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# PUMP-PROBE AND OTHER TIMING EXPERIMENTS IN SYNCHROTRON RADIATION

by

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There are many transient, kinetic, intermediate-state and other time-dependent scientific phenomena that remain poorly understood. Intense undulator radiation (UR) from insertion devices in third-generation synchrotron radiation sources creates new possibilities for high energy pump-probe-timing research. We propose a VUV/SXR dual beam, two-color facility for (1) pump-probe-type experiments with continuously variable pump-pulse-to-probe-pulse interval, (2) harmonic phase-shift experiments that should achieve a time resolution of better than 1 ps, and (3) Michelson-type interferometric experiments, such as Fourier-transform stimulated-emission spectroscopy for wavelengths shorter than 1000 Å. The proposed beamline utilizes a pump beam of high-intensity UR from a 61-period undulator with 8-cm periods, and a probe beam of monochromatized synchrotron radiation (SR) from the following bending magnet. A unique optical variable delay unit (which also greatly reduces the higher-order content of the SR) is used to delay the arrival of the SR pulse at the crossing point of the two beams. The SR pulse may be delayed to arrive between 0.1 and 2.5 ns after the UR pulse. Because the UR pulse from the next electron bucket of the LBL ALS is emitted 2.0 ns later, delay of the SR pulse by 2.0 ns allows superposition of two pulses, and utilization of the coherence properties of UR.

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# PUMP-PROBE AND OTHER TIMING EXPERIMENTS IN SYNCHROTRON RADIATION

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There are many transient, kinetic, intermediate-state and other time-dependent scientific phenomena that remain poorly understood. Intense undulator radiation (UR) from insertion devices in third-generation synchrotron radiation sources creates new possibilities for high energy pump-probe-timing research. We propose a VUV/SXR dual beam, two-color facility for (1) pump-probe-type experiments with continuously variable pump-pulse-to-probe-pulse interval, (2) harmonic phase-shift experiments that should achieve a time resolution of better than 1 ps [1], and (3) Michelson-type interferometric experiments, such as Fourier-transform stimulated-emission spectroscopy [2] for wavelengths shorter than 1000 Å. As conceived for the Advanced Light Source (ALS), Lawrence Berkeley Laboratory, the proposed beamline utilizes a pump beam of high-intensity UR from a 61-period undulator with 8-cm periods, and a probe beam of monochromatized synchrotron radiation (SR) from the following bending magnet. A unique optical variable delay unit (which also greatly reduces the higher-order content of the SR) is used to delay the arrival of the SR pulse at the crossing point of the two beams. The SR pulse may be delayed to arive between 0.1 and 2.5 ns after the UR pulse. Because the UR pulse from the next electron bucket of the ALS is emitted 2.0 ns later, delay of the SR pulse by 2.0 ns allows superposition of two pulses, and utilization of the coherence properties of UR.

## 1. The beamline layout

As illustrated in fig. 1, the beamline begins with a U8.0-type permanent-magnet undulator in a 5-m straight section of the ALS, providing high intensity UR for the pump beam. The second color probe beam is SR from the following bending magnet, BM, which is collected along an axis deviated 6° clockwise from the UR centerline. At a distance of about 5 or 6 m from the BM center, a mirror system, $M_0$, is placed for collection and deviation of the SR probe beam by about 7.5° counter-clockwise, as well as for providing necessary entrance optical requirements for the SR monochromator.

The centerline of the deviated SR beam intersects the centerline of the UR beam at the 30 m point of the UR beamline, where an experimental chamber, $E_1$, is placed. Along the way, the SR beam traverses first the SR monochromator, designed to provide monochromatic radiation over the range 5–1000 eV, and second, a unique optical delay and higher-order-filter unit designed to vary the time of arrival of the SR pulse between 0.1 and 2.5 ns after the UR pulse while at the same time

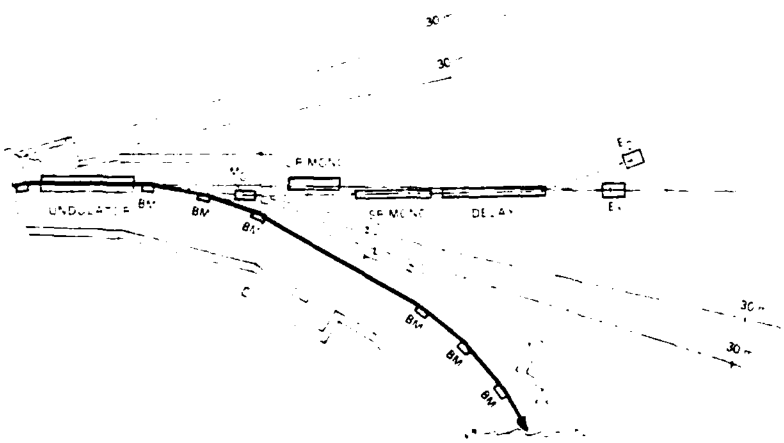

Fig. 1. Beam line layout. Abbreviations: UR ≡ undulator radiation; MONO ≡ monochromator; SR ≡ synchrotron radiation; $E_1$ ≡ experimental chamber #1; $E_2$ ≡ experimental chamber #2; BM ≡ bending magnet; $M_0$ ≡ deflection mirror for SR.

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greatly reducing its higher-order content. This device provides the timing control for pump-probe timing experiments.

An alternate experimental chamber, $E_2$, is located beyond and to the right of $E_1$ where experiments utilizing only SR may be conducted. Thus UR experiments and SR experiments may be conducted independently and simultaneously when the dual-beam capablity is not needed.

## 2. The time structure of the ALS

The ALS ring will have 328 electron buckets available [3]. If electrons are injected into only a single bucket, the SR or UR from any source point in the storage ring will be pulsed with a repetition frequency equal to the orbital frequency, 1.52 MHz. On the other hand, if all buckets are filled, the repetition frequency will be the rf frequency of the accelerating klystrons, 500 MHz.

The normal operation pattern is planned to be with 250 buckets filled, followed by 78 empty [3]. This gives a complex timing pattern with a fundamental frequency of 1.52 MHz. Within the 656-ns period, 250 light pulses, 2 ns apart and 35–55 ps wide, will be emitted into each beamline followed by 156 ns of "darkness". Other modes of operation of the ALS are possible, including the single-bunch operation that is needed for timing of slower processes.

The width of each light pulse is determined by several factors, including the peak electric field in the klystron, the rf frequency, and possible oscillations or perturbations of the electron orbit or of the individual bunches. It is predicted that the pulse width will be between 35 and 55 ps, depending on operating conditions [3].

Table 1
Tuning ranges for the U8.0 undulator

| | | |
|---|---|---|
| First harmonic: | 5.4– 220 eV | (usable intensity |
| Third harmonic: | 16.2– 660 eV | down to 3.6 eV |
| Fifth harmonic: | 27.0–1100 eV | while tuned to 5.4 eV) |

Table 2
Other characteristics of the U8.0 undulator

| | |
|---|---|
| Brightness range | $5.0 \times 10^{16}-1.2 \times 10^{18}$ photons/(s mrad$^2$ mm$^2$), 0.1% bw |
| Photon flux | |
| First harmonic: | $7.3 \times 10^{14}-3.4 \times 10^{15}$ photons/s, 0.1% bw |
| Third harmonic: | $1.5 \times 10^{13}-2.2 \times 10^{15}$ photons/s, 0.1% bw |
| Fifth harmonic: | $1.5 \times 10^{14}-1.8 \times 10^{15}$ photons/s, 0.1% bw |
| Average coherent power | |
| First harmonic: | 8 mw at 5.4 eV |
| | 160 mw at 100.0 eV |

Pump-probe timing experiments on ALS would thus have a temporal dynamic range of over $10^4$ (about 50 ps to 650 ns). Using other techniqus, the lower limit of timing should be less than 1 ps if the ALS electron bunch and its orbit are sufficiently stable.

## 3. Characteristics of the U8.0 undulator

Table 1 indicates the tuning ranges of U8.0. Other significant characteristics include those given in table 2.

Fig. 2 shows the tuning ranges of the U8.0 undulator in the first, third and fifth harmonics, plotted against the deflection parameter $K$ [4]. $K$ also determines the

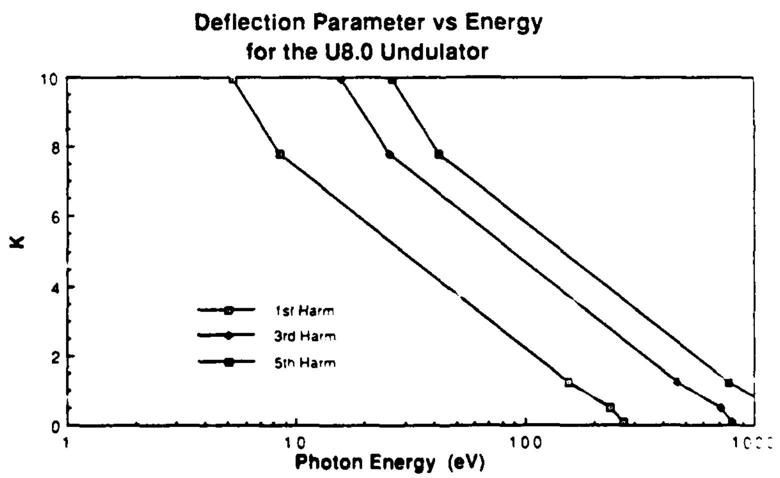


Fig. 2. Deflection parameter $K$ and tuning ranges for the U8.0 undulator.

spectral shape and breadth of the UR spectrum. The allowable tuning range is, for each harmonic, $9.9 \geq K \geq 0.7$. The central-lobe energy spread $\mathrm{d}E$ of unmonochromatized UR depends weakly on the harmonic number and strongly on the deflection parameter $K$. In general, $\mathrm{d}E$ varies inversely with $E$ and $K$. $\mathrm{d}E/E < 10\%$ at high photon energies, and $E > 50\%$ (asymmetrically tailing toward lower energies) at low photon energies. $K$, in turn, depends on the magnet gap, and therefore the photon energy selected.

The high flux, brightness and coherence of the U8.0 undulator of prime concern for the planned program of research in transient, dynamical and kinematic phenomena that are manifest in various valence- and core-level spectra within nanoseconds, picoseconds or less following stimulation or excitation (pumping). The pumping power available from the U8.0 undulator is adequate to enable experiments that otherwise would not be possible. Hence, in the selection of monochromators for use in the high-power UR beam, throughput efficiency is the prime concern, and resolving power is a secondary concern. Because of the very high total power emitted by the U8.0 undulator, as much as 4.9 kW at $K = 9.9$ [4], achievement of high resolving power in the monochromator will require new, untried cooling technology for the optical elements. Hence, in the current plan, resolving power will be sacrificed in the UR monochromator in favor of simpler, better tested power-handling technology, if necessary, so that the highest possible throughput efficiency is obtained.

## 4. Delay and higher-order-filter unit for the SR beam

The delay of the SR pulse is accomplished by increasing the optical path length, as illustrated in fig. 3. The four-mirror reflective design eliminates deviation of the outgoing beam, minimizes reduction of the maximum photon energy with increasing delay, as well as providing a sharp, low-pass transmission cutoff for higher-order rejection. All four mirrors have identical incidence angles. As delay is increased, mirrors 1 and 4 are rotated about their centers toward decreased incidence angle $\alpha$ while mirrors 2 and 3 are simultaneously rotated and translated downward. Thus, the incidence angles decrease from 86.6° to 75° as the delay is increased from 0.1 to 2.57 ns. The delay is tabulated below, along with the deviation angle $\theta$ and the height difference $H$. The values in table 3 are based on $M_0$ placement at 6.0 m from the bending-magnet center.

Table 3
Pulse delay for selected translations and rotations

| Delay (ps) | $\theta$ (deg) | $H$ (m) |
|---|---|---|
| 120 | 6.84 | 0.300 |
| 200 | 8.83 | 0.388 |
| 500 | 13.86 | 0.617 |
| 1000 | 19.37 | 0.879 |
| 1856 | 25.87 | 1.212 |
| 2000 | 26.76 | 1.261 |
| 2578 | 30.00 | 1.433 |

Without additional delay the SR pulse will arrive at the experimental point $E_1$ between 114 and 144 ps after the UR pulse, depending on the precise placement of $M_0$. Additional delay may be introduced by the optical delay unit at the experimenter's option.

For this $M_0$ placement, an additional delay of 1856 ps, added to the 144 ps delay intrinsic to the geometry, produces temporal superposition of the SR light pulse onto the UR light pulse that is emitted by the next following electron bunch (2 ns later). Spatial superposition of the two pulses occurs at the sample point in the experimental chamber $E_1$ where the effects of interference and coherence of the two beams may be studied in an experimental medium.

## 5. The UR monochromator

As shown in fig. 1, the UR monochromator placed at the 13-m point of the UR centerline. The primary concerns for the UR beam are: time structure, intensity, stability, and the reliability of the monochromator under the intense heat loading of UR. Optical resolving power is a secondary concern, and will be sacrificed in order to ensure accomplishment of the primary con-

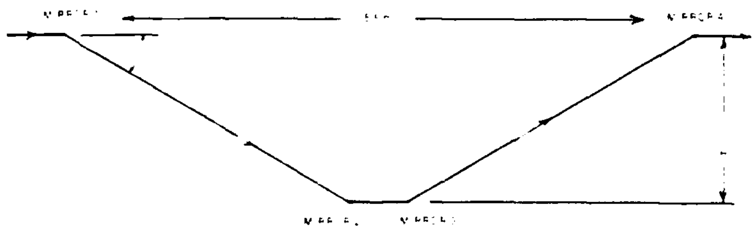

Fig. 3. Optical schematic of the four-mirror-optical delay unit.

cerns. At present, the best available monochromator technology meeting our primary concerns is the interference-enhanced (multilayer-coated) grating monochromator. Even so, the optical elements of the monochromator will be removable (automatically, under the experimenter's control) from the UR path, enabling the experimenter to utilize the full, raw power of UR when needed.

At higher energies, the undulator $K$ value is small enough that reasonable resolving power is intrinsic to the UR, and the monochromator may be unnecessary for many timing experiments. At lower photon energies, however, the large $K$ brings with it a broadened spectrum with much more energy in the harmonics, including the even-order harmonics, and the use of a monochromator will be necessary in most cases. Hence, the monochromator will be optimized for the lower energy range, and designed to provide mainly harmonic rejection when used at higher photon energies.

It should be noted that the survivability of the optical elements of any monochromator is not assured in the highest UR intensities expected. The maximum total power of the U8.0 undulator is projected to be 4.9 kW, with over 500 W in the high-brightness central portion of the beam [4]. The maximum power density is projected to be more than 1500 W/mrad$^2$. Development work at the Lawrence Berkeley Laboratory Center for X-Ray Optics, the Argonne National Laboratory Advanced Photon Source, and elsewhere is continuing, and progress on the heat-load problem is anticipated. However, stringent demands for optical figure stability in high resolution monochromators required to handle multi-kilowatt beams may not be met soon. At present, therefore, high resolution may be considered incompatible with the highest UR intensities.

A better solution of the problem of high resolution in undulator beams may be found eventually in vacuum ultraviolet and soft X-ray interferometric monochromators. Research in interferometric effects will be facilitated by the provision for superposition of UR and SR pulses in this beamline, coupled with the high degree of coherence available with third-generation undulator sources.

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