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TIME RESOLVED TECHNIQUES: AN OVERVIEW

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Time resolved techniques: an overview*

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ABSTRACT

Synchrotron sources provide exceptional opportunities for carrying out time-resolved x-ray diffraction investigations. The high intensity, high angular resolution, and continuously tuneable energy spectrum of synchrotron x-ray beams lend themselves directly to carrying out sophisticated time-resolved x-ray scattering measurements on a wide range of materials and phenomena. When these attributes are coupled with the pulsed time-structure of synchrotron sources, entirely new time-resolved scattering possibilities are opened. Synchrotron beams typically consist of sub-nanosecond pulses of x-rays separated in time by a few tens of nanoseconds to a few hundred nanoseconds so that these beams appear as continuous x-ray sources for investigations of phenomena on time scales ranging from hours down to microseconds. Studies requiring time-resolution ranging from microseconds to fractions of a nanosecond can be carried out in a triggering mode by stimulating the phenomena under investigation in coincidence with the x-ray pulses. Time resolution on the picosecond scale can, in principle, be achieved through the use of streak camera techniques in which the time structure of the individual x-ray pulses are viewed as quasi-continuous sources with ~100 - 200 picoseconds duration. Techniques for carrying out time-resolved scattering measurements on time scales varying from picoseconds to kiloseconds at present and proposed synchrotron sources are discussed and examples of time-resolved studies are cited.

2. INTRODUCTION

Synchrotron X-ray sources have had a major impact on x-ray scattering research through their unique combination of high photon fluxes, high degree of polarization, high angular collimation, wavelength tunability, and pulsed time structure. Of particular importance with respect to time-resolved measurements are the high photon fluxes and the pulsed time structure. The high fluxes have made it possible to investigate a wider range of dynamic phenomena on time scales varying from milliseconds to kiloseconds and the pulsed time structure has made it possible to extend time-resolved measurements into the microsecond and nanosecond ranges by synchronizing dynamic phenomena with the synchrotron x-ray pulses. The average photon fluxes available from synchrotrons are ~10^4 higher than those available using conventional sources, and the concentration of x-rays into ~150 picosecond pulses (with separations of several hundred nanoseconds) makes the instantaneous flux an additional ~10^3 times higher than the time average flux. This concentration of synchrotron x-rays into short bursts has made it possible to carry out time-resolved x-ray measurements on the nanosecond scale, and it also makes it feasible to consider the application of streak camera techniques to carry out picosecond resolution measurements within the x-ray pulses.

In this paper, techniques for carrying out time-resolved x-ray scattering on time scales varying from kiloseconds to picoseconds will be discussed, and examples of investigations will be cited. As the fields for which time-resolved investigations on synchrotrons are of interest range over biology, macromolecular crystallography, geology, materials science, fundamental physics, polymers, chemistry, etc, it is not feasible to consider work performed in all areas. Rather we will discuss general modes of measurement and discuss examples of experiments demonstrating the measurement techniques. Of particular interest will be the microsecond and shorter time range where the pulsed nature of synchrotrons offers fundamentally new opportunities.

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LARSON

3. TIME DOMAINS

Taking the synchrotron fill lifetime as an upper limit on time resolution and considering the timing jitter of individual synchrotron bunches as the lower limit, synchrotron sources provide for time resolution covering the range from tens of kiloseconds to a few picoseconds. This range corresponds to more than 15 orders of magnitude, and while incident beam fluxes, scattering rates of samples, triggerability of the phenomena, and detector capabilities ultimately determine the feasibility of individual experiments, unprecedented opportunities exist for time-resolved x-ray scattering measurements using synchrotrons.

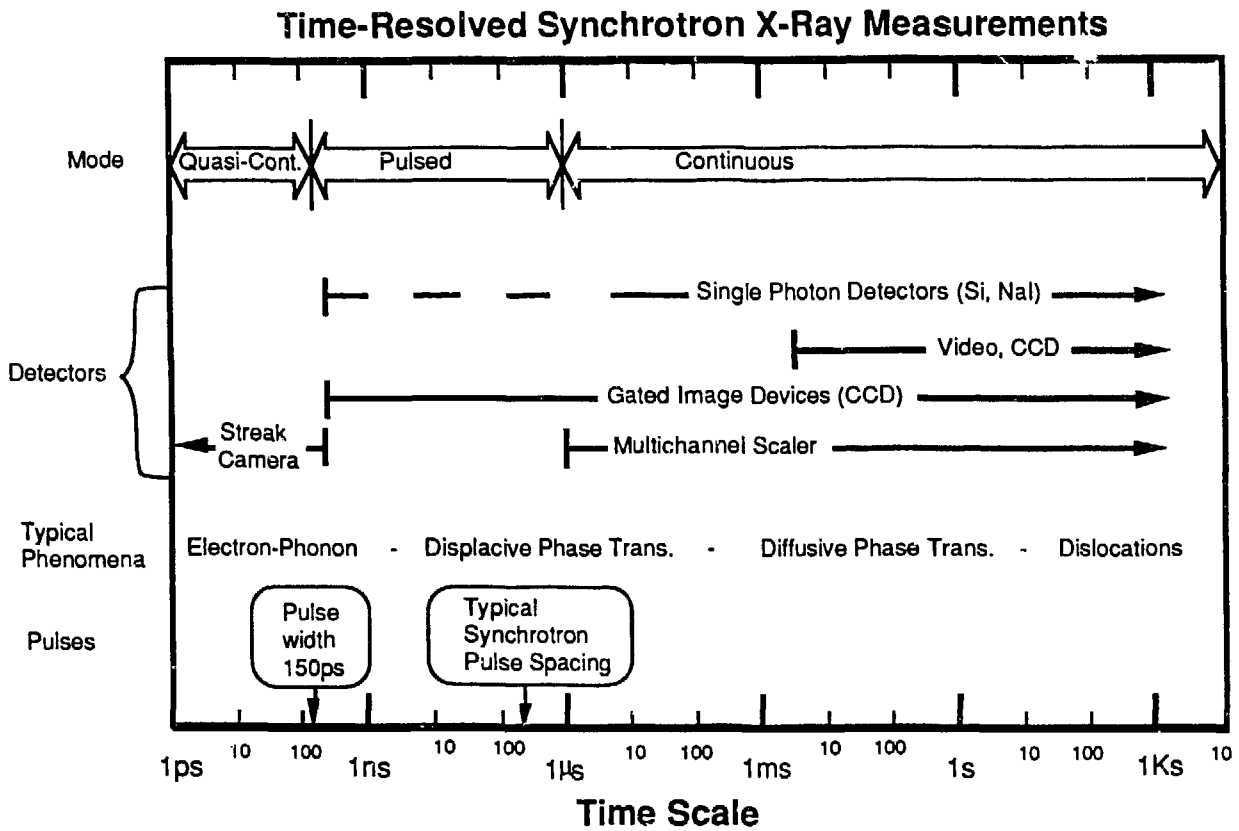


Fig. 1. General overview of time scales and time-resolved techniques associated with time-resolved synchrotron x-ray measurements.

Fig. 1 shows a composite sketch of time-resolved techniques as a function of the measuring time scale. At the longer time scales (milliseconds - kiloseconds) synchrotron beams can be considered continuous and measurements can be performed using arbitrary time slices without regard for the pulsed nature of the source. For these time scales, time-resolved techniques using synchrotron sources are similar to those using conventional x-ray sources; however, the advantages of higher incident beam fluxes, wavelength tunability, and angular collimation are, of course, present. For the shorter time scales (picoseconds - microseconds) the pulsed time structure of the incident x-rays becomes important and measurements must be synchronized with the arrival time of individual x-ray pulses. In this mode, time-resolved measurements are analogous to measurements made using pulsed sources such as electron discharge[1] or pulsed-laser[2] x-ray sources in that it is necessary to synchronize the triggering of the phenomena under investigation with the arrival of the probing x-ray pulses. The obvious advantage of synchrotrons sources over other pulsed sources is that the pulses are provided for the experimenter, eliminating the need for development or operation of a source. In some cases, laboratory pulsed x-ray sources are more intense than synchrotron pulses, but the collimation, tunability, and rapid repetition

associated with synchrotron pulses are in many cases essential to measurements. As indicated in Fig. 1, the repetition rate of synchrotron x-ray pulses is typically a few hundred nanoseconds so measurements can be performed in rapid succession if the phenomena under investigation can be stimulated on that time scale. The short interval between pulses can also be a source of difficulty, though, as x-ray detector systems may not be able to process photons scattered from successive synchrotron pulses. This aspect will be discussed later.

The pulse widths of most existing and planned synchrotrons vary from ~100-300 picoseconds; however, for discussions in this paper, the precise numbers are not critical and we will use 150 picoseconds as a representative number throughout the paper. The pulse repetition rates also vary considerably from one synchrotron source to another. Most notable is NSLS, which has a 19 nanosecond spacing in its ordinary mode of operation. Let it be understood that discussions relative to the pulsed nature of synchrotron sources would relate to NSLS in the single bunch mode (568 nanoseconds bunch spacing) rather than its normal multi-bunch mode.

Time resolution on the picosecond scale is indicated as a possibility in the region labeled quasi-continuous in Fig. 1. To our knowledge, such measurements have not yet been performed using hard x-rays, and the timing jitter associated with synchrotron pulses needs to be considered for the facility to be used. However, diffraction patterns have been obtained from a single synchrotron pulse using an undulator (to be discussed in Section 5 below), and streak camera techniques in principle provide the potential for time resolution down to a few picoseconds during the ~150 picosecond synchrotron x-ray pulses[3]. In such measurements, the synchrotron pulse would be thought of as a quasi-continuous x-ray source in which a continuous (but not constant) flux of x-rays is available on a periodic basis.

Some examples of physical science phenomena of interest for study in the various time domains are listed in Fig. 1. The topics indicated range from (a) investigations of the very sluggish thermodynamic or strain driven motion of dislocations or crystal grain-boundary motion; (b) to phase changes associated with atomic diffusion (i.e. ordering); (c) to the generally more rapid displacive structural transformations; and (d) to consideration of electron-phonon interactions such as the transfer of electronic energy to heat energy in the form of phonons on the picosecond time scale. Similar areas of interest can be compiled for other areas of research as well.

4. MEASUREMENT TECHNIQUES

We discuss here experimental configurations appropriate for investigations in the various time scales. Emphasis will be on the class of experiments that can be investigated and general experimental requirements rather than on particular experiments or detailed discussion of equipment capabilities. Measurements will be grouped into time scales of millisecond-to-kilosecond (continuous beam), nanosecond-to-microsecond (pulsed beam), and picosecond (quasi-continuous beam).

4.1. Millisecond-to-kilosecond range

Fig. 2 depicts experimental x-ray scattering configurations applicable to time-resolved measurements from milliseconds to kiloseconds. As noted above, these time scales are long compared to the separation between x-ray pulses and time-resolved experiments need only be concerned with the time scale of the phenomena to be studied. To the extent that sufficient scattering intensity is available, 2-D TV based vidicon or charge coupled device (CCD) detector systems can be used to make direct measurements with time frames of a few milliseconds. For cases where analog data is sufficient, such as in x-ray topographic imaging of dislocation motion or grain boundary migration, the data can be recorded directly on video tape. Direct use of video tape without image processing or averaging is normally possible only in cases where the imaging is from Bragg reflections or small angle scattering so that high intensities are available. CCD detector technology has been developing rapidly[4], as indicated by other papers in these proceedings, and new capabilities in this area will further stimulate the use of such devices. The use of micro channel plate (MCP) intensifiers with CCD detectors provides the possibility of electronicly gating the detector for data readout or for control of data collection integration times. As a result, 2-D time-resolved measurements in the millisecond and longer time scale can be carried out conveniently using time slice measurements and performing data readout with the input signal

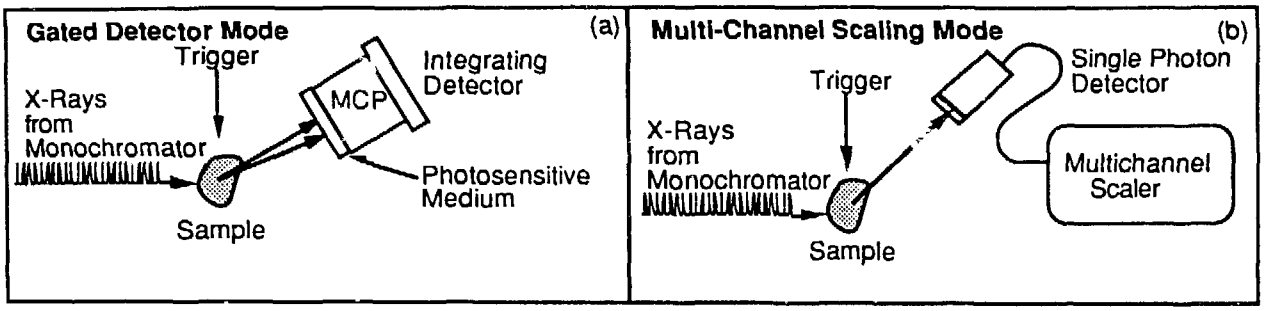


Fig. 2. Scattering configuration (a) for time-resolved measurements in the millisecond to kilosecond range using an integrating detector (such as CCD) and an optional MCP intensifier; (b) for time-resolved scattering measurements made using a single element detector and a multichannel scalar to bin measurements as a function of time.

electronically suppressed. To the extent that MCP intensifiers can be gated faster, even shorter time scales can be addressed using CCD detectors, however, the speed of phosphors must be commensurate with the gating as well. Gruner[5] has reviewed time-resolved detection issues in some detail in connection with biological structure measurements.

In cases where 2-D data are not required, but where precise angular resolution is needed, time-resolved work in the millisecond and longer time scales has been carried out with linear (i.e. 1-D) CCD detector arrays. Measurement cycles of ~3 milliseconds are possible by grouping a 1024 element array into 64 segments for readout[6]. Further, through the use of special readout electronics, time resolution down to 50 microseconds has been achieved for 1-D measurements through the use of 2-D CCD arrays in a mode similar to that of a streak camera. In this mode, a single row of the detector is exposed to photons and data from this single row is shifted in parallel to the unexposed rows of the device for readout after a sequence of single row measurements has been completed[4]. Time-resolved small angle scattering measurements on phase separation and lattice ordering in block copolymers have been reported recently using a linear CCD array using 1 second time slices[7]. Scattering from polycrystalline Cu_3Au during ordering phase transformations measurements were made[8] with time slices of 60, 100, or 500 milliseconds, depending on the resolution needed. Clarke discusses time resolution using the shifting row technique in his paper in these proceedings[9].

Fig. 2(b) emphasizes that single element detectors can be used for time-resolved measurements in this range as well. When it is not necessary to collect data in parallel, arbitrary time slices can be obtained using single detectors by electronically gating scalars under computer control or by using multi-channel analyzers in the multi-channel scaling mode. The obvious drawback of a single element (i. e. 0-D) detector is that measurements at various angles cannot be performed simultaneously so that time-resolved applications require either measurement scans on times short compared to the time scale of the phenomena, or continuous recycling of the phenomena under investigation. Although one- or two-dimensional data collection may be indispensable in cases where recycling of phenomena is difficult or impossible, in many cases the low noise, energy resolution, and inherent simplicity renders single element detectors the tool of choice. Nanosecond resolution measurements discussed in Section 5 below represent such a case.

For small angle scattering measurements and for measurements made on polycrystalline and amorphous materials, linear and area detectors collect data at the desired reciprocal space positions, so data collection is enormously aided by the use of 1- and 2-D detectors. However, scattering along symmetry directions from single crystals is a case in which single element detector measurements are not necessarily a large compromise. Since linear and area detectors perform measurements along the Ewald sphere, only one element on a linear or area detector array falls on a line of symmetry in reciprocal space. Therefore, angle scans are required for 1-D and 2-D detectors as well as single element detectors. Of course, if time-resolved measurements over planes or volumes in reciprocal space are needed, multi-element arrays are nearly essential. It should also be recognized that in many cases linear and area detectors provide measurements close enough to symmetry directions to be

useful or the additional data in the close proximity of the desired position can be used to improve the precision of the desired data by smoothing or extrapolation.

4.2. Nanosecond-to-microsecond range

The time-resolved techniques discussed in the previous section require either detector gating or data readout with speeds on the time scale of the desired time resolution. The experimental arrangements in Fig. 3 make explicit use of the pulsed nature of synchrotron x-rays to relax the requirement of gating detectors at the speed of the measurement resolution. As shown in the figure, the 150 picosecond synchrotron x-ray pulses arrive at intervals of ~200 nanoseconds so scattering occurs only at discrete times; therefore, the measurement resolution is determined by the width of the synchrotron pulse, not by the gating of the detector. That is, to obtain time resolution equal to the synchrotron pulse width, it is only necessary to synchronize the phenomena under investigation with the arrival time of an x-ray pulse. However, the number and frequency of measurements made during any one cycle of the phenomena are limited by the 200 nanosecond intervals between x-ray pulses. Since most measurements require more than one x-ray pulse to provide adequate statistical precision, the actual timing resolution will also depend on the reproducibility of the triggering.

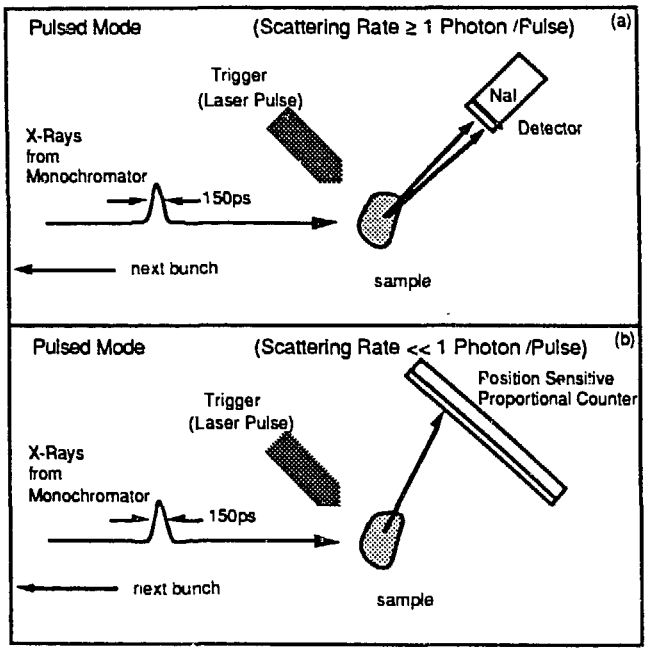


Fig. 3. Scattering configuration making use of the pulsed nature of synchrotron radiation to perform measurements on the nanosecond to microsecond scale. (a) Synchronization of the sample stimulus (laser) with the arrival of a probing x-ray pulse and determination of the number of scattered x-rays from a single bunch through measurement of the total pulse amplitude. (b) Configuration as in (a) with a position sensitive proportional detector operated in a mode so that no more than one photon is scattered from a single x-ray bunch.

Pulsed mode measurements can be subdivided into strongly and weakly scattering categories. For strongly scattering phenomena (more than one x-ray scattered from a single x-ray pulse), it is necessary to provide either an integrating position sensitive detector (such as a CCD device) or make use of a single photon detector that analyzes the total pulse height to determine the number of scattered photons per x-ray pulse. For ~150 picosecond x-ray pulses, all photons arriving at a detector from a single bunch will be detected as one pulse equal to the sum of the individual amplitudes. Since it is normally not possible to block the x-ray pulses before and after the desired x-ray pulse, it is necessary for the detector to be able to process and store (or exclude) x-rays from the preceding and following pulses in order to separate the x-rays scattered from the desired x-ray bunch. Therefore, pulsed mode measurements require a reasonably fast detector system or a gate (capable of operating faster than the synchrotron pulse frequency) for the photomultiplier or image intensifier. It is convenient that ordinary NaI scintillation detectors can be made to operate at these speeds and provide pulse height information to determine the number of photons detected from a single burst. We are not aware that CCD devices have been used in this fast mode, but it appears that MCP gating at these speeds should be

possible if appropriate phosphors exist. As discussed in Section 5 below, ultrafast rotating mechanical slits to block preceding and following x-ray pulses can be devised in some cases.

For the weakly scattering case (much less than one photon scattered per synchrotron pulse) and investigations of phenomena that can be triggered repeatedly without damage to the sample, it is possible to make use of gas filled position sensitive proportional counters and a gated data collection system to make nanosecond resolution measurements. In this configuration (depicted in Fig. 3(b)), only x-rays scattered in coincidence with the triggering of the phenomena are processed and again the precise timing is taken care of by the timing of the synchrotron pulse, not by the detector. The scattering rate must be low enough that two photons are not scattered from a single pulse, because they will be coincident in the detector and disrupt the position sensing ability. New detectors of this type are being developed that will increase the speed of such non-integrating proportional counters.

Examples of experiments using pulsed mode techniques can be found in nanosecond resolution investigations of pulsed melting in silicon by Larson[10,11] *et al* in the strong scattering regime, and by Bartunik[12] *et al* in the weak scattering regime. The pulsed laser melting measurements of Larson *et al* were carried out using a NaI scintillation detector and a step scanning method to compile nanosecond resolution scattering profiles as a function of time relative to the laser pulse. Bartunik *et al* made use of a gas filled position sensitive proportional counter to monitor a Bragg scattering profile in a nanosecond resolution study of an 18.6 nanosecond excited state of Ce^{3+} in laser excited CdP_5O_{14} single crystals

4.3. Picosecond range

The picosecond time scale represents a new frontier for x-ray diffraction measurements. While picosecond measurements have been carried out in connection with controlled fusion measurements[13], scattering experiments have not been done using hard x-rays from synchrotrons to our knowledge. Fig. 4 illustrates the requirement of dealing with an incident beam that is varying in intensity over the ~150 picosecond duration of the synchrotron pulses. This mode of operation is continuous (or quasi-continuous) in the sense that the x-ray pulses no longer act as strobes, but rather provide continuous illumination of the sample for the 150 picosecond duration of the pulse. This complicates time resolution measurements because detectors with picosecond resolution are needed, unlike the nanosecond resolution pulsed mode measurements described above in which the detector only records the scattering event and the precise timing is obtained from the arrival time of the x-ray pulse. This quasi-continuous mode of measurement may not be fully realizable until the advent of next generation synchrotrons such as the Advanced Photon Source (APS) and European Synchrotron Radiation Facility (ESRF) with undulator insertion devices and higher photon fluences.

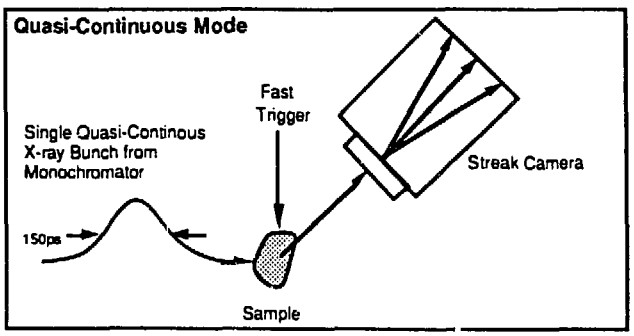


Fig. 4. Scattering configuration for time resolved scattering with a single synchrotron pulse. The sample is stimulated by the trigger and the streak camera is synchronized with the arrival of the x-ray pulse; the variation in the amplitude of the incident x-ray pulse must be considered in the analysis.

5. APPLICATIONS

Fig. 5 shows the results of time-resolved x-ray scattering measurements[14] made during the temperature-induced amorphous to crystalline transformation in $NiZr_2$ at NSLS. These measurements correspond to 200 ms time slices made using a linear (non-intensified) CCD detector in a geometry similar to that illustrated in

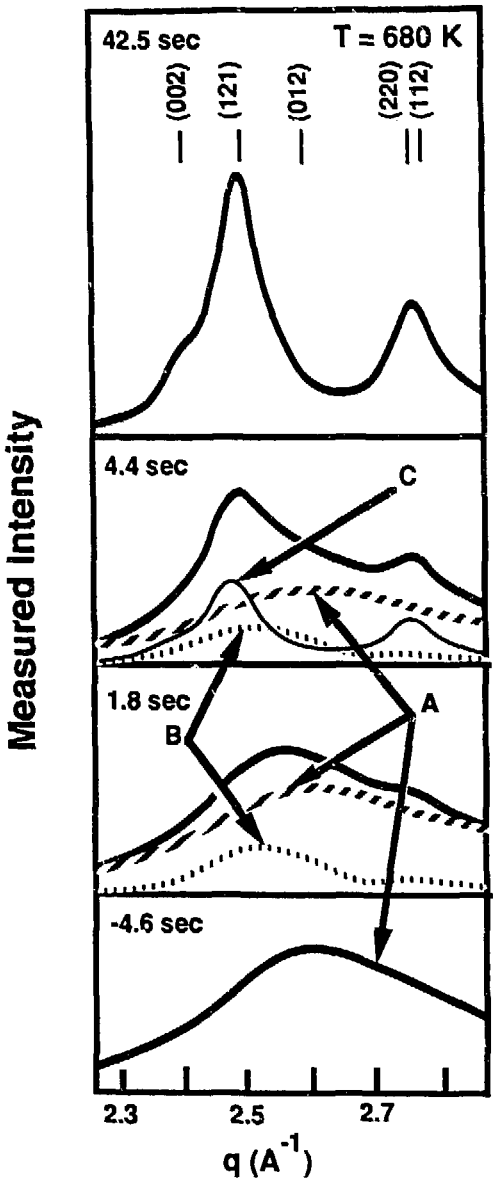
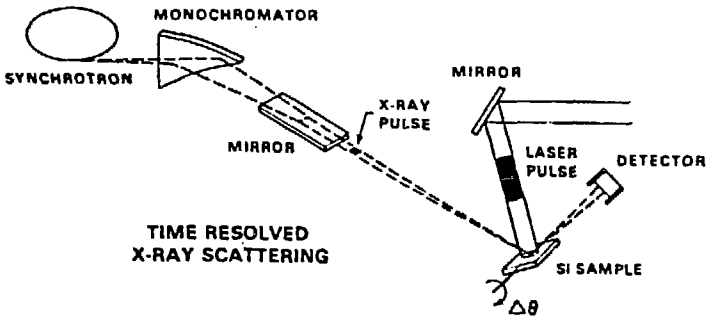


Fig. 5. X-ray scattering measurements on amorphous $NiZr_2$ made[14] during the thermal transformation to crystallinity. The -4.6 sec panel represents scattering before the transformation starts, curves A correspond to the amorphous scattering at the respective times, curves B correspond to precursor scattering at their respective times, and curve C corresponds to the crystalline scattering at 4.4 sec. The thick line represents the total observed scattering at each time and curves A, B, and C were infered from the analysis.

Fig. 2(a). The data displayed correspond to times where 0, 25, 50, and 100% of the volume was crystallized. The curves shown here are hand-drawn lines through original data that was composed of 64 separate data points per curve. Although only 200 millisecond time slices were needed for this study, the detector used was capable of 3 milliseconds resolution. These data show the initial amorphous scattering band (measured 4.6 seconds before the sample reached 680 K) and the emergence of Bragg scattering peaks as the sample transformed from an amorphous to a polycrystalline material. After careful analysis of these and other scattering profiles and by comparing these data with measurements at lower annealing temperatures, the authors concluded that the measurements at 1.8 and 4.4 seconds were not a linear combination of the crystalline phase plus the amorphous phase, but that a crystalline precursor phase with a diffraction pattern given by the dotted lines B in Fig. 5 formed in the sample before crystallization into the final structure. Kinetic details of the crystallization process at this level have not been addressable previously.

An example of nanosecond resolution time-resolved x-ray diffraction measurements is illustrated in Fig. 6, which depicts work performed at the Cornell High Energy Synchrotron Source (CHESS) by Larson[10] *et al.* Fig. 6(a) shows the scattering geometry and illustrates the synchronization of a laser pulse with the arrival time of a synchrotron x-ray pulse. In the experiment, 25 nanosecond laser pulses rapidly melt the near surface region of a silicon single crystal and thermal expansion induced strain is measured using the 150 picosecond x-ray pulses. Fig. 6(b) shows time-resolved Bragg reflection profiles measured during the melting process (15 ns after the beginning of the laser pulse) and during the crystal regrowth process (45 ns). These measurements were made along the [111] reciprocal lattice direction at the (111) reflection in a step scanning mode with each data point the average of 27 shots. The solid lines represent fits to the scattering curves to determine the thermal strain distribution. Since the scattering at angles away from the Bragg peak arise from thermal expansion induced strain in the near surface region, analysis of the scattering profiles provides a direct measure of the near surface temperature distribution. Measurements similar to these were used resolve a controversy regarding the mechanism of pulsed laser annealing in semiconductors, and an analysis of the measurements shown here were used to probe overheating and undercooling at the liquid-solid interface during melting and regrowth in silicon. These data indicate an unexpected asymmetry in overheating and undercooling in which silicon appears to melt with less than 2 K/m/s thermodynamic driving force, while regrowth was found to require undercooling of 6K/m/s on <100> and 11 K/m/s on <111> oriented crystals.



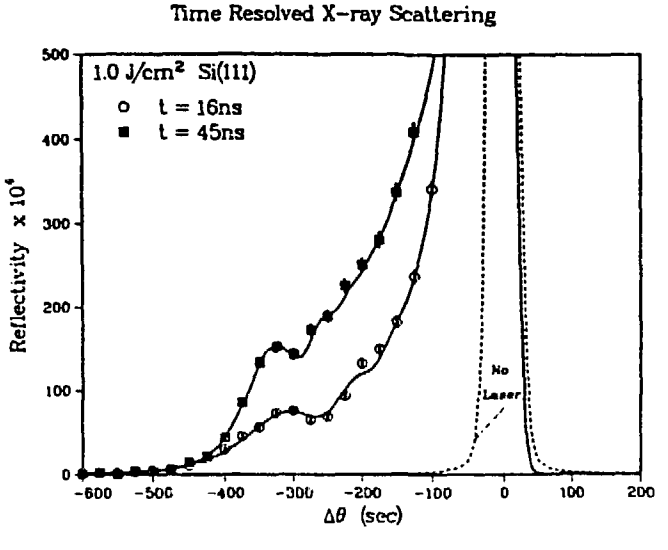


Fig. 6. (a) Schematic representation of the synchronization of a 25 nanosecond laser pulse with the arrival time of a probing synchrotron x-ray pulse for the study of silicon during laser melting and regrowth. (b) Time resolved x-ray scattering measurements made on silicon during the laser pulse (16 ns) and after the laser pulse (45 ns) during regrowth. The solid lines represent least squares fits to the data, from which the depth dependence of thermal strain and the lattice temperature distributions were determined.

Fig. 7 illustrates an experiment in which a crystal diffraction pattern from a lysozyme crystal was collected from a single (white) x-ray pulse during an undulator run at CHESS. As shown in the figure, two rapidly rotating slits[15] were used to limit the exposure to a single x-ray pulse, and the diffraction pattern was collected on a photostimulable, integrating image phosphor. The slit system used provided a window of 2 microseconds, which was less than the 2.56 microsecond bunch spacing of CHESS in the single bunch mode. It was found that the diffraction pattern from this single pulse could be successfully analyzed, and although this experiment did not include a time dependent stimulus to the crystal, it does demonstrate the potential for single pulse crystallography using undulator insertion devices. An extrapolation of this type of experiment to include excitation of a crystal with picosecond or shorter laser pulses during the x-ray pulse could, in principle, provide information on very rapid crystal dynamics or transformations. Since CHESS normally runs in a 7 bunch mode at present, and most synchrotrons have bunch spacings of only a few hundred nanoseconds, this shutter technique would have to be extended if it were to be used routinely.

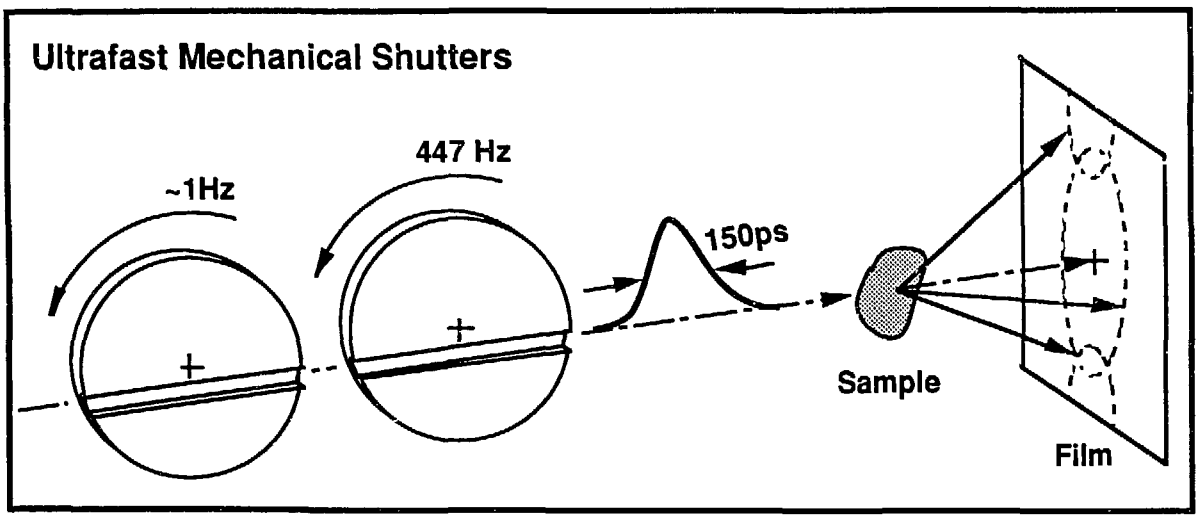


Fig. 7. Schematic view of the use of ultrafast mechanical shutters to select a single x-ray pulse from CHESS for the collection of a single synchrotron bunch diffraction pattern from a lysozyme crystal. The film material was a photostimulable storage phosphor.

The final application to be discussed illustrates a use of the pulsed nature of synchrotron sources in which the time resolution is used for spectroscopy rather than studying the kinetics of phenomena. Fig. 8 is an idealized representation of the time dependence of resonant nuclear (Mössbauer) scattering of synchrotron pulses from an isotopically enriched monochromator. Since resonant nuclear excitation of ^{57}Fe is limited in energy width to ~10^{-8} eV, the uncertainty principle requires photon emission in the resonant nuclear process to occur on a time scale of several tens of nanoseconds rather than on the 150 picosecond time scale of the incident pulses. The time delay separates the resonantly scattered beam (that has ~10^{-8} eV energy resolution) from any electronically (prompt) scattered photons that would have resolution of typically ~1 eV. More importantly, this resonantly scattered beam can be used to spectroscopically probe dynamic phenomena with energy resolution on the 10^{-8} eV scale. That is, analysis of the time structure of such a beam after scattering from a sample containing low energy excitations provides information on the energy scale of the fluctuations. In addition, nuclear resonant analyzers can be devised to separate elastic and inelastic scattering on the 10^{-8} eV scale as well. These concepts are more complicated in practice than has been depicted here as a result of Zeeman splitting of the energy levels and considerable development remains; however, the general principles have been demonstrated[16,17]. Due to the limited number of photons available in the narrow energy band, general application of such techniques will require the intense undulator beams that will be available from next generation synchrotrons, however, development work can be carried out on present machines.

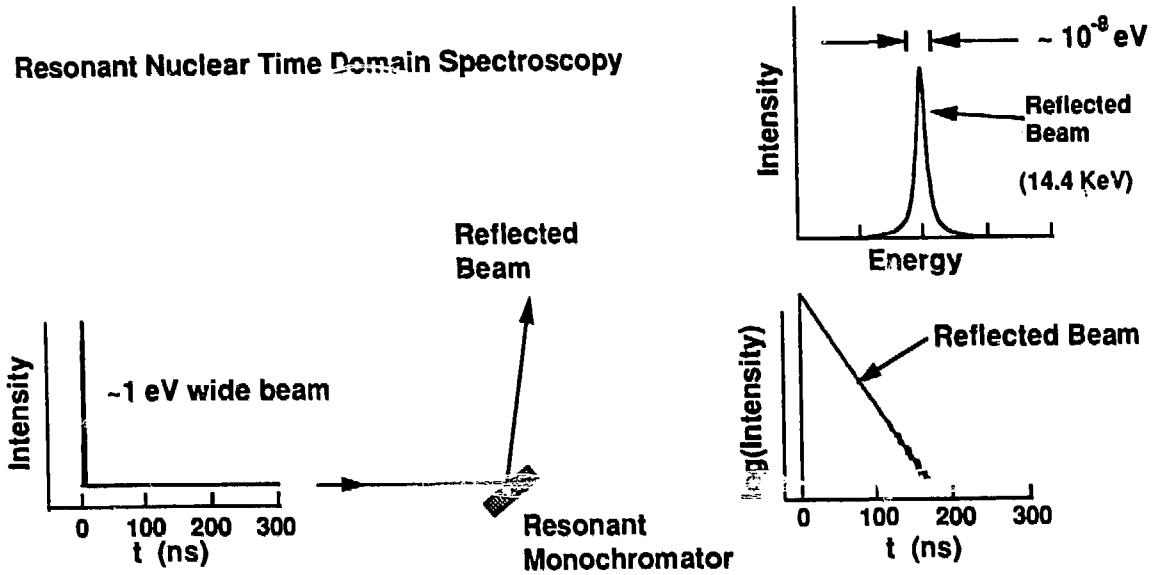


Fig. 8. Schematic view of the time structure and energy width associated with resonant nuclear monochromatization of 150 picosecond synchrotron x-ray pulses. The 150 picosecond incoming pulses are presumed to have ~1 eV widths from a premonochromator; after scattering resonantly from the resonant monochromator, the energy width is reduced to ~10^{-8} eV and the time structure is broadened due to the lifetime of the resonant state.

6. CONCLUSIONS

The pulsed time structure as well as the high photon fluxes available from synchrotron x-ray sources provide new opportunities for time-resolved x-ray scattering investigations, and next generation synchrotron facilities now under construction will even further increase these possibilities. The development of new and improved experimental techniques and the continued development of photon detectors with increased capabilities will have a significant influence on progress in time-resolved studies as well. In particular, the development of detectors capable of selecting single synchrotron pulses or the development of shutter mechanisms to select single x-ray pulses will be important for full exploitation of the pulsed time structure of synchrotrons.

Time-resolved measurements have been carried out in disciplines varying from biology to fundamental physics, and results have led to new and unanticipated information on physical systems as well as new insight into the kinetics of physical processes. As time-resolved techniques and measurement equipment mature, increasingly sophisticated experiments can be expected and non-equilibrium investigations will be possible on a wider range of systems.

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