2-4-2013

Optoelectronic measurement of x-ray synchrotron pulses: A proof of concept demonstration

Stephen M. Durbin
Purdue University, durbin@purdue.edu

Aamer Mahmood
Birck Nanotechnology Center, Purdue University, amahmood@purdue.edu

Marc W. Caffee
Purdue University, mcaffee@purdue.edu

Sergei Savikhin
Purdue University, sergei@purdue.edu

Eric M. Dufresne
Argonne National Laboratory

See next page for additional authors

Durbin, Stephen M.; Mahmood, Aamer; Caffee, Marc W.; Savikhin, Sergei; Dufresne, Eric M.; Wen, Haidan; and Li, Yuelin, "Optoelectronic measurement of x-ray synchrotron pulses: A proof of concept demonstration" (2013). *Birck and NCN Publications.* Paper 1342.
http://dx.doi.org/10.1063/1.4791559

Authors
Stephen M. Durbin, Aamer Mahmood, Marc W. Caffee, Sergei Savikhin, Eric M. Dufresne, Haidan Wen, and Yuelin Li

AIP Applied Physics Letters

Optoelectronic measurement of x-ray synchrotron pulses: A proof of concept demonstration

Stephen M. Durbin, Aamer Mahmood, Marc Caffee, Sergei Savikhin, Eric M. Dufresne et al.

Citation: Appl. Phys. Lett. **102**, 051109 (2013); doi: 10.1063/1.4791559
View online: http://dx.doi.org/10.1063/1.4791559
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v102/i5
Published by the AIP Publishing LLC.

APPLIED PHYSICS LETTERS **102**, 051109 (2013)

Optoelectronic measurement of x-ray synchrotron pulses: A proof of concept demonstration

Stephen M. Durbin,[1,a)] Aamer Mahmood,[2] Marc Caffee,[1] Sergei Savikhin,[1]
Eric M. Dufresne,[3] Haidan Wen,[3] and Yuelin Li[3]
[1]*Department of Physics, Purdue University, West Lafayette, Indiana 47907, USA*
[2]*Birck Nanotechnology Center, Purdue University, West Lafayette, Indiana 47907, USA*
[3]*Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA*

(Received 11 December 2012; accepted 28 January 2013; published online 7 February 2013)

Optoelectronic detection using photoconductive coplanar stripline devices has been applied to measuring the time profile of x-ray synchrotron pulses, a proof of concept demonstration that may lead to improved time-resolved x-ray studies. Laser sampling of current vs time delay between 12 keV x-ray and 800 nm laser pulses reveal the ~50 ps x-ray pulse width convoluted with the ~200 ps lifetime of the conduction band carriers. For GaAs implanted with 8 MeV protons, a time profile closer to the x-ray pulse width is observed. The protons create defects over the entire depth sampled by the x-rays, trapping the x-ray excited conduction electrons and minimizing lifetime broadening of the electrical excitation. © *2013 American Institute of Physics*.

[http://dx.doi.org/10.1063/1.4791559]

Optoelectronic techniques have greatly expanded the capabilities for subpicosecond measurement and generation of optical pulses. Optoelectronics bypasses the challenges of terahertz bandpass electronics by relying instead on ultrafast optical pulses from readily available lasers to provide superior time resolution.[1–3] The time profile of laser pulses can be measured optoelectronically with subpicosecond precision, limited only by the ability to reduce photo-induced carrier lifetimes in the substrate to sufficiently low levels. This would be a very attractive capability for determining x-ray synchrotron pulse profiles, since a laser pump x-ray probe experiment can produce changes in x-ray reflectivity in less than a picosecond.[4–6] Standard x-ray detectors, however, are unable to resolve time responses faster than the synchrotron pulse duration (~50–100 ps). Subpicosecond resolution is possible using time slicing techniques where an ultrafast laser pulse interacts with an electron bunch in the storage ring, or bypassing synchrotrons for an x-ray free electron laser or a laser-induced plasma source.[7–9] Efforts to obtain higher time resolution at standard x-ray synchrotron beam lines include the development of x-ray streak cameras, which have demonstrated 2 ps resolution.[10] For an alternative approach, we report here on the optoelectronic detection of x-ray synchrotron pulses using GaAs substrates whose carrier lifetime is reduced by deep implantation with energetic protons.

Subpicosecond measurement of high repetition rate laser pulses begins with illuminating a biased coplanar stripline pair on the surface of a semi-insulating semiconductor (see Fig. 1, top). The photo-induced current pulse across the gap launches an electrical pulse down the stripline whose spatial profile follows from the laser pulse time profile, lengthened by the photo-induced carrier lifetime. A time-delayed sampling laser pulse, split off from the original laser pulse, illuminates the photoconductive gap to a sampling electrode. The current across the sampling gap is proportional to the instantaneous value of the original voltage pulse at the sampling electrode, but also convoluted by the photo-induced carrier lifetime in the sampling gap. Standard electronics are used to measure this current vs time delay between the two laser pulses, controlled with picosecond precision.

The photoconducting material is typically a thin layer of a semi-insulating semiconductor that has a negligible carrier density until illuminated by photons with an energy above the band gap. High absorption of these photons results in sub-micron penetration depths that determine the minimum device thickness. Because the photo-excited carrier lifetimes can be much longer than the desired time resolution, the semiconductor must have excess defects that trap the photocarriers with subpicosecond rates. This can be achieved using 0.5 μm silicon on sapphire substrates with oxygen ion implantation.[11] Similar results have been achieved by doping of various semiconductors, including InN and GaSb.[12,13] Another common approach is to employ "low temperature" gallium arsenide (LT-GaAs), in which a GaAs epilayer is grown by MBE such that excess As is incorporated, producing the needed trapping sites; these can also be fabricated as freestanding, micron thick devices.[14,15]

None of these approaches, however, are well-matched to the x-ray penetration depths in the ~10–100 μm range. We minimize the depth using GaAs substrates with 12 keV x-rays, just above the Ga and As *K* absorption edges, which reduces the exponential absorption length to ~11 μm. (For comparison, the absorption length in Si would be ~230 μm.) Standard implantation methods cannot reach this depth, and MBE growth is challenging for a thickness of the several absorption lengths needed, so instead we implanted with 8 MeV protons using the PRIME Lab van de Graaf accelerator at Purdue.[16] The implanted dose was 10^{15}cm^{-2}; simulations predict a near-surface vacancy density of $\sim 10^{18}$cm^{-3}; the relative penetration depths are indicated in Figure 2.[17] Proton bombardment of GaAs at 200 keV has reduced

---

[a)] durbin@purdue.edu.

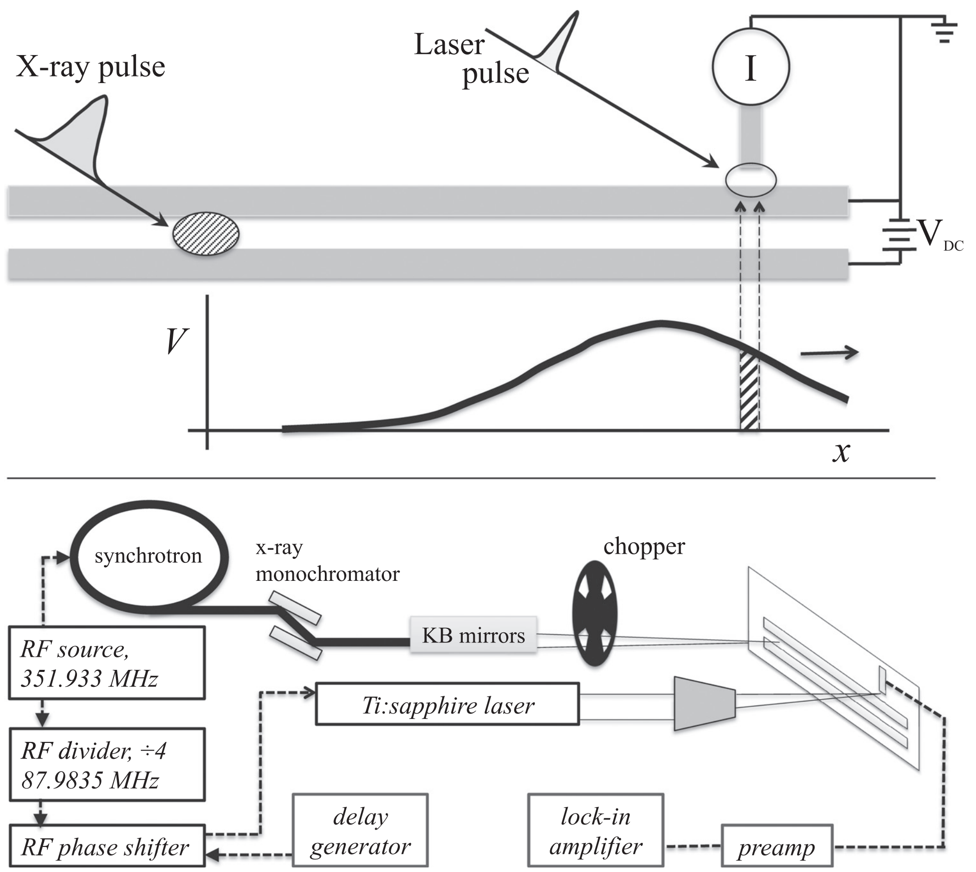

FIG. 1. Optoelectronic detection of x-ray synchrotron pulses. Top: Layout of the coplanar stripline on a GaAs substrate surface. A DC voltage is applied across the 25 μm gap. The x-ray pulse produces carriers that become a current pulse shorting the gap, creating a propagating voltage pulse. A time-delayed laser pulse creates photocarriers in the sampling gap, leading to a brief current proportional to the voltage at the gap. The current versus x-ray/laser pulse delay ($I_s(t)$) maps out the time profile of the original x-ray pulse, convoluted with carrier lifetime effects. Bottom: Layout of the synchrotron experiment. X-ray pulses are monochromated to 12 keV and focused by Kirkpatrick-Baez (KB) mirrors to a 50 μm spot size and intensity of $\sim 6 \times 10^{11}$ photons/s centered on the coplanar stripline gap, after passing through a 1 kHz chopper. The RF signal that controls the synchrotron electron bunches (351.933 MHz) is divided by four (87.9835 MHz) to provide the reference signal for the Ti:sapphire laser oscillator. The laser produces one laser pulse per x-ray pulse at a fixed time delay determined by a delay generator that controls a RF phase shifter. The laser pulse is focussed with a microscope objective onto a 10 μm photoconducting gap between the striplines and a sampling electrode, with an average power of ~80 mW. The collected current is converted to voltage by a preamplifier, and measured by a lock-in amplifier referenced to the chopper frequency.

laser-excited carrier lifetimes from 200 ps to as low as 0.5 ps for comparable doses, suggesting similar results are possible at 8 MeV.[18,19]

The goal of optoelectronic x-ray detection is to determine the time profile of the incident x-ray pulse, $X(t)$. What is measured is the sampling current versus x-ray/laser pulse time delay, $I_s(t)$ (Fig. 1). Making this current proportional to $X(t)$ requires the following: (1) The x-ray induced carriers in the stripline gap, which produce the shorting current, should have subpicosecond lifetimes. (2) The laser induced carriers in the sampling gap also should have subpicosecond lifetimes. (3) The propagating electrical pulse should remain undistorted by dispersion before being detected at the sampling electrode; this is readily achieved by minimizing the distance travelled (~100 μm).

Coplanar striplines were fabricated with CMOS compatible microfabrication processes on a semi-insulating GaAs wafer. X-ray pulses are generated at the Advanced Photon Source (APS), an x-ray synchrotron that operated at an 88 MHz repetition rate with nominal 50 ps (FWHM) pulses; x-rays were monochromated to an energy of 12 keV at the Sector 7-ID beamline.[20] Laser sampling was accomplished with a Ti:sapphire oscillator laser producing 800 nm pulses of ~100 fs duration. Synchronization of x-ray and the laser pulses is achieved by phase locking the 4th subharmonic of the 351.933 MHz rf signal that drives the electron bunches in the synchrotron to the laser, to produce output pulses at that frequency (87.9835 MHz). The phase error signal is fed back to a piezo motor that controls the laser cavity to stabilize the cavity length. To change the time delay, a delay generator (Stanford SDG 645) and a mechanical rf phase shifter (Colby PDL-100A, resolution 0.25 ps) are used to shift the phase of the synchrotron rf signal. The estimated jitter between x-ray and the laser pulses is about 1 ps.[21]

A side effect of implantation with 8 MeV protons is the transmutation of some of the target Ga and As nuclei into radioactive isotopes. In particular, ^{69}Ga transmutes into ^{69}Ge (half-life 39h), and ^{75}As into ^{75}Se (half-life 120d).[22] Initial activity determined by gamma spectroscopy found levels below 200 kBq. This corresponds to a negligible dopant density, less than 10^{12}cm^{-3}.

The measured sampling current for a standard semi-insulating GaAs substrate is shown in Figure 3 (top). A pronounced drop in the baseline current is observed for a duration consistent with the 50 ps x-ray pulse duration broadened by a carrier lifetime of ~200 ps. Note that in optoelectronic

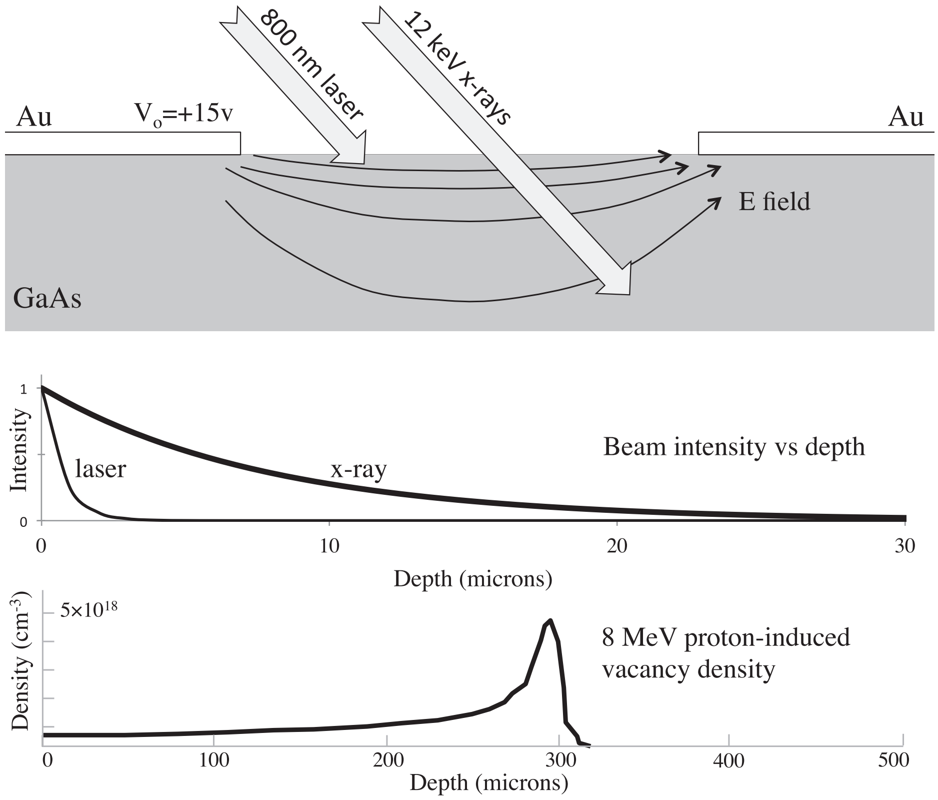

FIG. 2. Penetration depths into the GaAs substrate. Top: Cross-section view of GaAs substrate and the biased coplanar striplines, indicating the much greater penetration of 12 keV x-rays relative to the 800 nm laser light (not to scale). Middle: Exponential decay curves for the laser and x-ray radiation. Bottom: Vacancy density versus depth for 8 MeV protons, showing a nearly constant density for the top region exposed to x-rays.

measurements of laser pulses, the photocarriers are directly excited into the conduction band by the absorption of a single photon, whereas for x-rays there is a cascade of events following *K* shell absorption that leads to electrons in the conduction band. These data establish that the additional x-ray de-excitation processes still lead to conduction band dynamics similar to direct laser absorption, at least at picosecond time scales.

Figure 3 (bottom) shows the result of 8 MeV proton bombardment of the GaAs substrate. Note that there are now three separately resolved peaks in this time profile, each of them broadened somewhat compared to the nominal 50 ps (FWHM) gaussian synchrotron pulse width (although with low count rate limited statistics). This is very strong evidence that the proton bombardment has created trapping sites that significantly reduce the carrier lifetimes from the intrinsic value (~200 ps). Analysis of the actual electrode configuration provides an explanation of the three peaks. As shown in Figure 4, there are two impedance mismatches in the stripline circuit, where previous time domain reflectometry measurements show first a drop from $Z = 60\,\Omega$ to $40\,\Omega$ where the electrode width abruptly increases (and would produce a reflected pulse of the same polarity), and then a jump to $Z = 150\ \Omega$ from the wires bonded to the electrodes (which would produce a reflected pulse of opposite polarity). These mismatches are consistent with the small positive peak (labelled "b" in the figure) and the larger negative peak (labelled "c"), assuming a pulse group velocity ($v = c/n$) determined by an index of refraction $n = 4.2$.

Proton implantation in GaAs caused the x-ray induced sampling current to drop by nearly a factor of 10^{-4}, significantly greater than expected if the carrier lifetime were simply reduced by a factor of 10^{-2}; the small signals made

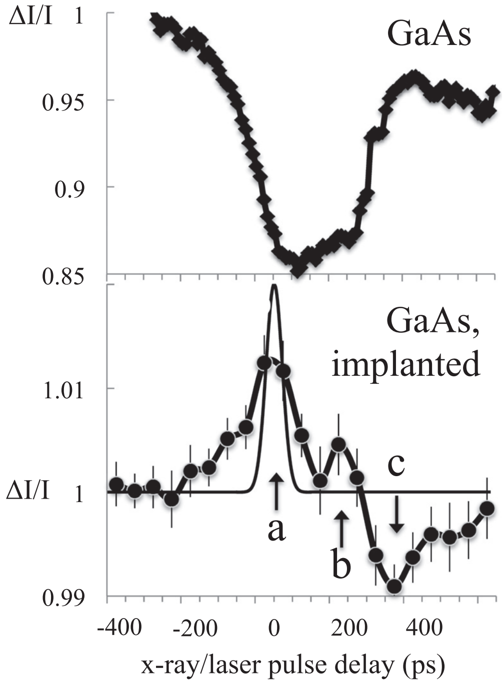

FIG. 3. Relative sampling current versus x-ray/laser pulse time delay. Top: Sampling current for semi-insulating GaAs, showing a pronounced signal (a drop from the baseline) caused by the x-ray generated voltage pulse passing the sampling gap. The width is consistent with 50 ps x-ray pulses convoluted with ~200 ps carrier lifetime in GaAs. Bottom: Results for GaAs after bombardment with 8 MeV protons. The baseline current is much reduced after bombardment; the absolute deviation from baseline is smaller by 10^{-4}. Three well-resolved structures are visible (a)-(c), corresponding to the x-ray induced voltage pulse and its reflections from impedance mismatches in the stripline. The bare line corresponds to the nominal 50 ps (FWHM) profile of the synchrotron pulses.

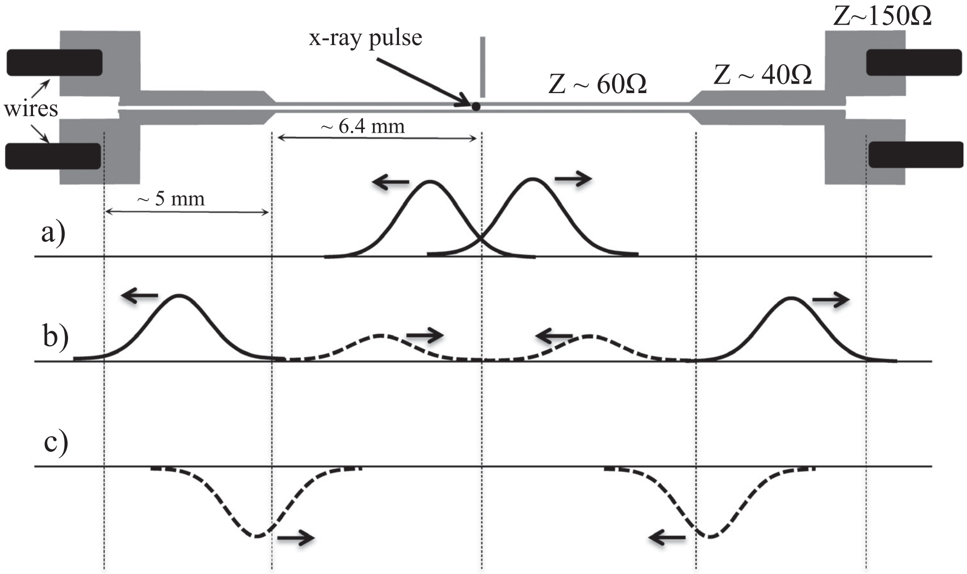

FIG. 4. Generation and reflection of voltage pulses in the coplanar stripline. Top: Layout of the coplanar stripline circuit (not to scale). The central region has a measured impedance of $Z = 60\,\Omega$, bracketed by regions of $40\,\Omega$ and $150\,\Omega$. (a) X-ray pulse excitation near the center of the coplanar stripline circuit (above) creates voltage pulses that propagate in both directions along the wave guide. (b) The initial pulse is partially reflected at the first impedance mismatch, with positive amplitude. (c) The pulse is then reflected at the second impedance mismatch with negative amplitude.

larger time step sizes necessary as well as the larger error bars. In standard optoelectronic measurements, the laser photon energy is only slightly greater than the band gap, so the direct promotion of a valence electron results in a conduction electron with little initial kinetic energy. When an x-ray is absorbed, however, the energetic photoelectron and subsequent Auger electrons produce electron-hole pairs with an average energy over 10 eV.[23] These electrons may also interact with the defect sites created by proton bombardment, and become trapped before they thermalize in the conduction band. This would effectively remove them from the carrier population, possibly accounting for the large suppression of current. This model needs to be tested with future studies of x-ray interactions with proton-implanted GaAs.

These results are a proof of concept demonstration that synchrotron x-ray pulses can be detected using optoelectronic techniques, and several avenues are apparent for improving on these results. The impact of reflected pulses can be minimized by increasing the length of the striplines and improved impedance matching on the device. The proton-induced loss of signal needs to be studied as a function of dose and perhaps annealing to remove certain defects. That problem might also be solved by switching from proton implanted GaAs to MBE-grown LT-GaAs or other thin film devices with shorter lifetimes, where greater efficiency may compensate for thicknesses less than the x-ray absorption lengths. With sufficient progress in these areas, measurements of picosecond x-ray time responses in laser pump x-ray probe studies "inside" the synchrotron pulse duration should become feasible, opening up a new ultrafast time domain for x-ray synchrotron sources.

We acknowledge Tom Miller, Shih-Chieh Liu, Jason Boomsma, Calvin Ebinger, and Tony Clevenger for assistance with the experiments. This research was supported by the U.S. Department of Energy, Office of Basic Energy Science, through Award No. DE-SC0004078. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under Contract No. DE-AC02-06CH11357. The PRIME Laboratory at Purdue University is supported by NSF Award No. 1153689-EAR.

[1] D. R. Grischkowsky, "Ultrafast THz photonics and applications," in *Springer Handbook of Lasers and Optics* (Springer, Berlin, 2012), pp. 1399–1459.

[2] M. B. Ketchen, D. Grischkowsky, T. C. Chen, C. C. Chi, I. N. Duling, N. J. Halas, J. M. Halbout, J. A. Kash, and G. P. Li, "Generation of subpicosecond electrical pulses on coplanar transmission-lines," Appl. Phys. Lett. **48**, 751–753 (1986).

[3] D. R. Grischkowsky, M. B. Ketchen, C. C. Chi, I. N. Duling, N. J. Halas, J. M. Halbout, and P. G. May, "Capacitance free generation and detection of subpicosecond electrical pulses on coplanar transmission-lines," IEEE J. Quantum Electron. **24**, 221–225 (1988).

[4] D. H. Auston, R. R. Freeman, P. R. Smith, D. M. Mills, and R. H. Siemann, "High-speed x-ray sensitive photoconducting detector," Appl. Phys. Lett. **42**, 1050–1052 (1983).

[5] A. Cavalleri, M. Rini, H. H. W. Chong, S. Fourmaux, T. E. Glover, P. A. Heimann, J. C. Kieffer, and R. W. Schoenlein, "Band-selective measurements of electron dynamics in VO2 using femtosecond near-edge x-ray absorption," Phys. Rev. Lett. **95**, 067405 (2005).

[6] L. X. Chen, "Probing transient molecular structures in photochemical processes using laser-initiated time-resolved x-ray absorption spectroscopy," Annu. Rev. Phys. Chem. **56**, 221–254 (2005).

[7] R. W. Schoenlein, S. Chattopadhyay, H. H. W. Chong, T. E. Glover, P. A. Heimann, C. V. Shank, A. A. Zholents, and M. S. Zolotorev, "Generation of femtosecond pulses of synchrotron radiation," Science **287**, 2237–2240 (2000).

[8] P. Emma, R. Akre, J. Arthur, R. Bionta, C. Bostedt, J. Bozek, A. Brachmann, P. Bucksbaum, R. Coffee, F. J. Decker, Y. Ding, D. Dowell, S. Edstrom, A. Fisher, J. Frisch, S. Gilevich, J. Hastings, G. Hays, P. Hering, Z. Huang, R. Iverson, H. Loos, M. Messerschmidt, A. Miahnahri, S. Moeller, H. D. Nuhn, G. Pile, D. Ratner, J. Rzepiela, D. Schultz, T. Smith, P. Stefan, H. Tompkins, J. Turner, J. Welch, W. White, J. Wu, G. Yocky, and J. Galayda, "First lasing and operation of an angstrom-wavelength free-electron laser," Nature Photon. **4**, 641–647 (2010).

[9] C. Rose-Petruck, R. Jimenez, T. Guo, A. Cavalleri, C. W. Siders, F. Raksi, J. A. Squier, B. C. Walker, K. R. Wilson, and C. P. J. Barty, "Picosecond-milliangstrom lattice dynamics measured by ultrafast x-ray diffraction," Nature **398**, 310–312 (1999).

[10] B. Ahr, M. Chollet, B. Adams, E. M. Lunny, C. M. Laperle, and C. Rose-Petruck, "Picosecond x-ray absorption measurements of the ligand substitution dynamics of Fe(CO) in ethanol," Phys. Chem. Chem. Phys. **13**, 5590–5599 (2011).

[11] F. E. Doany, D. Grischkowsky, and C. C. Chi, "Carrier lifetime versus ion-implantation dose in silicon on sapphire," Appl. Phys. Lett. **50**, 460–462 (1987).

[12] R. Ascazubi, I. Wilke, S. H. Cho, H. Lu, and W. J. Schaff, "Ultrafast recombination in Si-doped InN," Appl. Phys. Lett. **88**, 112111 (2006).

[13] R. Ascazubi, C. Shneider, I. Wilke, R. Pino, and P. S. Dutta, "Enhanced terahertz emission from impurity compensated GaSb," Phys. Rev. B **72**, 045328 (2005).

[14] A. C. Warren, N. Katzenellenbogen, D. Grischkowsky, J. M. Woodall, M. R. Melloch, and N. Otsuka, "Subpicosecond, freely propagating electromagnetic pulse generation and detection using GaAs-As epilayers," Appl. Phys. Lett. **58**, 1512–1514 (1991).

[15] R. Adam, M. Mikulics, A. Forster, J. Schelten, M. Siegel, P. Kordos, X. Zheng, S. Wu, and R. Sobolewski, "Fabrication and subpicosecond optical response of low-temperature-grown GaAs freestanding photoconductive devices," Appl. Phys. Lett. **81**, 3485–3487 (2002).

[16] P. Sharma, M. Bourgeois, D. Elmore, D. Granger, M. E. Lipschutz, X. Ma, T. Miller, K. Mueller, F. Rickey, P. Simms, and S. Vogt, "PRIME Lab AMS performance, upgrades and research applications," Nucl. Instrum. Methods Phys. Res. B **172**, 112–123 (2000).

[17] J. F. Ziegler, "SRIM-2003," Nucl. Instrum. Methods Phys. Res. B **219–220**, 1027–1036 (2004).

[18] M. B. Johnson, T. C. Mcgill, and N. G. Paulter, "Carrier lifetimes in ion-damaged GaAs," Appl. Phys. Lett. **54**, 2424–2426 (1989).

[19] M. Lambsdorff, J. Kuhl, J. Rosenzweig, A. Axmann, and J. Schneider, "Subpicosecond carrier lifetimes in radiation-damaged GaAs," Appl. Phys. Lett. **58**, 1881–1883 (1991).

[20] E. M. Dufresne, B. Adams, D. A. Arms, M. Chollet, E. C. Landahl, Y. L. Li, D. A. Walko, and J. Wang, "Time-resolved research at the Advanced Photon Source beam line 7-ID," AIP Conf. Proc. **1234**, 181–184 (2010).

[21] E. M. Dufresne, B. Adams, M. Chollet, R. Harder, Y. L. Li, H. D. Wen, S. J. Leake, L. Beitra, X. J. Huang, and I. K. Robinson, "A technique for high-frequency laser-pump X-ray probe experiments at the APS," Nucl. Instrum. Methods Phys. Res. A **649**, 191–193 (2011).

[22] V. A. Didik, R. S. Malkovich, E. A. Skoryatina, and V. V. Kozlovski, "Profiles of transmutation isotopes formed in solids by irradiation with charged particles: Formation, analysis and use," Nucl. Instrum. Methods Phys. Res. B **160**, 387–396 (2000).

[23] B. Ziaja, R. A. London, and J. Hajdu, "Ionization by impact electrons in solids: Electron mean free path fitted over a wide energy range," J. Appl. Phys. **99**, 033514 (2006).