Synchronizing fast electrically driven phenomena with synchrotron x-ray probes

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Time scales of long-range physical processes in solids are typically in the range of picoseconds to nanoseconds. These times are commensurate with the time resolution of structural probes based on modern synchrotron x-ray sources. Several processes of technological and scientific interest can be driven by applied electric fields, but synchronizing electrically driven phenomena with an x-ray probe poses a technical challenge. We describe the synchronization of a well-defined number of fast electrical pulses with the time structure of synchrotron x rays to probe the dynamics of thin films and nanostructures. This synchronization technique yields x-ray transient signals with 600 ps transitions in ferroelectric thin films, with a contribution of approximately 320 ps due to timing jitter in the synchronization. © *2007 American Institute of Physics.* [DOI: 10.1063/1.2668989]

I. INTRODUCTION

Synchrotron x rays can yield structural information with picosecond time resolution, which is an excellent match for a wide range of physical phenomena.[1–4] Experiments in which x-ray scattering probes the structure of materials driven by ultrafast lasers, for example, have resolved dynamical phenomena at time scales approximately equal to the bunch duration at electron storage rings and linear accelerators.[5,6] There is a continuous effort to synchronizing laser pulses with synchrotron x rays.[7–10] The electromagnetic field generated by laser pulses is, however, a poor match for the electrical or magnetic signals required to initiate events such as ferroelectric polarization switching or magnetization reversal.[1,3,11] These phenomena require electrical driving signals that must be synchronized with synchrotron x-ray pulses.

Synchronizing the electric fields applied to a thin film with short synchrotron x-ray pulses involves two instrumental challenges: (1) producing the electric field pulses driving the sample and synchronizing them with x-ray pulses, and (2) selecting the signal arising from x-ray pulses with a well-defined delay with respect to the electrical pulses. Since electrical devices such as ferroelectric capacitors often operate for a limited total number of polarization switching cycles, it is important to produce an accurate total number of electrical pulses and to collect scattering information only during these pulses.[12] This cannot be done using the existing approaches to synchronizing the laser pulses with synchrotron x rays since such approaches are based on locking the repetition frequency of the *continuous* laser pulses to the phase of the synchrotron radio frequency.[7–9]

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II. SYNCHRONIZATION

Electrons at the Advanced Photon Source (APS) of Argonne National Laboratory make a complete orbit around the storage ring in 3.678 μs. At the APS, the signal associated with the orbit is referred to as the revolution clock signal P0, which is distributed to experimental stations.[13] This signal can be used for synchronizing experiments with x-ray pulses. Synchrotron x rays are produced in short pulses with a full width at half maximum (FWHM) that is typically less than 100 ps, and techniques are being developed to lower this time to a few picoseconds and even into the femtosecond range.[10,14] Pulses are separated in time by up to a few hundred nanoseconds depending on the operation mode of the electron storage ring. In the standard operating mode of the APS, the x-ray pulse FWHM is $\sim$100 ps and there are 24 electron bunches equidistantly spaced in the ring resulting in 153 ns time between any two consecutive x-ray pulses.[15] Each bunch carries a nominal current of 4.25 mA.

Electrically driven phenomena can often be repeated only at rates lower than the rate at which x-ray pulses are produced by the synchrotron source. Electrical pulse rates, that are appropriate for experiments probing dynamics in ferroelectric or magnetic materials, are in the kilohertz regime, much lower than the x-ray pulse rates of 6.5 MHz or more. We thus designed our synchronization strategy so that only the signal resulting from a single electron bunch out of the 24 stored bunches contributed to the measured intensity [Fig. 1(a)]. The signal was further gated so that only scattering with a well-defined time relationship with respect to the electrical pulses contributed to the data.

A block diagram of the timing arrangement used to synchronize the electrical events with x-ray pulses is shown in Fig. 1(b). The measurement is started by triggering a pulse

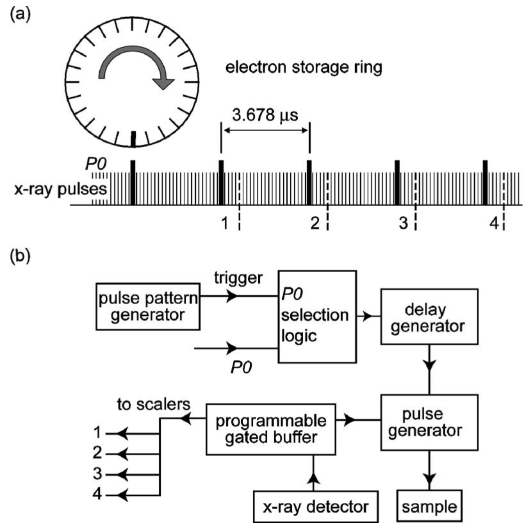

FIG. 1. (a) The sequence of x-ray pulses arising from a synchrotron radiation source. Four x-ray pulses associated with the orbit of single electron bunch are indicated with dashed lines. (b) A block diagram of the timing setup. The gated detection circuitry sends the diffraction signals arising from x-ray pulses 1–4 to separate channels of a scaler.

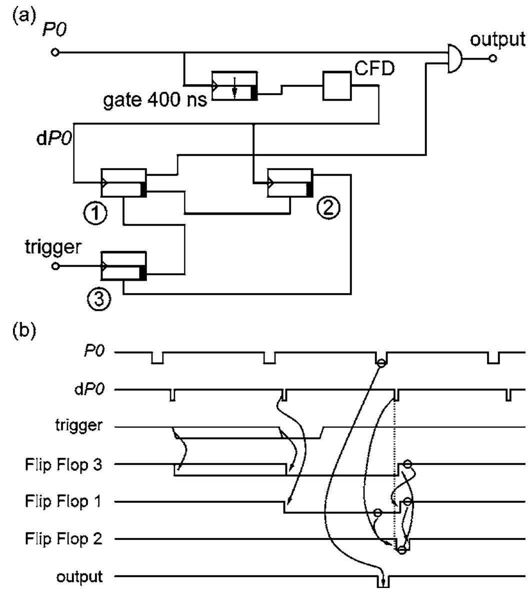

FIG. 2. (a) Schematic of the gating circuit to produce a pulse synchronized to P0 using a series of logical flip-flops. CFD is a constant fraction discriminator. The flip-flops are logical elements numbered 1–3. (b) Timing diagram of the P0 selection circuit. Note that branching the signals on the diagrams in Figs. 2 and 3 is accomplished using separate outputs of a logic translator (Phillips Scientific., Inc., model 7126) for P0 signals or the CFD for dP0 signals to avoid multiple triggering and signal reflections.

pattern generator to produce a sequence of pulses at the repetition rate of the electrical experiment; we have used rates from 0.5 to 10 kHz.

A series of logic gates produces pulses that mark the coincidence of the rapidly repeating signal of the storage ring orbiting period P0 and the pulses generated by the pattern generator. The output of this arrangement is a series of pulses at a low repetition rate that have a well-defined time relationship with the synchrotron x-ray pulses. Some degree of complexity is involved in generating these pulses because the P0 signal is tens of nanosecond long and it thus has a significant probability of colliding with the edge of the pattern-generator pulses. In a poorly designed logical arrangement, if a trigger pulse were to arrive *during* a P0 pulse, the resulting output pulse could be synchronized with the edge of the gate pulse rather than the edge of the P0 signal. This would result in the synchronization time uncertainties of up to the duration of the pulses used to distribute the P0 signal.

The challenge of synchronizing the electrical pulses with P0 accurately can be addressed by designing a pulse logic circuit to eliminate the possibility that the experiment is synchronized with the edge of the asynchronous trigger signal. We present here two circuits that accomplish this synchronization. In the first, a series of flip-flops assures that the output pulses are generated only at the edge of P0 rather than at the edge of the trigger signal. Figure 2 shows a schematic of the circuit and a timing diagram. In this approach, the P0 signal from the APS is AND gated with an enable signal that is guaranteed to become active long (400 ns) after the previous P0 pulse. The subnanosecond timing of the gated P0 signal is determined solely by the jitter of the P0 signal and that of the output AND gate (LeCroy, Inc., model 365AL). The enable signal is generated by three flip-flops (Philips Scientific, Inc., model 794) that go through a well-defined sequence of states following a trigger edge from the kilohertz signal generator. A gate and delay generator, configured as a monostable circuit, outputs a 400-ns-long gate pulse following a trigger by P0. Its trailing edge is detected by a constant fraction discriminator (Ortec, Inc., model 935) which outputs a delayed P0 signal (dP0). The exact timing of dP0 is not critical, as long as it is guaranteed to occur after the trailing edge of P0.

An edge in the trigger signal sets flip-flop 3 (FF3), which removes the reset signal from FF1. The next dP0 following this event sets FF1, which puts an enabling level at one input of the AND gate that forms the output signal. Because the output AND gate is enabled in response to dP0, P0 itself is already inactive. In the next cycle of the storage ring, just under 3.678 μs after the beginning of dP0, P0 becomes active again, and the AND gate outputs follows with low jitter. After P0 has become inactive, the next dP0 signal occurs, which sets FF2 (due to its reset signal having been removed by FF1). As soon as the FF2 is set, it resets FF3, which, in turn, resets FF1 resetting FF2. The circuit is thus reset completely and waits for the next kilohertz trigger.

Our implementation of the second approach to producing a low repetition rate signal with a well-defined time relationship to P0 uses two AND gates. A schematic of the circuit and a timing diagram are shown in Fig. 3. The first AND gate produces a long pulse that is formed at coincidence of the kilohertz trigger and P0. This long pulse is delayed relative to P0 as a result of the propagation delays of the AND gate electronics (Ortec, Inc., CO4020) and cables. The duration of this pulse is tuned to be longer than one period between P0 pulses and shorter than two of these periods. This guarantees that using this long signal as the gate pulse for the second AND gate produces an output signal synchronized with the clean edge of the second P0 signal. In comparison with the circuits more complex scheme shown in Fig. 2

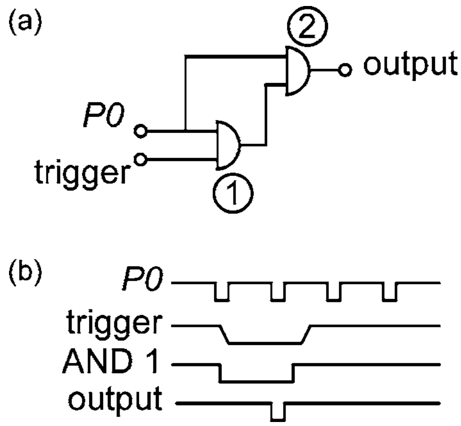

FIG. 3. (a) Schematic of a gating circuit producing an output pulse synchronized to P0 using two AND gates. (b) Timing diagram of the P0 selection circuit using two AND gates.

above, the approach shown in Fig. 3 depends on the propagation delay of the logic circuits, which may be undesirable in some experimental setups.

The gated P0 pulses produced by either of these arrangements in turn trigger a delay generator (Stanford Research Systems, Inc., model DG-535) which then triggers the pulse generator sending pulses to the sample. Varying the delay allows the electrical pulses to reach the sample with a fixed time relationship to x rays resulting from one of the 24 electron bunches. The relative timing of the x-ray probe and the electrical pulse can be adjusted by changing the delay.

The diffraction signals resulting from two adjacent x-ray pulses can be distinguished using an avalanche photodiode (APD) x-ray detector. This detector consists of an avalanche photodiode (EG&G Optoelectronics, Inc., model C30626F) biased to respond to single x-ray photons. Each 10 keV x-ray photon results in a current pulse consisting of approximately 10^3 electrons. A transimpedance amplifier converts these current pulses to voltage pulses with a gain of 5×10^3 V A^{-1} and a response time of less than 10 ns. The voltage pulses converted to digital signals using a constant fraction discriminator (Ortec, Inc., model 935) which produces an output pulse when one or more x-ray photons are detected. The APD detector is thus operated in a photon counting mode in which it reports either 0 or 1 photon for each x-ray pulse produced by the storage ring.

The detection circuitry is gated to select those x-ray pulses that arise from the electron bunch of interest. We have done this using a gated buffer implemented using a programmable digital signal processor. The buffer separates signals resulting from consecutive rotations of the electron bunch of interest into queue of four buffers. A similar function can be achieved using four channels of a conventional gate-delay generator.

The buffer is started after each of the trigger pulses used to generate electric field pulses. In the top up operating mode of the APS the current in each of the 24 electron bunches can be different because the filling scheme fills only a small fraction of the bunches at each top up event.[16] It is essential therefore to select x-ray pulses that are produced by the same electron bunch. To address this problem the software of the programmable buffer is designed to follow several subsequent rotations of the electron bunch of interest around the ring. The buffer then outputs pulses to separate channels of a scaler for each rotation of the electron bunch in which a photon was observed. Each event triggering the experiments results in the addition of either 0 or 1 for each of four bunches. After a number of repetitions, the accumulated signal in the scaler corresponds to the intensity in the four consecutive rotations of the same electron bunch around the synchrotron.

The buffer was programed to read the state of the detector for the same bunch exactly one orbital period before the trigger pulse and three rotations afterwards. The first channel of the scaler corresponds to the time preceding the electrical pulses to the sample. This channel is useful for intensity normalization as it represents a sample structure that is undisturbed by the electric field pulse. The three following bunch rotations probed the sample at time intervals of the orbital period.

The x-ray pulse duration sets the ultimate limit of time resolution for experiments using detectors with response times longer than the x-ray pulse duration. In the present case, however, the time resolution was limited by timing jitter and the rise times of the circuitry driving the sample rather than by the width of the x-ray bunches. Several triggering signals and instruments have jitter times that may affect the time resolution of the experiment. These include P0, the level translator copying the P0 signal, the logic AND gate, the delay generator, and the pulse generator. The total jitter time is the square root of the sum of the squares of individual uncorrelated jitter times, which emphasizes the contributions of a few dominant sources of jitter.

We have found that the electrical P0 signal alone has a jitter of 91 ps, which is comparable with the 100 ps FWHM duration of the x-ray pulses.[17] This jitter time can be reduced if the P0 signal is used only to phase lock a precision oscillator, such as a mode-locked laser, that serves as a high accuracy local copy of the P0 signal with jitter possible in the few-picosecond time scale.[5] The timing jitter of the synchronization circuit (including the jitter in the P0 signal itself) was measured using a series of arrival time spectra of electrical pulses that are applied to the sample relative to x-ray pulses detected by an ultrafast InGaAs pin photodiode. The total timing jitter measured in this way was approximately 320 ps. Using the arrangement described here, we have probed the piezoelectric response associated with applying electric field pulses to a ferroelectric PZT thin film with a time resolution of 600 ps (Fig. 4). This resolution is a convolution of 320 ps timing jitter, 300 ps rise time of the applied electric field pulse and the response time of the ferroelectric film.

III. SUMMARY

Synchronizing the synchrotron x-ray pulses with electrical signals results in time resolutions that are already comparable with the duration of single x-ray pulse. This capability will lead to the realization of new x-ray techniques that, for instance, can uniquely combine both x-ray microfocusing

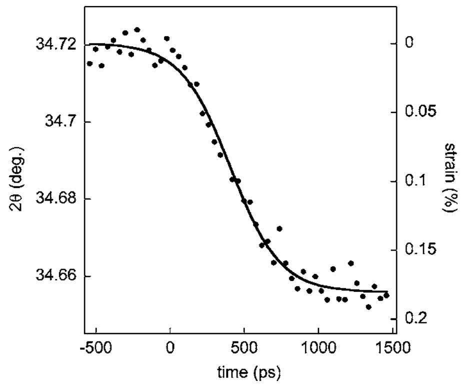

FIG. 4. The structural transient signal associated with the piezoelectric distortion of a $Pb(Zr,Ti)O_3$ thin film by a voltage pulse, after Ref. 1.

and ultrafast scattering to provide the basis for imaging fast transient processes with spatial resolution appropriate to resolve individual switching domains and nanostructures.[1] The scientific problems that can readily be addressed using the synchronization scheme described in this article include polarization switching in ferroelectric thin films, magnetization reversal, and heat dissipation in nanostructures.

ACKNOWLEDGMENTS

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