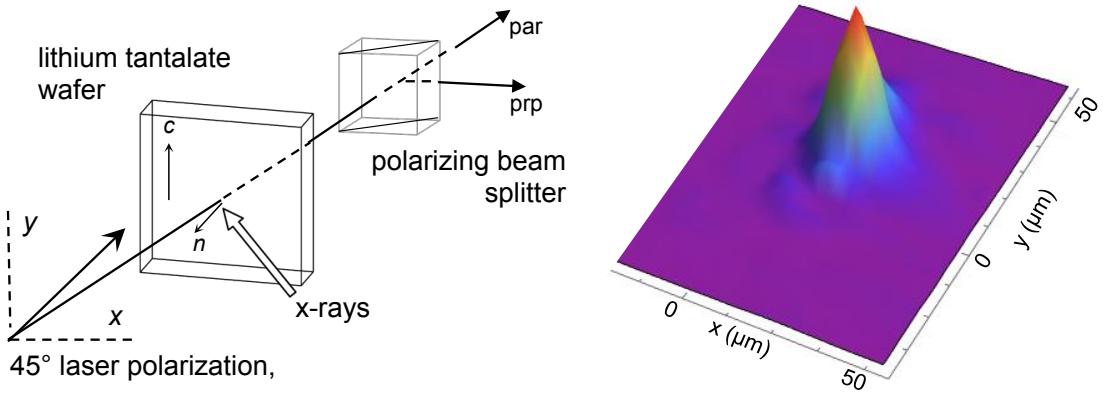

Figure 4: Optical birefringence imaging of x-ray excited lithium tantalate. Left: Schematic of experimental arrangement. A focused x-ray beam is absorbed by a lithium tantalate crystal, which is then probed by a linearly polarized laser pulse. The transmitted laser signal is split into the perpendicular and parallel components, and their x-ray induced difference is recorded. Right: results of a 2D raster scan of the laser across the x-ray spot, showing excellent signal to noise ratio and high spatial resolution.

What should be evident from Figure 4 is that the same x-ray beams used in the GaAs detector studies, obtained at APS Sector 7, produce a very strong and easily detected optical polarimetry signal. These data were taken in 324 bunch mode, where the time between pulses is only 11 ns. The signal showed no dependence on the time delay between the x-ray and laser pulses, sugesting that the time decay of the excitation must be much longer than 11 ns. Further investigations were conducted at APS Sector 14, where the specialized "Jülich" shutter reduces the time between x-ray pulses to 2 milliseconds or longer. This allows the full time dependence to be determined, from picoseconds to milliseconds.

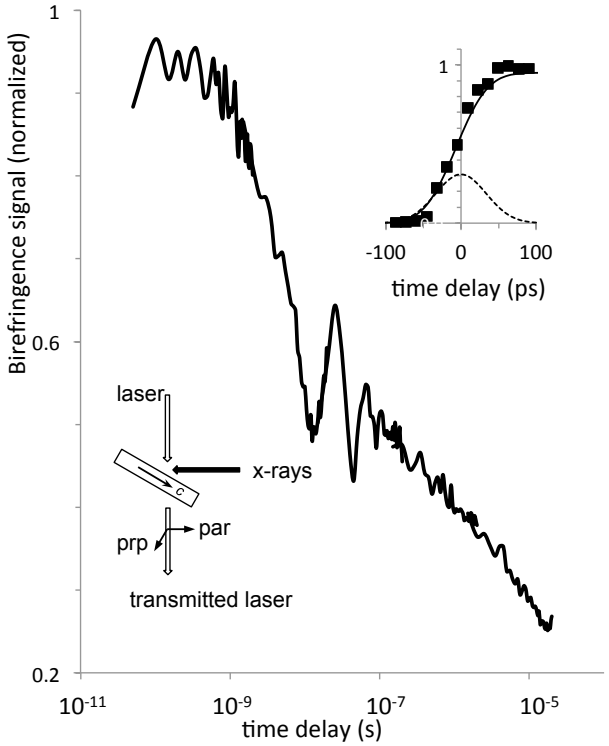

Figure 5: Time response of x-ray induced birefringence in lithium tantalate. Using 90 ps x-ray pulses, birefringence exhibits a non-exponential decay; the oscillation near 10^{-8} s is attributed to a bulk acoustic wave. Upper inset: initial response compared to the x-ray pulse profile (dashed line) and its integral (solid line). Lower inset: side-view of laser and x-ray beams at the sample, with c-axis denoted by arrow. X-rays are polarized horizontally (out of the page), and the linear polarization direction of the laser bisects the orthogonal "prp" and "par" directions.

The full dynamics of the birefringence response to x-rays is displayed in Figure 5. The geometry is shown in the lower inset; this is essentially the same as at APS Sector 7, except the laser beam is vertical instead of being in a horizontal plane. The upper inset shows the picosecond response of the x-ray induced optical birefringence. Note that it tracks very closely with the integrated x-ray dose, i.e. the integral of the Gaussian x-ray pulse profile. This proves that the phenomenon observed here is capable of measuring x-ray temporal profiles with picosecond sensitivity, the over-arching goal of this research project. In addition, the measurement is much simpler and easier the the GaAs optoelectronic approach, in part because it is all optical. There is no intermediate stage where the x-ray time profile must be mapped onto an electrical pulse profile, which then is detected optically. The all-optical approach also requires no microfabrication of the sensor head; commercially available lithium tantalate crystals are sufficient, without further processing.

The main data plot in Figure 5 shows the slow time decay of the x-ray induced optical birefringence in lithium tantalate. This reveals that tens of microseconds (at least) are required for the indcuced charges to decay, meaning that for this material the x-ray pulses must be separated by tens of microseconds. This is well suited for such specialized beamlines as APS Sector 14, but this would not be suitable for standard running modes (e.g. 24 bunch mode at the APS, with pulses separated by only 150 ns).

It should be stressed that lithium tantalate is the only ferroelectric, optoelectronic material that has been tested in this manner. It is certainly conceivable, and perhaps likely, that other materials may be superior to lithium tantalate, both in terms of sensitivity and in faster time decay. For example, both barium titanate and potassium titanate have larger electro-optic coefficients than lithium tantalate, meaning they may have greater sensitivity. Other materials with smaller bandgaps (e.g. semiconductors) are likely to have faster recombination times. Further materials research could provide a material that could work at high rep-rate synchrotron beamlines, not just at specialized ones like APS Sector 14.

Furthermore, there is much room for improvement in experimental sensitivity of the measurement. We have employed lock-in techniques and extensive signal averaging, which is more than adequate for the intense x-ray beams used so far. Sensitivity could be improved by orders of magnitude by doing pulse-by-pulse detection of the optical beams using high speed, high resolution video cameras.

In summary, the following results have been achieved with lithium tantalate:

- X-ray induced optical birefringence can detect x-ray pulse profiles with picosecond resolution.
- The measurements are easy to make, and no specialized detector fabrication is required.
- This has only been demonstrated at a specialized beamline with long times between x-ray pulses, and very high x-ray flux.
- Improved materials would allow use at standard synchrotron beamlines.
- Improved off-the-shelf detector technologies would improve sensitivity by several orders of magnitude.

C. Other significant results

During the course of this research project, a series of fundamental studies on the interaction of laser and x-ray pulses was conducted. These provide some of the knowledge base needed to understand detector development. Much of this is being compiled into a manuscript being submitted for publication shortly. For this report, we only include the first observation of x-ray pumped stimulated emission in a semiconductor:

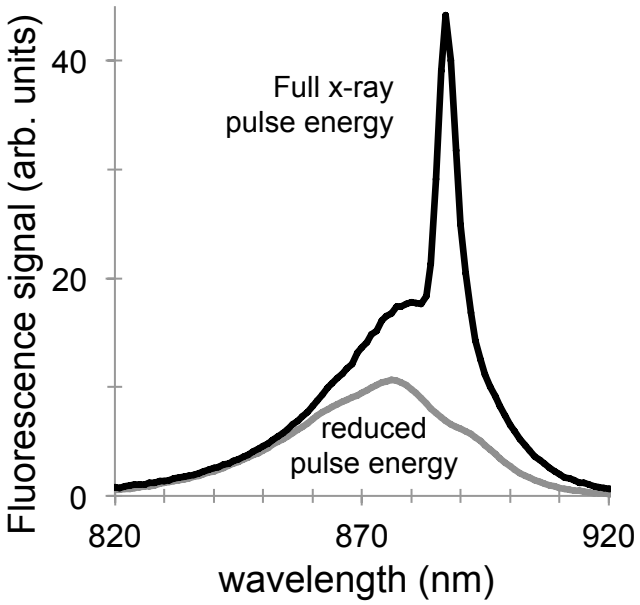

Figure 6: X-ray induced band fluorescence from GaAs. Upper curve is the optical spectrum obtained from x-ray excitation with 10 μJ pulses, a radiant exposure of ~8 kJ/m^2 per pulse; lower curve is the scaled output from x-ray pulse energies reduced by a factor of ~7. The prominent line at 885 nm indicates x-ray induced stimulated emission from the GaAs surface.

This figure shows the optical fluorescence spectrum from GaAs for two different x-ray fluxes. X-ray absorption generates a high density of electron-hole pairs, whose recombination creates optical fluorescence over a range of wavelengths corresponding to the pair energies. The lower curve, at reduced x-ray pulse flux, is a standard spectrum corresponding to band fluorescence in excited GaAs. The upper curve, however, is the spectrum after a factor of seven increase in x-ray flux. In addition to an alter background spectrum, the prominent spike is the clear indicator of a form of stimulated emission. Technically this is know as surface amplified spontaneous emission (ASE), since there is no incident optical beam to be amplified. That effect, however, has also been inferred from other measurements. In summary, we can report the first observation of x-ray pumped stimulated emission in gallium arsenide.

5. Publications

"Optical birefringence imaging of x-ray excited lithium tantalate," S. M. Durbin, A. Landcastle, A. DiChiara, Haidan Wen, D. Walko, and B. Adams, Appl. Phys. Lett. Photonics **2**, 2017. [http://dx.doi.org/10.1063/1.4997414]

"Time-delay measurement in the frequency domain," S. M. Durbin, S. C. Liu, E. M. Dufresne, Y. L. Li, and H. D. Wen, Journal of Synchrotron Radiation **22**, 1293 (2015).

In preparation:

"X-ray and optical beam interactions in GaAs," S. M. Durbin, T. Nagalu, and A. DiChiara (to be submitted to APL Photonics during August, 2017).

"Plasma-edge dynamics in photo-excited GaAs." S. M. Durbin and A. DiChiara

Other manuscripts are in earlier stages of preparation.

6. Project Personnel

Amanda Landcastle is a graduate student in the Purdue physics department who was supported in part by this grant during Fall 2016 and Spring 2017. She contributed significantly to many experimental aspects, including staffing the synchrotron beamlines at the Advanced Photon Source for two different experimental periods. She is the co-author of one publication.

Previously Shih-Chieh Liu, a graduate student in the Purdue Physics Department, was also supported as a graduate research assistant, and grad student Jeremy Munsell was involved with a courtse-based research project but was not financially supported by this project.

7. Final Year Activities

Data was acquired during beamtime at the Advanced Photon Source on different occasions during the last year: July, August, and November of 2016, and during February of 2017. These were crucial for the results described above. Much additional time has been spent with data analysis and manuscript preparation.

8. Other Support

No other support was utilized for this project.

9. Cost Status

All funds have been expended. During the final no-cost extension period, remaining funds were allocated to support for one graduate research assistant, synchrotron travel expenses for multiple beamtimes at the Advanced Photon Source, a small amount of supplies and expenses to substrates, optics, etc., one month of PI salary support, and the required indirect costs.