Development of a new instrument to observe time-resolved neutron diffraction intensities in association with phase transitions

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Introduction

The study of time-resolved diffraction intensities during a phase transition is important for gaining an understanding of the kinetics involved. Unfortunately the flux needed in order to achieve good counting statistics for a diffraction experiment during a single phase transition is very high. This problem is then compounded if the time scales which need to be observed are very small. Therefore techniques which can take advantage of the reversibility of some phase transitions are potentially beneficial in order to build up statistics. Stroboscopic techniques have been used in recent times to observe diffraction peak intensities with very good time resolution during the application of some type of perturbing field, driving a transition. Most commonly, and as will be the case with this instrument, high electric fields are used to switch single-crystal samples between their paraelectric and ferroelectric phases. Diffraction peaks are observed as a function of time during the switching period [1]. Other types of cycles which have also been used are stress [2], and temperature [3].

In order to carry out these types of experiments, a new capability has been added to The Australian Stress Scanner (TASS) instrument [4] at the High Flux Australian Reactor (HIFAR) operated by the Australian Nuclear Science and Technology Organisation (ANSTO). TASS is the former Triple Axis Spectrometer which, within the last couple of years, has been significantly refurbished (including the introduction of a multi-line, position-sensitive detector) to function as a two-axis diffractometer for strain mapping in engineering materials. This capability will allow for such stroboscopic experiments to be performed using the switching of high-voltage electric fields. The exact operation of the device is outlined in the experimental details section below.

The common ferroelectric material, triglycine sulphate, will be used as a model crystal for the commissioning of the stroboscopic technique at HIFAR since, in a previous study involving stroboscopic diffraction intensity measurements at the ISIS [5], most unusual time dependencies for diffraction peak intensities from a TGS crystal in response to the switching of high-voltage electric fields applied to the crystal were observed.

Experimental Details

TASS is equipped with a 32 line, position sensitive detector (PSD). This detector covers a two theta angle of 2.24°, making it capable of observing an entire Bragg peak while stationary at any one position.

Additional hardware needed to be added to the instrument in order for the time-stamping of detector events to be possible, this is outlined in the schematic diagram of the experimental setup, shown in Figure 1.

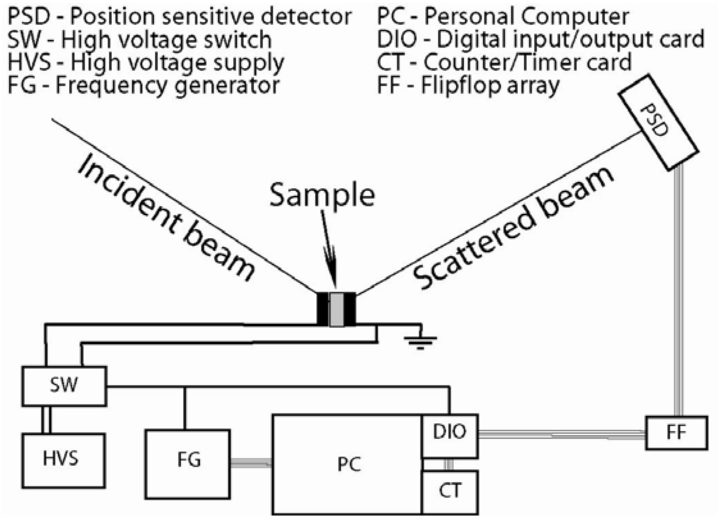

Figure 1: Schematic diagram of the hardware setup used to perform stroboscopic measurements.

During the collection of the stroboscopic data, two systems operate in parallel. Firstly, the Electric Field Application System (EFAS) is used to provide a user-defined electric field pulse to the sample. Secondly, the Data Acquisition System (DAS) is used to detect neutron events and time-stamp them.

The EFAS operates by outputting the user-defined pulse waveform from a frequency generator (FG). This FG output is then used to trigger a high-voltage switch (SW) which replicates the waveform but with the output high value now at the voltage set on the high-voltage power supply (HVS) (1-10kV). This field is applied across the sample as shown in Figure 1. The output of the FG is also passed back to the DAS and the DAS time is reset to zero on each rising edge of the pulse waveform.

The DAS operates by first passing the detector pulses through an array of flip-flops in order to transform each transistor-transistor logic (TTL) pulse from the detector into a change in digital line state, a requirement of the digital input/output (DIO) card. The DIO card then detects any change in state on its input lines. Each time a change in state occurs a request signal is sent to the counter/timer (CT) card. This request signal is stamped by the CT card with the number of clock pulses that have occurred on the internal counter of the CT card since time was set to zero by the EFAS. The internal counter on the CT card used during these experiments has an 80 MHz signal, thus allowing for a hardware timing resolution of 12.5 ns. However, experimentally the timing resolution is ultimately governed by the thickness of both the sample and detector and is of the order of 200 ns.

Software is then used to determine both the line and the time from the beginning of the last EFAS waveform at which detector events occurred. These results are binned in user-defined time frames, which can be as small or large as required.

Results

The time-stamping system has been tested using a neutron beam chopper (a spinning disk of neutron absorbing material with slits cut into it at regular intervals) to produce timed neutron pulses. During the test the chopper simply replaced the EFAS, with a photo-gate that produces a TTL-type pulse to reset the timing of the DAS to zero each time one of the slits passed through the neutron beam. In this test the PSD was aligned to a two theta value which corresponded to a Bragg peak from a steel sample. The incident beam was "chopped" and the time variations of detected neutrons recorded for a short period of time. Results from this test are shown below in Figure 2.

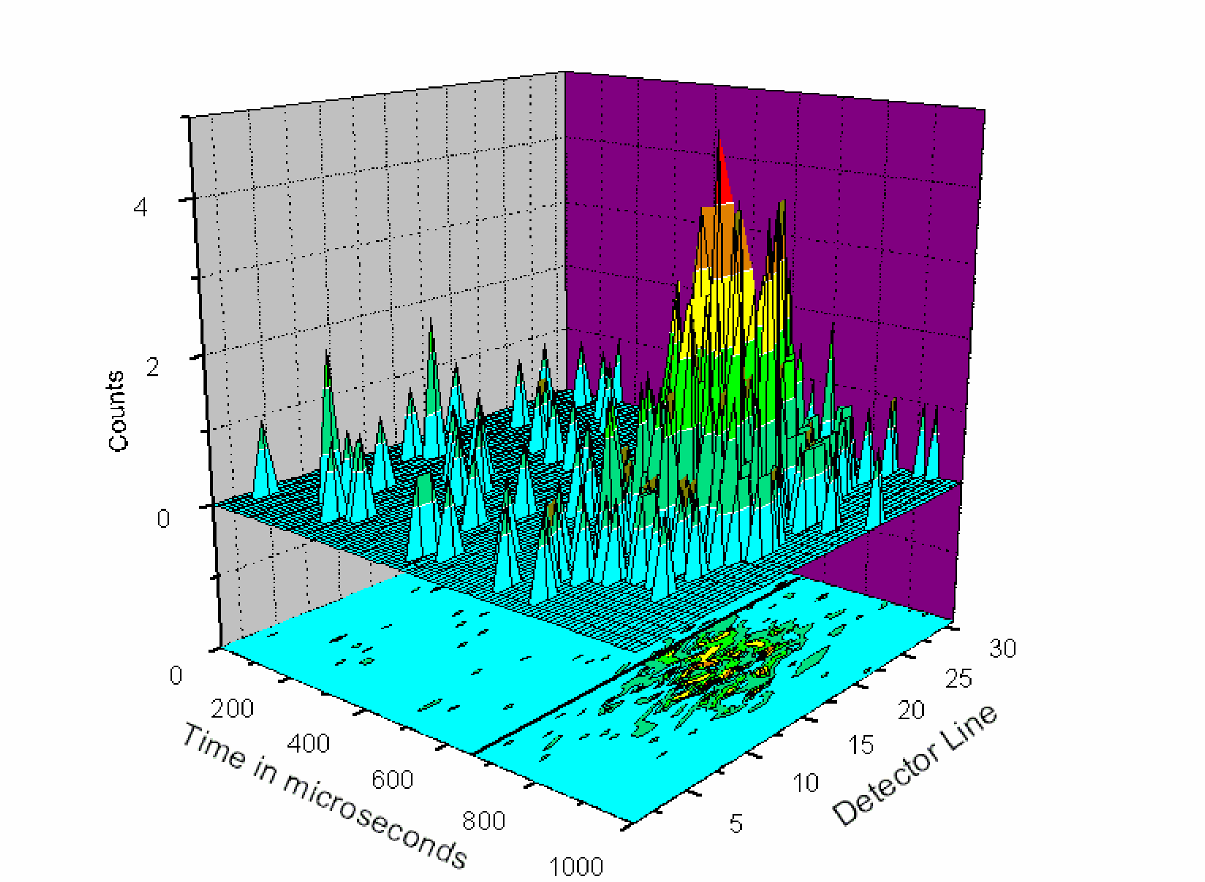

Figure 2: Bragg peak from a steel sample observed as a function of time using a pulsed neutron beam.

As can be seen in Figure 2, the Bragg peak is beginning to emerge approximately 650 μs after the slit in the chopper had passed through the neutron beam. For the 1.4 Å neutrons used during this experiment, this time delay corresponded to a flight distance of 1.8 m, approximately the distance from the chopper to the detector via the sample.

The system was also tested, observing the (060) Bragg peak of TGS, using a five second cycle time from the FG but with no high voltage applied to the sample. It is expected that because the sample is unperturbed during the time cycle that the Bragg peak intensity should remain constant, as was observed (Figure 3). Each five second cycle resulted in a total of approximately 500 counts. The data in Figure 3 show clearly how the counting statistics within each time frame can be added over a number of cycles. Also evident is the fact that the low signal-to-noise ratios for the PSD detector on TASS are maintained whilst operating in stroboscopic mode. The variations in the maximum peak height within each time bin are greater than the noise of the PSD. These larger variations have been attributed to the fact that the peak tips are spread across 2-3 detector lines, thus fitted peak curves would reduce these variation to within the natural noise of the PSD.

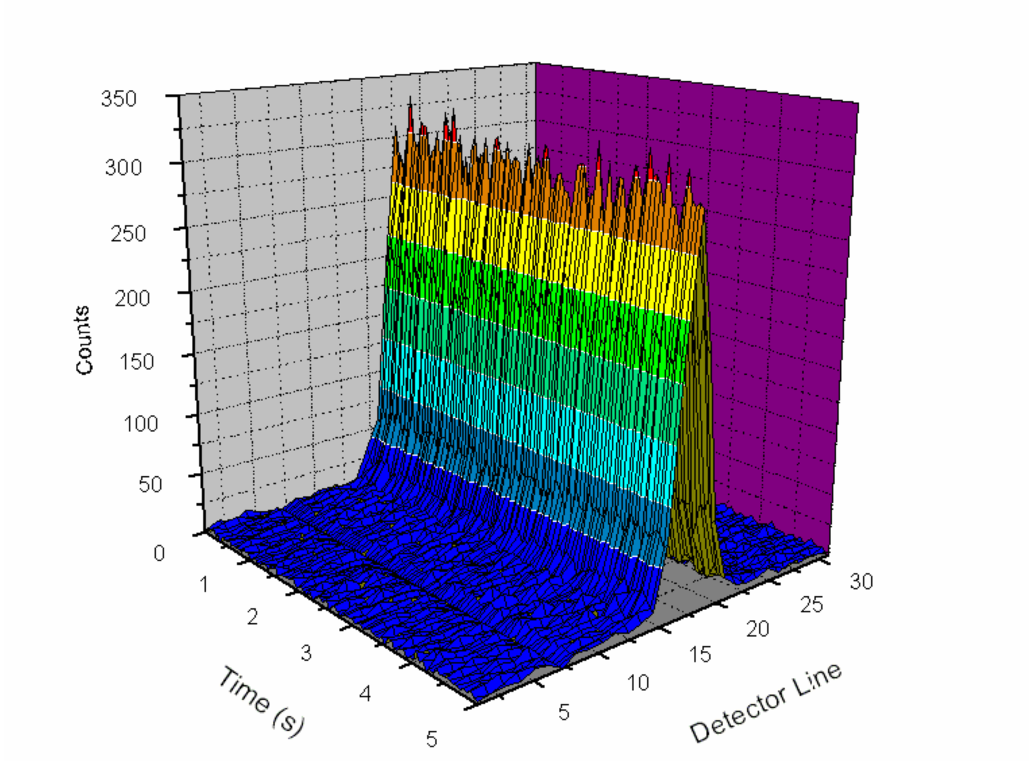

Figure 3: (060) Bragg peak from TGS, observed as a function of time, with no perturbing electric field.

Conclusion

The capabilities of a new instrument for the observation of time resolved diffraction intensities during a strobed electric field have been demonstrated. The apparatus will initially be used to observe the unusual relaxation effects seen in the ferroelectric TGS.

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