Femtosecond Synchrotron Radiation Pulses Generated in the ALS Storage Ring

Scientists from the Lawrence Berkeley National Laboratory (Berkeley Lab) have generated 300-femtosecond pulses of bend-magnet synchrotron radiation at the Advanced Light Source (ALS) with the aid of a laser "time-slicing" technique. This technique allows an ultrashort portion of an electron bunch in the ALS storage ring to be spatially displaced in such a way that the synchrotron radiation from the displaced portion can then be collected separately. Their proof-of-principle experiment demonstrates that this technique is a viable one for producing ultra-short pulses of x rays. An ALS bend-magnet beamline is already under construction that will be dedicated to time-resolved x-ray diffraction, EXAFS, and other techniques capable of probing the long-range and local structure of matter on a femtosecond time scale. A proposed undulator beamline based on the same technique would further enhance the flux and brightness by orders of magnitude.

Via the making and breaking of chemical bonds and the rearrangement of atoms, the time evolution of condensed-matter structure occurs on the fundamental time scale of a vibrational period, or about100 fs. Atomic motion and structural dynamics on this time scale ultimately determine the course of phase transitions in solids, the kinetic pathways of chemical reactions, and even the efficiency and function of biological processes. A thorough understanding of such dynamic behavior is a first step to being able to control structural evolution, and it is expected to have important scientific applications in solid-state physics, chemistry, materials science, and biology.

X rays can provide the requisite structural information, and ultrafast x-ray science is an emerging field of research in which x-ray techniques are used in combination with femtosecond lasers to probe structural dynamics. However, the tremendous potential scientific impact of this research area is so far largely unfulfilled, owing to the lack of adequate sources. For example, the pulse length of a synchrotron x-ray source is limited by the bunch length of the electron beam in the storage ring. In particular, the bunch length at the ALS of about 30 ps is some 300 times to long to reach the 100-fs time scale. Shorter bunches are not feasible because it is not possible to simultaneously store high currents and ultra-short bunches for long times, owing to the effects of bunch-induced wakefields that give rise to bunch lengthening and other instabilities.

In early 1996, Alexander Zholents and Max Zolotorev of Berkeley Lab's Center for Beam Physics proposed the laser time-slicing technique as a way to achieve effective bunch lengths bounded by the laser pulse length, while avoiding the need for bunch compression and its accompanying difficulties. At the heart of the proposed technique is the use of a high-power, femtosecond laser that is synchronized with the electron bunches so that a pulse of laser light passes collinearly with an electron bunch through an undulator or wiggler. The high electric field of the shorter laser pulse modulates a portion of the longer electron bunch, with some electrons gaining energy and some losing energy. The condition for optimum energy modulation occurs when the laser wavelength matches the wavelength of fundamental emission from the insertion-device. Subsequently, when the energy-modulated electron bunch reaches a bend magnet (or other section of the storage ring with a non-zero dispersion), a transverse separation occurs. In a bend magnet, for example,, the high- and low-energy electrons are deflected through paths with larger and smaller radii of curvature, respectively, than the unmodulated portion of the bunch, resulting in a separation several times the rms transverse width (σ_x) of the electron bunch. A collimator or aperture selects the synchrotron radiation from the displaced bunch slices (see Figure 1).

Several factors limit the flux and brightness of femtosecond pulses to less than the normal flux and brightness of synchrotron radiation. Among these are that the slicing technique utilizes only a fraction of the electrons in a given bunch, and furthermore, not all the bunches are used since this technique operates at the repetition rate of the laser, which is much less than the frequency of electron bunches in the storage ring. Nonetheless, Zholents and Zolotorev estimated that the angular flux density obtainable at the ALS by their technique could provide femtosecond pulses with enough x-ray photons to be useful for experiments.

The pulse length of the femtosecond synchrotron radiation, while bounded by the length of the laser pulse, actually is somewhat larger. There is a pulse stretching that results from the different path lengths traveled by the electrons of slightly different energy in a slice as it progresses around the storage ring from the interaction region where it was created to the region where it generates synchrotron radiation. Zholents and Zolotorev calculated that 100 fs pulses should be possible.

To demonstrate the laser bunch-slicing technique, a team led by Robert Schoenlein was established with members drawn from the Berkeley Lab Materials Sciences Division, the Center for Beam Physics, the ALS, and the University of California, Berkeley (UC Berkeley). The team made use of the previously installed 16-cm-period wiggler that illuminates a protein crystallography beamline (Beamline 5.0.2), a test chamber on existing bend-magnet beamline (Beamline 6.3.2), and a high-power Ti-sapphire laser. They conducted their experiments with the ALS operating at 1.5 GeV, the wiggler gap adjusted to match the laser wavelength, and the laser operating at 800 nm with a pulse length of 100 fs and a repetition rate of 1 kHz. Following the interaction region, a mirror directed the fundamental spontaneous wiggler emission and the laser beam out of the storage ring for diagnostic purposes (temporal and spectral overlap, and spatial mode matching between the laser beam and the wiggler emission).

The femtosecond time structure of the synchrotron radiation was directly measured using cross-correlation techniques. Visible light (about 2 eV photon energy) from Beamline 6.3.2 was imaged onto a nonlinear optical crystal along with a delayed 50-fs pulse from the laser system. Photons at the sum frequency were counted as a function of delay between the modulating laser pulse (propagating through the wiggler) and the laser pulse used for cross-correlation. An adjustable knife edge located in the beamline at an intermediate image plane of the synchrotron radiation (before the nonlinear optical crystal) provided a means to select radiation from different transverse regions of the electron beam. Figure 2 shows a dark 300-femtosecond hole in the central cone of the synchrotron radiation and a bright 300-femtosecond peak in the wing of the synchrotron radiation (knife edge at $3\sigma_x$).

As the next step in the growing femtosecond x-ray science program at the ALS under the leadership of Schoenlein and Roger Falcone of UC Berkeley, a bend-magnet beamline (Beamline 5.3.1) is under construction with an anticipated completion date of May. The experiment hutch will have a chopper to block synchrotron radiation from electron bunches that are not modulated by the laser, a double-crystal monochromator, and slits to select the femtosecond pulses. As the radiating region is in the curved sector immediately after the Beamline 5.0.2 wiggler, the pulse stretching will be smaller than in the demonstration experiment. Performance goals include 100-fs pulses at a repetition rate of 5 kHz with a flux of about 10^5 photons/second/0.1% bandwidth and a brightness of about 10^8 photons/second/mm^2/mrad^2/0.1% bandwidth for photon energies up to 10 keV. A high-speed streak camera will also be available for time-resolved measurements. Initial experiments include time-resolved x-ray diffraction, EXAFS, and NEXAFS (XANES). A proposed undulator beamline would increase the flux and brightness by a factors of about 100 and 10,000, respectively, for photon energies up to about 6 keV and somewhat less for higher photon energies.

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References

1. R. W. Schoenlein et al., *Science* **287**, 2237 (2000).

Figure Captions:

Figure 1. Schematic diagram outlining laser bunch-slicing technique for generating femtosecond pulses of synchrotron radiation at a bend-magnet beamline.

Figure 2. Femtosecond synchrotron-radiation pulses measured by cross-correlation between a delayed laser pulse and the synchrotron radiation.(top) Synchrotron radiation from the central core of the electron bunch shows a dark femtosecond hole. (bottom) Synchrotron radiation from a wing of the electron bunch shows a bright femtosecond peak.