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Time structure measurement of the SSRF storage ring using TRXEOL method (postprint)

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Abstract

To achieve temporal alignment between the timing signal and the synchrotron X-ray pulse at the sample position in the time domain, measuring the temporal structure of the storage ring at the sample location within the experimental hutch is a fundamental step in time-resolved experiments utilizing pulsed synchrotron X-rays whose temporal structure is defined by the storage ring. In this work, the time-resolved X-ray excited optical luminescence (TRXEOL) method was designed and implemented to perform this measurement. This method is based on the principle of time-correlated single-photon counting techniques. The measurement system comprises a spectrometer equipped with a photomultiplier tube detector, a timing system, a set of nuclear instrumentation modules, and a zinc oxide luminescent material. The measurement was conducted at the X-ray absorption fine structure spectroscopy beamline of the Shanghai Synchrotron Radiation Facility. The results demonstrate that this method can measure the temporal structure of the storage ring with sub-nanosecond precision. The measurement system can also be applied to time-resolved studies of optical luminescent materials.

Full Text

Preamble

Time Structure Measurement of the SSRF Storage Ring Using TRXEOL Method

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Abstract: In time-resolved experiments using pulsed synchrotron X-rays, aligning the timing signal with the synchrotron X-ray pulse on the sample spot in the time domain requires accurate measurement of the storage ring' s time structure at the sample position inside the experimental hutch. This work designs and implements a time-resolved X-ray excited optical luminescence (TRXEOL) method for this measurement, based on the principle of time-correlated single photon counting techniques. The measurement system comprises a spectrometer with a photomultiplier tube detector, a timing system, a set of nuclear instrument modules, and zinc oxide as the luminescent material. Measurements performed on the X-ray absorption fine structure spectroscopy beamline at the Shanghai Synchrotron Radiation Facility demonstrate that this method can measure the storage ring' s time structure with a precision of less than 1 ns. The system can also be applied to time-resolved research on optical luminescent materials.

Keywords: Synchrotron ring time structure, X-ray excited optical luminescence, Time-resolved X-ray excited optical luminescence, Shanghai Synchrotron Radiation Facility

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INTRODUCTION

The 432-m storage ring of the Shanghai Synchrotron Radiation Facility (SSRF) has a time width of 1440 ns divided into 720 filling buckets [1], with each bucket containing a single electron bunch of less than 100 ps time width. Consequently, the width of X-ray pulses produced by these electron bunches along the beamline is less than 100 ps, and the shortest interval between two consecutive pulses is 2 ns. This time structure of pulsed X-rays enables numerous time-resolved experimental methods [2].

In time-resolved experiments [3], accurate measurement of the storage ring' s time structure at the sample spot inside the experimental hutch is essential for aligning the timing signal with the synchrotron X-ray pulse in the time domain, thereby obtaining the time structure of X-ray pulses produced by electron bunches. The conventional approach involves placing a fast photodiode at the sample spot [4]; each X-ray pulse generates an electrical pulse, and the time structure is acquired by recording these pulses in the time domain using an ultrahigh-performance oscilloscope.

In pump-probe nanosecond time-resolved luminescence experiments [5], a single X-ray pulse induced by a single electron bunch serves as the pump source, with electronic instruments detecting signals during the dark period of several hundred nanoseconds following the pulse. Since luminescent decays span a broad range from nanoseconds to milliseconds [6], nanosecond time resolution suffices

for such experiments. However, the SSRF Phase-II project proposes hundred-picosecond time-resolved experiments, requiring ring time structure measurement with resolution better than 100 picoseconds using an avalanche photodiode for X-ray detection [7].

This paper implements a time-resolved X-ray excited optical luminescence (TRXEOL) method [8] to measure the storage ring' s time structure. XEOL describes photon emission following synchrotron X-ray absorption [9]. In a TRXEOL cycle, the sample is excited by a single X-ray pulse, and emitted photons are detected and recorded before the next pulse arrives. The method employs time-correlated single photon counting techniques (TCSPC) [10]. The measurement system consists of a spectrometer with a photomultiplier tube (PMT) detector, a timing system, nuclear instrument modules (NIM), and fast optical luminescent ZnO material ($<$ 1 ns at 390 nm) [11, 12]. The ring' s time structure is indirectly measured by detecting and recording luminescent pulse timestamps. Measurements on the X-ray absorption fine structure spectroscopy (XAFS) beamline (BL14W1 [13]) demonstrate a system time resolution of $<$ 1 ns.

II. PRINCIPLE OF THE MEASUREMENT

A. Time Structure of the Storage Ring

As shown in Fig. 1 [Figure 1: see original paper], the SSRF storage ring features four filling patterns: single bunch, multi-bunch, hybrid, and full filling. Following the Phase-II project, SSRF will conduct 100 ps resolution pump-probe experiments in hybrid filling pattern, where a single electron bunch occupies a specific bucket while a series of multiple bunches fills consecutive buckets. The interval between the single bunch and multiple bunches can be adjusted by modifying the number and position of multiple bunches, with dark time gaps on both sides of the single bunch ranging from 2 to 1440 ns.

B. Interactions Between X-rays and Luminescent Material

XEOL is a complex process of interactions between X-rays and luminescent material [14]. As illustrated in Fig. 2 [Figure 2: see original paper], Step 1 involves creation of core holes and additional electrons in the conduction band within femtoseconds [8]. In Step 2, electrons from the valence band fill the core hole via X-ray fluorescence or Auger decay, generating additional valence band holes—also an ultrafast process. In Step 3, electrons in the conduction band and holes in the valence band from secondary processes recombine to produce optical luminescence.

TRXEOL monitors optical luminescent lifetime [15]. Since luminescent decays span lifetimes from nanoseconds to milliseconds [14], using a material with $<$ 200 ns optical luminescent lifetime as a sensor to detect luminescence from

samples excited by pulsed X-rays enables proper measurement of storage ring time structure, given that electron bunches have ~100 ps width and minimum inter-bunch intervals of 2 ns.

C. Theory of Luminescence Detection

In an excitation cycle, the X-ray pulse strikes the ZnO nanowire sample, which emits optical luminescence detected and recorded by the PMT and electronics. After numerous repeated cycles, the time structure is obtained indirectly through statistical analysis of optical luminescence decay data. This method is based on TCSPC technique.

TCSPC essentially measures luminescence lifetime. As shown in Fig. 3 [Figure 3: see original paper], the trigger pulse synchronized with the X-ray pulse indicates when the pulse hits the sample and serves as the START input for the time-to-amplitude converter (TAC). The PMT outputs an electrical pulse each time it detects a photon from the spectrometer, which feeds into the TAC' s STOP input. The TAC converts the interval between START and STOP pulses into a proportional voltage amplitude. In each excitation cycle, once the detector outputs a valid pulse, the multichannel analyzer (MCA) registers a single photon detection as a count in the corresponding time channel, ignoring pulse amplitude. After many excitation cycles, the MCA generates an array of photon detection numbers within short time intervals, yielding a photon arrival histogram that represents the sample' s luminescent decay curve.

For an MCA channel interval of $t_i+\Delta t$, the average photon number (ω_i) reaching the detector per interval must be less than one, as these are pulsed weak nuclear processes. The probability of z photons reaching the detector per interval follows Poisson distribution:

$$p_i(z) = \exp(-\omega_i)$$

Specifically:

$$p_i(0) = \exp(-\omega_i)$$

$$p_i(1) = \omega_i \exp(-\omega_i)$$

$$p_i(z>1) = 1 - p_i(0) - p_i(1) = 1 - (1+\omega_i)\exp(-\omega_i)$$

After N_E excitation cycles, N_i counts are detected in the ith interval:

$$N_i \sim N_E[p_i(1) + p_i(z>1)]$$

Since $\omega_i \ll 1$, $p_i(1) \sim \omega_i$ and $p_i(z>1) \sim \omega_i^2$, thus:

$$N_i \sim N_E[\omega_i + \omega_i^2] \sim N_E\omega_i$$

With N_E constant, counts in the ith interval are proportional to intensity in that interval $(t_i + \Delta t)$, making the MCA software histogram approximately equal to the theoretical optical luminescence curve.

III. THE MEASUREMENT SETUP

The measurement setup (Fig. 4 [Figure 4: see original paper]) comprises a spectrometer, timing system, NIM system, and ZnO material.

A. Timing System

The timing system generates high-precision trigger pulses synchronized with X-ray pulses at the sample spot to indicate when X-rays strike the sample. Designed using VME bus, FPGA, and high-speed serial communication technology, it includes a timing event generator (EVG) module, fanout modules, timing event receiver (EVE) modules, and an optical fiber transmission network [16]. The EVG reference clock source is the SSRF accelerator' s 500 MHz radio frequency (RF) clock. Timing events and trigger clocks are generated in the EVG and coded into serial frames according to 8b10b protocol for fiber network transmission. Fanout modules can duplicate and distribute these coded frames, providing triggers and clocks wherever the fiber network reaches. An EVE module is installed near the sample stage in the BL14W1 experimental station hutch.

The EVE receives timing frames from the fiber network, decoding them to obtain a 125 MHz clock synchronized with the 500 MHz RF clock. After frequency division processing, trigger pulses synchronized with X-ray pulses from the single electron bunch are obtained. Timing jitter is about 6 ps, and EVE delay step is 5 ps [17, 18].

B. Spectrometer System

The spectrometer collects photons from the sample and disperses them at different wavelengths. The PMT detects dispersed photons and outputs electrical pulses to the DAQ system. A 0.3-m focal length triple grating imaging spectrometer (Princeton Instruments Acton SP2358) was selected, fitted with three gratings: 1200 lines/mm blazed at 500 nm and 300 nm, and 150 lines/mm blazed at 500 nm. The main exit port accommodates a CCD (PI-MAX3). A Hamamatsu R928 PMT with 120–900 nm spectral response is attached to the spectrometer.

To reject PMT dark current, the experimental hutch must be dark during measurements, and the experimental area covered with thick aluminum foil.

C. NIM System

The NIM system measures XEOL lifetime via TCSPC by determining the time difference between timing pulses and PMT output pulses. It includes a fast timing amplifier (ORTEC-FTA820), constant fraction discriminator (CFD, ORTEC-935), time-to-amplitude converter and single-channel analyzer (TAC/SCA, ORTEC-567), multichannel analyzer (MCA, ORTEC-927), and ratemeter (ORTEC-661).

In the CFD, the signal splits into two branches: one inverted and delayed, the other attenuated to 20% of original amplitude. Delay time is selected via external cable equal to the time needed for the input pulse to rise from 20% to maximum amplitude [19]; in this measurement, delay time is 1.7 ns. The attenuated signal adds to the inverted delayed signal, forming a bipolar signal with zero crossing occurring at ~0.4 ns when the inverted delayed signal reaches 20% of maximum amplitude. The CFD' s zero-crossing discriminator detects this point and generates the corresponding timing output pulse, placing the timing point at 0.4 ns on the leading edge of the inverted delayed pulse with only tens of picoseconds timing jitter. To further reduce PMT dark current counts, the CFD threshold is adjusted until few signals appear with the photon shutter closed and good signals appear with it open.

The TAC generates a linear DC voltage proportional to the interval between the timing system' s trigger pulse and the CFD' s timing pulse.

D. The ZnO Sample

ZnO nanostructures are strong optical emitters used in fast X-ray detectors. ZnO' s wide bandgap (~3.37 eV) produces blue emission, and optical dynamics in ZnO nanowires show ~400 ps lifetime in band-gap transition [11], making it suitable for detecting storage ring time structure. The ZnO nanowire sample was provided by Professor SUN Xu-Hui of Soochow University.

IV. RESULTS AND DISCUSSION

A. XEOL Spectrum of the Sample

The fast luminescence decay center wavelength of ZnO must be known accurately to confirm spectrometer position and measure storage ring time structure. The DAQ system collected PMT output signals while scanning the spectrometer from 190 nm to 790 nm. The obtained spectrum (Fig. 6 [Figure 6: see original paper]) shows two luminescence centers: at 390 nm corresponding to ZnO bandgap luminescence, and at 500 nm corresponding to defect structure in the ZnO nanowire.

B. Timing Mode Test

Measurements were conducted on Beamline BL14W1 at SSRF with an X-ray beam spot of 0.3 mm × 0.3 mm. The storage ring operated in hybrid filling pattern with a 5 mA single bunch and 230 mA multiple bunches composed of 500 consecutive single bunches (Fig. 5 [Figure 5: see original paper]), providing 220 ns dark time gaps on both sides of the single bunch. X-rays were tuned to 9.7 keV, just above the Zn K absorption edge.

Two timing modes are possible: TAC started by XEOL signal and stopped by timing trigger (Mode 1), or vice versa (Mode 2). The trigger pulse rate far exceeds the luminescent signal pulse rate, with ratio depending on experimental efficiency and total measurement dead time. The two modes exhibit different efficiencies. Decay curves measured under identical conditions in both modes (Fig. 7 [Figure 7: see original paper]) show significantly better efficiency when XEOL signal serves as start and timing signal as stop, ensuring a stop signal corresponds to each start signal. Otherwise, the instrument remains continuously busy, increasing data acquisition dead time.

C. Luminescent Decay Curve at 390 nm Wavelength

With TAC range set to 200 ns and MCA channels to 16,384, MCA time resolution was ~12 ps. The spectrometer was positioned at 390 nm in inverted timing mode. Upper decay curves measured over 30 and 2 minutes are shown in Fig. 8 [Figure 8: see original paper]. The dynamic process' s full width at half maximum is ~500 ps, demonstrating that < 1 ns resolution can be achieved using ZnO nanowire to detect X-ray time structure, with signal-to-noise ratio improving through longer measurement times. Figure 8 also reveals impurity in the single bunch—a weak peak appears at 2 ns (see insets), induced by satellite weak bunches around the main single bunch. While such a bunch can track nanosecond lifetime decay events, it is clearly undesirable for sub-nanosecond and shorter events [20]. The SSRF accelerator physics group has confirmed this issue and begun cleaning the single bunch.

D. Time Structure of the Storage Ring

With TAC range set to 2000 ns (covering the 1440 ns full time scale) and MCA channels at 16,384, MCA time resolution was ~120 ps. The spectrometer was positioned at 390 nm in inverted timing mode. Results from 10-minute measurements (Fig. 9 [Figure 9: see original paper]) show single bunch width of ~500 ps, dark time gaps of 220 ns on both sides, and multiple bunch width of 999 ns—data identical to that provided by the SSRF accelerator physics group.

V. CONCLUSION

Measuring SSRF storage ring time structure at the sample spot in the experimental hutch is fundamental for time-resolved experiments using pulsed synchrotron X-rays. The conventional method employs a fast photodiode at the sample spot to sense and record X-ray pulses with timestamps. This paper proposes and implements a new TCSPC-based method using a spectrograph with PMT detector, timing system, nuclear instrument modules, and ZnO sample. Experimental results demonstrate achieved time resolution of < 1 ns. The measurement system can be further applied to pump-probe timing experiments and optical luminescent materials research.

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