Science “inside” the synchrotron pulse width: X-ray pump/optical probe cross-correlation study of GaAs

Proposal Title: Picosecond X-ray Detector for Synchrotrons
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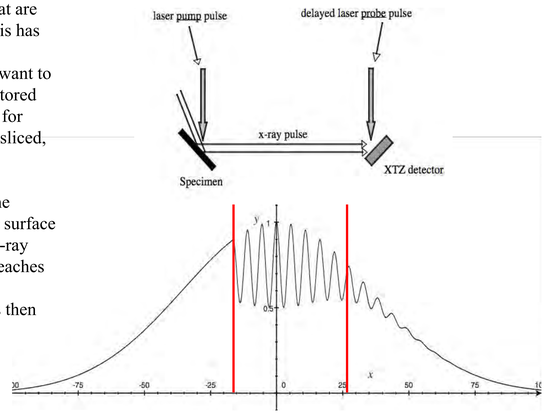
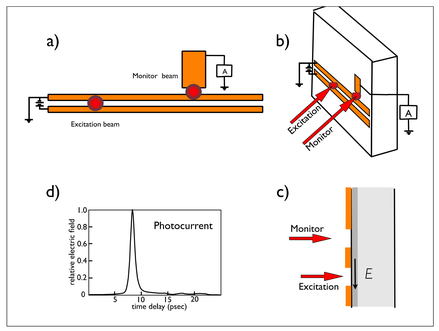
The challenge is to measure x-ray responses of materials that are much faster than the 100 ps FWHM synchrotron pulses. This has beed done with laser “slicing” techniques, especially at the Advanced Light Source and at the Swiss Light Source. We want to develop an alternative that does not require perturbing the stored particle beam. We are developing a semiconductor detector for pump/probe synchrotron experiments where the detector is sliced, instead of the particle beam.

Consider an x-ray pulse being reflected from a crystal. In the middle of the pulse duration, a 100 fs laser pulse strikes the surface and seriously perturbs the x-ray reflectivity. The reflected x-ray profile now carries a time-dependent response. When this reaches the detector, a time-delayed laser pulse triggers it, in effect allowing a single slice of the profile to be measured. This is then repeated as the pump/probe time delay is varied.

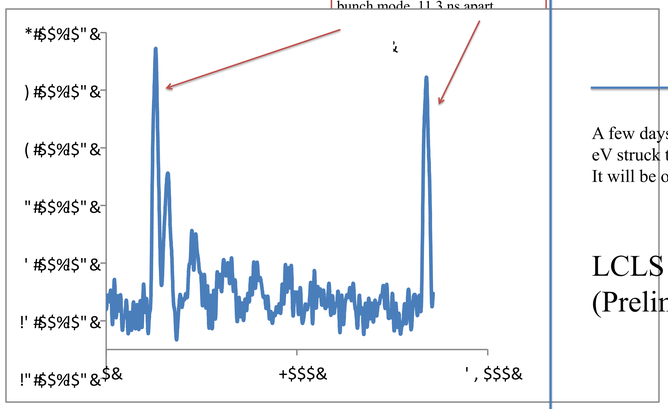




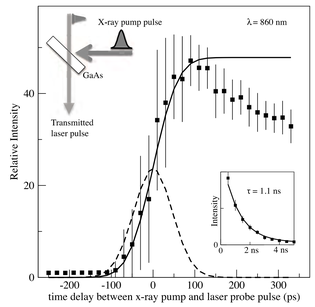
The detection scheme is based on a method for measuring laser pulse profiles for THz emitters (Fattinger & Grischkowsky APL 1988). A coplanar stripline is evaporated onto a high-resistance semiconductor substrate, and a DC bias potential is applied. The initial laser pulse excites an electrical pulse down the waveguide. A “slice” of this is detected when a monitor pulse briefly makes the gap to the pick-up electrode conducting. Sub-picosecond time resolution is possible when the carrier recombination lifetime is reduced by ion implantation or other means. We simple replace the initial laser pulse with the x-ray pulse reflected by the sample. The detector is triggered by a time-delayed portion of the laser pulse that excited the sample.

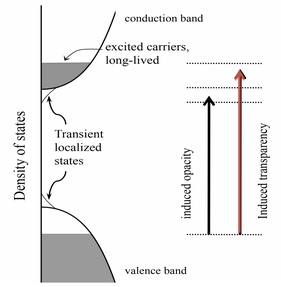
We have fabricated prototype detectors from semi-insulating GaAs. During initial tests with a focused hard x-ray beam at the APS Sector 7 undulator line, we simply measured the time-dependent voltage induced at the end of the coplanar stripline by connecting it to a high speed digital oscilloscope. The figure at right shows a 15 ns data set when the APS was in 324 bunch mode. These signals were readily obtained without RF amplification. The obvious ringing is largely due to the mounting structure for the device, which can be easily remedied. The spectrum below is the signal from all 324 bunches recorded in a single pass (i.e. 3.68 microseconds total).

The nest stage will incorporate the monitor laser coupled to a chopper and lock-in detection to permit operation in a full “slicing” mode.



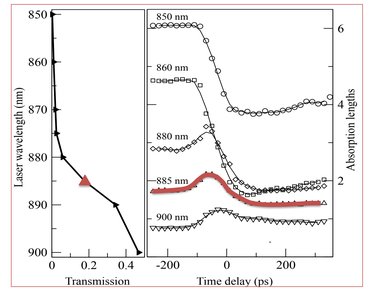
X-ray induced optical transparency

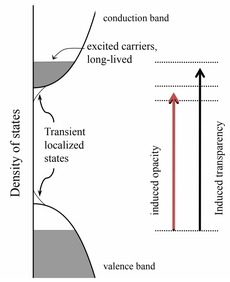




To demonstrate doing science “inside” the synchrotron pulse width, we exposed a thin GaAs crystal to a very intense focused “pink” beam at 12 keV, using the APS 14-ID-D beamline. This strong x-ray pump pulse was followed by a weak optical probe beam of 1.2 ps duration, at photon energies close to the semiconductor band gap. The measured transmission of this optical probe shows dramatic increases during and after x-ray illumination, *i.e.* x-ray induced optical transparency. The complex dynamical processes that follow hard x-ray absorption eventually leave a high density of electrons excited from the valence band into the conduction band. For photons at the band gap, the blocks all available final states, thus inducing transparency in an otherwise strongly absorbing crystal.

X-ray induced optical opacity



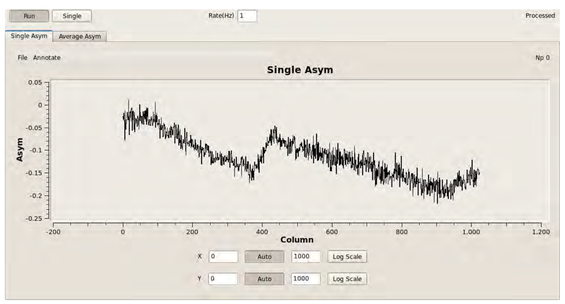


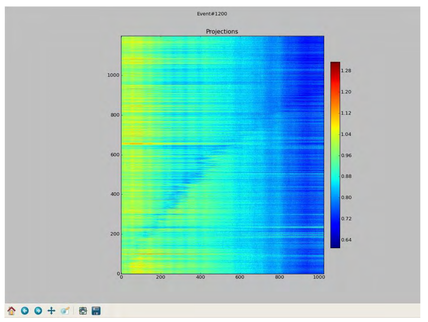
When we selected the laser photon energy to be slightly below the semiconductor band gap energy, the sample is normally very transparent to these laser pulses. As the 885 nm curve above shows, however, the absorption of these photons doubles while the x-rays are illuminating the crystal. The curve closely follows the instantaneous x-ray intensity, indicating that whatever process is involved here, it involves induced states that have lifetimes much shorter than the 100 ps x-ray pulse duration. For x-ray induced transparency, on the other hand, the response depends on the accumulated x-ray flux, because the excited electrons have lifetimes much longer than 100 ps.

The origin of these induced states causing the excess absorption is not yet clear, offering an interesting puzzle in the many-body response of semiconductors to hard x-ray absorption.

A few days ago (Aug 14) I joined Ryan Coffee *et al.* for 12 hours of LCLS beam time. Optical absorption through thin SiN membranes was recorded as ~10 fs LCLS pulses at 800 eV struck the specimen. Clear responses were seen in the raw data, which may lead to improved techniques for measuring the arrival time and pulse width of these ultra-short pulses. It will be of great interest to repeat the GaAs cross-correlation study with LCLS hard x-rays to observe the sub-picosecond response of induced opacity.

LCLS Pulse Length Study (Preliminary Results)





All 324-bunches, single pass: