Generation of Femtosecond Synchrotron Pulses from the Advanced Light Source

R. W. Schoenlein[1], H.H.W. Chong[1,2], T. E. Glover[3], P.A. Heimann[3], C.V. Shank[1,2], A. Zholents[4], M. Zolotorev[4]

[1]Materials Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory, University of California, Berkeley, California 94720, USA
[2]University of California Berkeley, Berkeley, California 94720, USA
[3]Advanced Light Source, Ernest Orlando Lawrence Berkeley National Laboratory, University of California, Berkeley, California 94720, USA
[4]Accelerator and Fusion Research Division, Ernest Orlando Lawrence Berkeley National Laboratory, University of California, Berkeley, California 94720, USA

INTRODUCTION

The application of x-ray techniques to understand fundamental atomic motion in condensed matter is a rapidly emerging area of research in chemistry, solid-state physics, and biology. The dynamic properties of materials are governed by atomic motion which occurs on the fundamental time scale of a vibrational period, ~100 fs. This is the time scale of interest for ultrafast chemical reactions, non-equilibrium phase transitions, vibrational energy transfer, surface desorption and reconstruction, and coherent phonon dynamics. To date, our understanding of these processes has been limited by lack of appropriate tools for probing the atomic structure on an ultrafast time scale. X-ray techniques such as diffraction and EXAFS yield detailed information about "static" atomic structure. However, the time resolution of high-brightness synchrotron x-ray sources such as the Advanced Light Source (~30 ps) is nearly three orders of magnitude too slow to directly observe fundamental atomic motion. Conversely, femtosecond lasers measure transient changes in optical properties of materials on a 10 fs time scale, but optical properties are only indirect indicators of atomic structure.

We report our first results using a novel scheme[1] to generate femtosecond pulses of synchrotron radiation. We directly measure femtosecond pulses in the visible synchrotron emission from bend-magnet beamline 6.3.2 at the ALS. This work is a first step toward developing a synchrotron-based source for ultrafast x-ray spectroscopy.

EXPERIMETNAL METHODS

The temporal duration of the synchrotron light pulses at the ALS is determined by the duration of the stored electron bunches, ~30 ps FWHM. The duration of the x-ray pulses may be reduced by more than two orders of magnitude by selecting radiation which originates from only a thin (~100 fs) temporal slice of an electron bunch[1]. Such a slice can be created through the interaction of a femtosecond laser pulse co-propagating with an electron bunch in an appropriate wiggler (Figure 1a). The high electric field present in the femtosecond laser pulse produces an energy modulation in the electrons as they traverse the wiggler (some electrons are accelerated and some are decelerated depending on the optical phase, ϕ). The optimal interaction occurs when the wiggler period λ_W satisfies the resonance condition $\lambda_L=\lambda_W(1+K^2/2)/2\gamma^2$ where γ is the Lorentz factor, K is the deflection parameter, and λ_L is the laser wavelength[1]. In addition, the far field laser radiation must overlap with the far field spontaneous radiation from the electron passing through the wiggler, and the laser spectral bandwidth (number of optical cycles per pulse, M_L) must match the spectrum of the fundamental wiggler radiation (determined by the number of wiggler periods, M_W). Under these conditions, the energy absorbed by the electron from the laser field (or transferred to the laser field), ΔE, is calculated by considering the superposition of the laser radiation and the spontaneous electron wiggler radiation[1]:

$$(\Delta E)^2 = 4\pi\alpha\hbar\omega_L \frac{K^2/2}{1+K^2/2}\frac{M_W}{M_L} A_L \cos^2\phi$$

where A_L is the laser pulse energy and ω_L is the laser frequency. We estimate that a 35 fs laser pulse with a photon energy of 1.55 eV, and a pulse energy A_L=100 μJ will produce an energy modulation amplitude ΔE~10 MeV, using the protein-crystallography wiggler, W16 (19 periods, λ_w=16 cm) at the ALS.

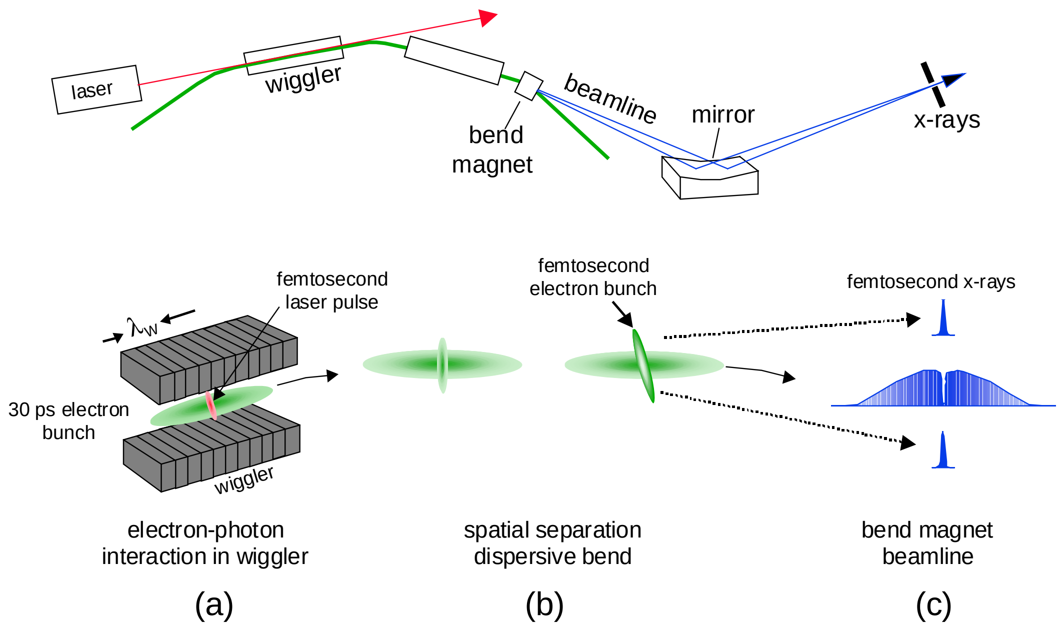

Figure 1. Schematic illustration of the technique for generating femtosecond synchrotron pulses. (a) laser/electron beam interaction in resonantly-tuned wiggler, (b) separation of accelerated femtosecond electron slice in a dispersive (bend) section, (c) generation of femtosecond x-rays at a band-magnet beamline.

The laser-induced energy modulation is several times larger than the rms beam energy spread at the ALS (1.2 MeV), and only a thin slice of the electron bunch (the portion which is temporally overlapped with the laser pulse) experiences this modulation. The accelerated and decelerated electrons are then spatially separated from the rest of the electron bunch (in a dispersive bend of the storage ring) by a transverse distance that is several times larger than the rms transverse size of the electron beam (Figure 1b). Finally, by imaging the displaced beam slice to the experimental area, and by placing an aperture radially offset from the focus of the beam core, we will be able to separate out the radiation from the offset electrons (Figure 1c). Since the spatially offset electrons result from interaction with the laser pulse, the duration of the synchrotron radiation produced by these electrons will be approximately the same as the duration of the laser pulse, and will be absolutely synchronized. Furthermore, the extraction of an ultrashort slice of electrons leaves behind an ultrashort hole or dark pulse in the core of the electron bunch (Figure 1c). This time structure will be reflected in the generated x-rays, and can also be used for time-resolved spectroscopy.

The average flux of the femtosecond radiation is defined by three factors: $\eta_1 = \tau_L/\tau_e$ = 35 fs/30 ps, $\eta_2 = f_L/f_{synch}$ =10 kHz/500 MHz, and η_3 = 0.2, where τ_L and τ_e are the laser pulse and electron bunch durations, f_L and f_{synch} are the laser and synchrotron repetition rates, and η_3 accounts for the fraction of electrons that are in the proper phase of the laser pulse to get the maximum energy exchange suitable for creating the large transverse separation. Accounting for the above factors, we estimate a femtosecond x-ray flux of ~$3x10^5$ photons/sec/0.1% BW at 2 keV ($5x10^4$ photons/sec/0.1% BW at 10 keV) from a bend-magnet beamline at the ALS with a 3 mrad x 0.4 mrad collection optic.

Synchrotron radiation damping provides for recovery of the electron beam between interactions. Since the laser interacts sequentially with each bunch, the interaction time is given by n/f_L where n is number of bunches in the storage ring. Furthermore, the bunch slice is only a small fraction of the total bunch. Thus, the storage ring damping time is more than sufficient to allow recovery of the electron beam between laser interactions (even for laser repetition rates as high as 100 kHz [1]) and other beamlines at the ALS can be operated at the same time without adverse effects.

A Ti:sapphire femtosecond laser system is located near beamline 6.3.2, and the laser pulses (~500 μJ, 60 fs pulse duration, 1 kHz repetition rate) are projected across the storage ring roof blocks to sector 5, where they enter the main vacuum chamber through a back-tangent optical port. Amplified femtosecond pulses co-propagate with the electron beam through wiggler W16 in sector 5. A photon stop/mirror following the wiggler reflects the laser light and the visible wiggler emission out of the vacuum chamber for diagnostic purposes. Images of the near field and far field wiggler emission are observed on a CCD camera, and the near and far field modes of the laser propagating through the wiggler are matched using a remotely adjustable telescope at the back tangent port. Timing between the laser pulse and the electron bunch is synchronized to better than 2 ps by phase-locking the laser-cavity repetition rate to the RF master oscillator for the storage ring. The spectrum of the laser is also matched to the fundamental wiggler emission spectrum. The efficiency of the laser/e-beam interaction is tested by measuring the gain experienced by the laser beam passing with the electron beam through the wiggler. The gain is a direct indication of the energy exchange, ΔE, between the laser and the electron beam and is equivalent to the gain that occurs in a free electron laser.

RESULTS

Femtosecond duration synchrotron pulses are directly measured by cross-correlating the visible light from bend-magnet beamline 6.3.2 at the ALS with the synchronized laser pulses in the nonlinear crystal, BBO. Figure 2a shows a laser synchrotron cross-correlation measurement on a long time scale. The measured pulse duration, σ=16 ps, corresponds to the overall electron bunch duration. Measurement with higher time resolution (Fig. 2b) shows the femtosecond "dark" pulse (264 fs FWHM, σ=112 fs) which appears as a narrow hole in the main pulse, and originates from the central core of the sliced electron bunch. Figure 2c shows a measurement of the femtosecond pulse (379 fs FWHM, σ=161 fs) originating from the spatial wings of the sliced electron bunch. An important point is that the femtosecond time structure will be invariant over the entire spectral range of bend-magnet emission from the near infrared to the x-ray regime, making this a very powerful tool for femtosecond spectroscopy.

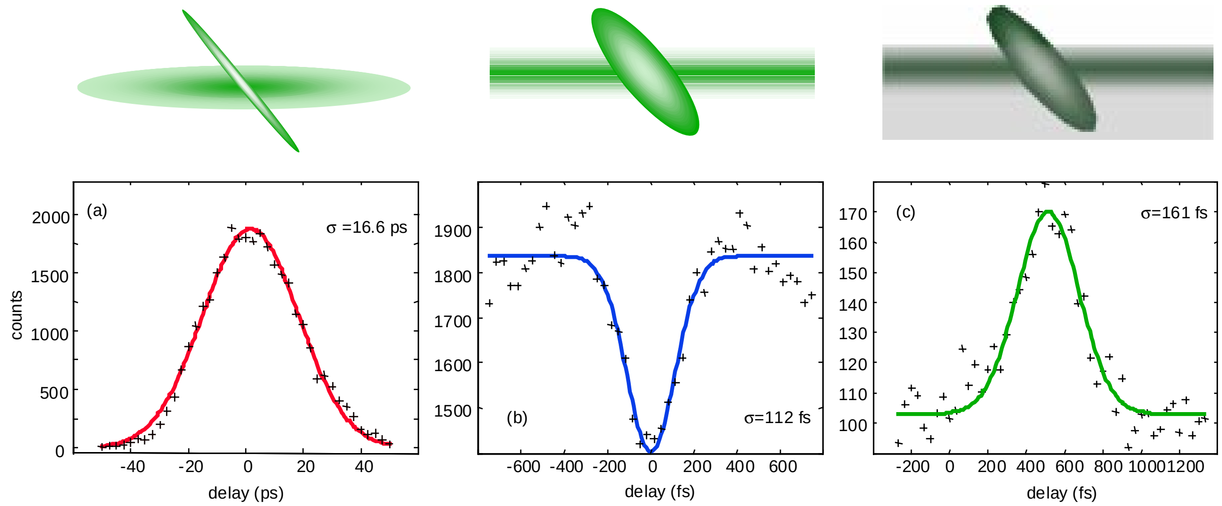

Figure 2. Cross correlation of visible synchrotron pulse with femtosecond laser pulse (and schematic illustration of the corresponding region of the electron bunch): (a) overall synchrotron pulse, (b) femtosecond dark pulse from on-axis radiation, (c) femtosecond pulse from off-axis radiation.

In conclusion, femtosecond x-ray pulses are generated from the ALS using a novel laser slicing technique. A pulse duration of ~300 fs is measured in the visible using cross-correlation techniques. The pulse duration is determined by the storage ring dispersion integrated from sector 5 (wiggler location) to the center bend magnet in sector 6. The limit on the pulse duration that could be generated from a bend-magnet in sector 5 is less than 100 fs. This technique also generates complementary "dark" femtosecond x-ray pulses, and is the first step toward development of a high-brightness femtosecond source for ultrafast x-ray spectroscopy.

REFERENCES

1. A. A. Zholents, M. S. Zolotorev, "Femtosecond pulses of synchrotron radiation," *Phys. Rev. Lett.*, **76**, 916-918 (1996).

This work was supported by the Director, Office of Science, Office of Basic Energy Sciences of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098

Principal investigator: Robert W. Schoenlein, Materials Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory.
Email: rwschoenlein@lbl.gov. Telephone: 510-486-6557.