Subnanosecond piezoelectric x-ray switch

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We report an ultrafast piezoelectric switch for synchrotron x rays. A thin epitaxial film of piezoelectric Pb(Zr, Ti)O_3 works as a diffractive optical switch at frequencies from dc to >1 GHz. The broad frequency range allows single bunches of synchrotron x rays to be selected in an arbitrary sequence. The piezoelectric effect introduces mechanical strains of a fraction of 1% in the Pb(Zr, Ti)O_3 film, which can be used for blocking or passing diffracted x rays. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2219342]

Time-resolved synchrotron x-ray scattering investigations of the dynamics of solids and the transient structures of molecules in nonequilibrium states have recently become possible at the characteristic time scales of these phenomena.[1–4] The sequence of x-ray pulses generated by modern synchrotron radiation sources allows experiments to be synchronized with short x-ray pulses. For example, at the Advanced Photon Source (APS) of Argonne National Laboratory the full width at half maximum of a single x-ray pulse is less than 100 ps, which defines the time resolution of experiments.[5] This time resolution, however, can be used only with either a fast x-ray detector or an optical chopper that passes a single x-ray pulse. In many applications, the use of fast detectors is not acceptable due to the need to detect monochromatic x rays scattered in many directions simultaneously, or to resolve the energy of the detected photons.[6–8] In this letter, we show that piezoelectric lattice distortion of a thin film ferroelectric capacitor can be used to create a diffractive x-ray switch able to select a single synchrotron x-ray pulse for arbitrary synchrotron operation modes.

Mechanical choppers can isolate a single synchrotron x-ray pulse if the pulse is separated from others by a sufficient interval.[9,10] Only the most sophisticated choppers, such as those described by Lindenau *et al.*, can isolate bunches with submicrosecond separations.[11] In multibunch synchrotron operation modes the time between successive x-ray pulses ranges from 2 to 328 ns. At the APS, for example, pulse separations in 24, 324, and 1296 bunch modes are 153, 11.37, and 2.85 ns, respectively.[5] These modes are normally inaccessible for time-resolved experiments with all but the most advanced rotating mechanical choppers. In comparison with the diffractive approaches to isolating single bunches, mechanical choppers based on apertures have the advantage of functioning with polychromatic x-ray pulses, which are desirable in some experiments.

The idea to use the piezoelectric effect to modulate a beam of radiation is over 30 years old.[12] Originally, beams were modulated by a macroscopic translation of large optical elements by a piezoelectric ceramic. Recently acoustic standing waves in a piezoelectric crystal were used to modulate synchrotron x rays at time scales of tens of nanoseconds.[13] Our approach is to employ the piezoelectricity of a thin epitaxial Pb(Zr, Ti)O_3 (PZT) film to modulate the intensity of diffracted x rays.

The diffractive piezoelectric switch was a 300-nm-thick film of PZT, in the tetragonal crystal phase, between 100-nm-thick conducting $SrRuO_3$ (SRO) electrodes. The bottom electrode and PZT layers were deposited onto a single-crystal $SrTiO_3$ (001) substrate using off-axis sputtering.[14] The top electrode was a polycrystalline SRO film deposited through a shadow mask with 200 μm apertures. The device diameter of 50 μm was defined using focused-ion-beam lithography to isolate a region of the top electrode. An electric field was applied to the PZT film between the grounded bottom electrode and the top electrode, which was connected to the output of a pulse generator using an electrical probe. Synchrotron x rays of 10 keV photon energy from station 7-ID of the APS were forced to a submicrometer size spot by Fresnel zone plate optics. The efficiency of 160-μm-diameter Fresnel zone plate at 10 keV was about 7% with a focal length of 6.45 cm. The experiment was performed in the 24 bunch synchrotron operation mode, so x-ray pulses were separated by 153 ns. The diffracted x-ray beam was detected using an avalanche photodiode.[15] The detector electronics were gated to examine the signal arising from a single synchrotron bunch.

The piezoelectric switch was subjected to electrical pulses of 15 V amplitude and 10 ns duration. The timing could be adjusted to overlap the electrical pulse in time with the probing x-ray pulse (Fig. 1). The intensity and the position in reciprocal space of the PZT (002) Bragg reflection were recorded as a function of the delay time between the electrical pulse and the x-ray pulse. This Bragg reflection has its scattering vector in the direction of the applied electric field. The (002) Bragg reflection is presented in Fig. 1 as a function of scattering angle 2θ for three different time positions of the electrical pulse relative to the probing x-ray pulse. The angular width of these peaks is set by a combination of factors including the angular divergence introduced by the zone plate and the structural mosaicity of the PZT thin film. The intensity was normalized to the maximum at each time step. The PZT c-axis lattice constant increases in response to the electric field, and this lattice expansion shifts the Bragg reflection to a lower scattering angle.

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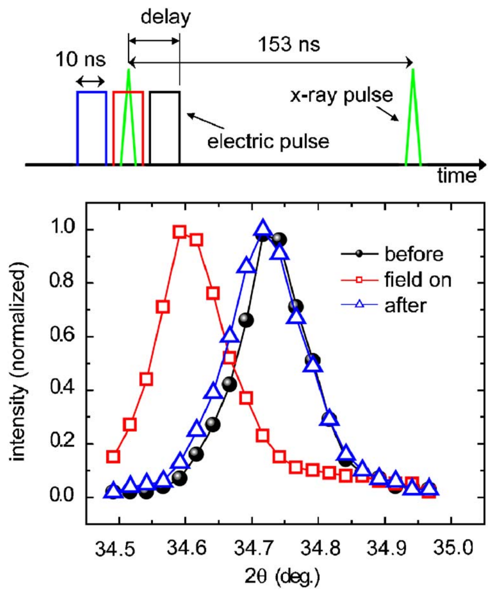

FIG. 1. (Color online) θ-2θ scans of the (002) Bragg reflection of PZT before, after, and during an electrical pulse applied to the piezoelectric switch. The timing diagram is shown in the upper part of the figure.

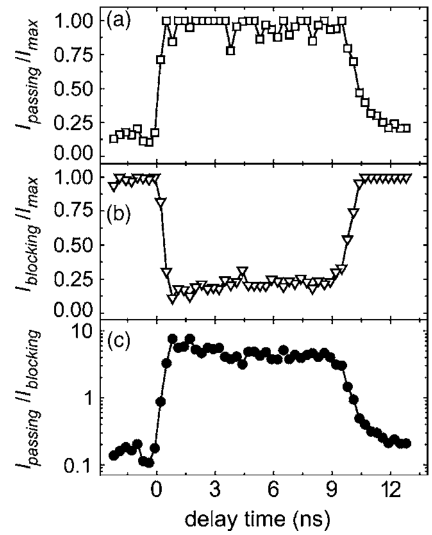

FIG. 3. Intensities $I_{\text{passing}}/I_{\text{max}}$ (a) and $I_{\text{blocking}}/I_{\text{max}}$ (b) for passing and blocking operation modes of the switch, as they are shown in Fig. 2, as a function of the delay time. I_{max} is the maximum intensity at each delay time. The intensity ratio $I_{\text{passing}}/I_{\text{blocking}}$ between the passing and blocking operation modes is shown in (c).

The scattered intensity as a function of the Bragg angle is shown in Fig. 2 in a range of angles covering the PZT (002) Bragg reflection. The intensity is again normalized to the maximum at each time point. The time interval in Fig. 2 covers the complete 10 ns electrical pulse. Turning on the electric field shifts the Bragg reflection to a lower scattering angle. When the field is turned off, the Bragg reflection returns to its initial position. The response time of the PZT *c*-axis lattice constant to the electric field is considerably shorter than 10 ns. The magnitude of the lattice constant distortion corresponds to 0.38% strain that is of a magnitude comparable to the highest strains reported for piezoelectric ceramics and within a factor of 5 of the largest reported for single crystals.[16–18] The high strain allows the piezoelectric effect to be used to block or pass an x-ray pulse depending on the selected scattering angle (Fig. 2).

The x-ray intensities at two fixed scattering angles corresponding to (002) Bragg peak positions of relaxed (I_{blocking}) and strained (I_{passing}) PZT are shown as a function of time in Figs. 3(a) and 3(b), respectively. Each time step is 300 ps, and it takes only two time steps, ~600 ps, to switch between the blocking and passing operation modes in response to the electrical pulse. This time is twice as large as the electrical pulse leading edge time of 300 ps. The difference is likely due to a combination of jitter in the timing circuit and the impedance mismatches between the capacitive load and the coaxial connection between the switch and the pulse generator. The switching occurs over 3 ns at the trailing edge of the electrical pulse; this reflects only the long trailing edge of the electric field pulse and not the response time of the piezoelectric switch. With a response time less than 1 ns the piezoelectric switch can isolate any single synchrotron x-ray pulse of any filling pattern at existing synchrotron radiation sources.[5]

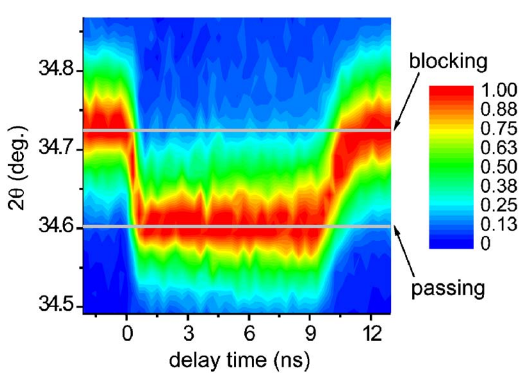

FIG. 2. (Color online) The intensity of diffracted x rays near the (002) Bragg reflection of PZT as a function of 2θ and time during an electrical pulse of 15 V with 10 ns duration. The intensity is normalized to the maximum at each step in time. Two fixed 2θ angles, marked with lines, correspond to the blocking and passing operation modes of the switch.

The effectiveness of the switch can be quantified by the evolution as a function of time of the intensity ratio $I_{\text{passing}}/I_{\text{blocking}}$ [Fig. 3(c)]. This ratio is limited by the divergence of x rays focused with the Fresnel zone plate optics, and can be significantly improved by reducing the divergence introduced by the focusing optics and by optimizing the piezoelectric switch scattering geometry and the PZT film thickness and structural perfection. To understand the roles of the crystalline quality and the device structure, we can estimate the contrast that would be observed using a structurally ideal PZT thin film with zero beam divergence. The thickness of the part of a perfect single crystal that contributes to reflecting x rays at the Bragg condition is defined by the extinction depth:[19]

$$\Lambda_{\text{ext}} = \frac{1}{4}\left(\frac{m}{d}\right)\frac{V_c}{r_0|F|},$$

where m is the order of the reflection, d is the lattice constant, V_c is the unit cell volume, r_0 is the classical electron radius, and $|F|$ is the unit cell structure factor. The extinction depth corresponding to the e^{-1} attenuation of the primary x-ray beam intensity of the (002) Bragg reflection of PZT at 10 keV photon energy is 350 nm. This defines the approximate optimum thickness for a diffractive switch. Increasing the thickness will also eventually limit the response time by increasing the time required for the piezoelectric deformation.

Neglecting the divergence of synchrotron x rays, the maximum possible ratio $I_{\text{passing}}/I_{\text{blocking}}$ for a film subjected to 0.38% strain is 6×10^4 as estimated from calculated Darwin curves for a perfect bulk PZT crystal.[20] This ratio will be influenced by the film thickness and epitaxial quality and by the magnitude of the piezoelectric strain. If fewer than every other bunch is used the contrast decreases as the fraction of the pulses used for the experiment decreases. With a 10 kHz experimental repetition rate at the APS, only 1 in 600 pulses is used and the total contrast is reduced by this factor.

The size of an unfocused synchrotron x-ray beam is typically a few mm^2. The *RC* response time of the switch increases linearly with the area and large-area switches are thus not acceptable for fast operations. By focusing on a small switch, for example, using Kirkpatrick-Baez mirrors, the total flux from each x-ray pulse can be increased. At the APS this should let 10^3 or more photons pass a 50 μm switch for each x-ray pulse. In experiments that require micron-size x-ray beams with low angular divergence the switch can be positioned between a focusing mirror and a sample. A 1 m focal length mirror can easily focus a 50 -μm-diameter beam at its input to 5 μm at the focal spot. At 50 cm from the mirror, the beam will have a diameter of 25 μm, which is small enough for the piezoelectric switch to be useful, although a large amount of the beam flux at the input of such a setup will be traded for a small beam size and higher x-ray intensity at the output. Further improvements of the contrast ratio can potentially be achieved by using higher order Bragg reflections [i.e., the (004) reflection of the PZT instead of the (002) reflection used in this work] or another ferroelectric material, for example, $Pb(Zn_{1/3}Nb_{2/3})O_3 - PbTiO_3$ (PZN-PT) that undergoes a field-induced phase transition.[21]

The repetition frequency of the electrical pulses applied to the piezoelectric switch can be at least between 0 and 10 kHz. At frequencies above 10 kHz we found that the PZT switch performance begins to be compromised by long time-constant charging of the PZT film. Similarly, operation at voltages higher than 15 V also lead to charging effects that offset the potential gain in contrast from higher strains. The piezoelectric switch can be synchronized with any nonperiodic experimental event, which is impossible to do with any of presently available synchrotron x-ray choppers and switches. We anticipate that the flexibility of operation and the subnanosecond response time of the switch could improve the performance of time-resolved experiments at synchrotrons and allow the use of operation modes previously inaccessible for time-resolved measurements.

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