

# Advances in Experimental Techniques in Synchrotron Radiation X-ray Research

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As capabilities of synchrotron x-ray sources continue to advance, so too do new experimental techniques that better utilize the source characteristics. Also, existing experimental techniques have been improved in their performances and extended in their applications. The photon flux and the brilliance (photon flux per area per solid angle per photon energy) from synchrotron radiation sources did not improve in a small integer factors, but leapfrogged in the order of magnitude from one generation to the next. Also, beam stability, the range of available photon energies and the time structure of the beam, purity in polarization improved remarkably in recent years. These advances in source properties stimulated the development of many new experimental techniques. Among these new or improved techniques, several examples will be covered to show how various aspects of high brilliance sources can be made useful. These techniques introduced here are relevant to the area of condensed matter physics and materials science.

## I. Source Characteristics and Optics

Before the experimental techniques are reviewed, attention should be drawn to the improvement realized in the third-

generation synchrotron x-ray sources. The characteristics of the type A undulator of the Advanced Photon Source (APS) were measured recently. The undulator A can provide very strong x-rays in the range of 2.9 KeV to 45.0 KeV using its first, third and fifth harmonics. The measured source sizes and divergences were  $359 \mu\text{m}$  (h)  $\times$   $21 \mu\text{m}$  (v) and  $24 \mu\text{rad}$  (h)  $\times$   $6.9 \mu\text{rad}$  (v) in  $\sigma$ . A typical undulator beamline with a Si(111) double crystal monochromator at APS can deliver about  $3 \times 10^{13}$  photons/sec at 8 KeV with 2 to 3 eV (full width at half maximum) energy spread.

A Si(111) crystal is still the best choice as a monochromator. The Darwin width (equivalent to angular acceptance) of Si(111) matches very closely with the vertical divergence of the APS undulators. Therefore there is no loss in brilliance through the Si(111) monochromators. Only difficulty with Si(111) was how to prevent thermal distortion under enormous thermal load (the total power, a few kW maximum and the power density,  $167 \text{ kW/mrad}^2$  or  $185 \text{ W/mm}^2$  at 30 m away from the source). This was solved by cooling the crystal to the liquid nitrogen temperature. At the liquid nitrogen temperature, the thermal expansion of Si almost disappears. Although one can use a thin diamond crystal as a monochromator to deal with the thermal load, the intensity will drop by the factor of 2.5.

In high energy third generation synchrotron sources, such as ESRF, APS and Spring-8, the harmonic contamination (A Si(111) double crystal monochromator can tune to  $\lambda$ ,  $3\lambda$ ,  $5\lambda$ , ... simultaneously.) can be removed to a practical level only by a pair of mirrors. Even with progresses in fabrication and measurement methods, it is very difficult to acquire a mirror with a figure error below a few  $\mu\text{rad}$ . This figure error will add the vertical beam size in the usual vertically reflecting geometry. When the mirror is located 10 m away from the sample position, this means additional tens of  $\mu\text{m}$  broadening (in half width). With the present technique, the only way to preserve the phenomenal brilliance is to locate reflecting optics close to the sample position. The conserved brilliance through the modern x-ray optics benefited most of the examples presented next.

## II. Thermal Diffuse Scattering (TDS) Imaging

Although phonon dispersion curves have been measured mainly by neutron scattering, x-ray thermal diffuse scattering was also used to determine dispersion curves in early days. However, thermal diffuse scattering as a tool of phonon measurements has been almost forgotten. The conventional measurement of x-ray thermal diffuse scattering is a time consuming and tedious process. Also, data reduction is not simple. However, with

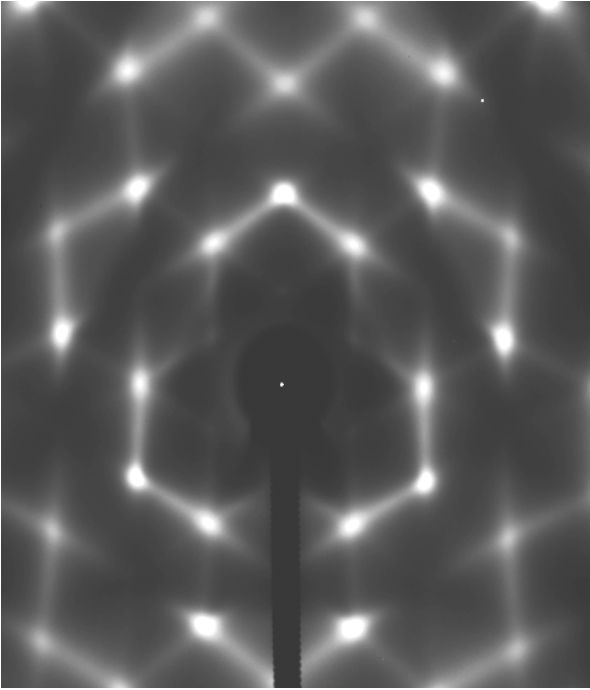


FIG. 1 Transmission x-ray thermal diffuse scattering images taken from a Si(111)

enormous number of photon flux available and the advances in 2-D detectors, such as, low-noise high-dynamic-range CCD detectors and x-ray imaging plates, data acquisition time was shortened from days (with a second generation synchrotron source) to less than a minute. Fig. 1 shows a TDS patterns [1] from a Si wafer recorded on an imaging plate. X-rays of 28 KeV were used. Fig. 2 also shows the phonon dispersion curves obtained through a least-squares analysis in terms of lattice dynamics.

The fast data acquisition rate, simplicity of the experiment, and a minimal requirement on sample volume make this method attractive to a wide range of applications in condensed matter physics. This will be an alternative to neutron scattering or inelastic x-ray scattering to measure lattice vibrational characteristics. TDS patterns from various materials such as metal, oxides, high  $T_c$  superconductors have been already recorded. It seems that there is no limit on atomic species in contrast to the neutron technique where the scattering cross-section varies very widely atom to atom. The data collection time can be reduced as small as 10 msec with an optimized setup. This rate is fast enough to allow dynamic studies such as ones in phase transition. In conjunction with microbeam techniques, phonon studies from single grains in practical material should be possible.

### III. Microbeam Diffraction

With the shrunken source size of high-brilliance synchrotron sources, diffraction with micron or sub-micron size beams can be performing more readily. Efforts to

produce efficient microfocusing optics are pursued by many different ways. Two popular methods are Fresnel zone plates and Kirkpatrick-Baez mirrors. Both methods have reported to achieve sub-micron focusing. In Kirkpatrick-Baez optics, one uses two cylindrical mirrors (or elliptical mirrors to eliminate spherical aberrations) to focus beams in both vertical and horizontal direction. Because there is no way to focus the full undulator beam to a sub-micron size, the incoming beam should be reduced to a size of less than 100  $\mu\text{m}$  before focusing. Zone-plate method is also not so simple. Because zone plates methods are diffraction originated, it produce multiple focused spots and it requires a carefully aligned order-sorting aperture.

One of the difficulties with microbeam diffraction is that there is no diffractometer with sub-micron sphere-of-confusion. In other words, depending on the angle of the diffractometer, the micro-beam will illuminate different volumes of the sample. To overcome this problem, one can fix the sample and find four reflections from white-beam Laue diffraction to obtain the grain orientation. Then, the x-ray energy of one reflection can be measured to determine the lattice volume. An ingenious way to alternate white-beam and monochromatic beam to the same sample diffraction volume with very small positional error has been developed. [2]

### IV. High Energy Resolution Experiment

There are a few different ways to achieve milli-eV energy resolution in x-ray. The principle is to utilize high order reflections (such as; Si(11 11 11) )of crystals. There is geometrical constrain for high order reflections and loss by reduction of angular acceptance. By using a back-scattering geometry, one can achieve highest order reflection for given energies.[3] The improvement in throughput can be realized by a nested channel-cut monochromator. This monochromator is a combination of a lower index asymmetric channel-cut crystal and a higher index channel-cut crystal nested by the other. The

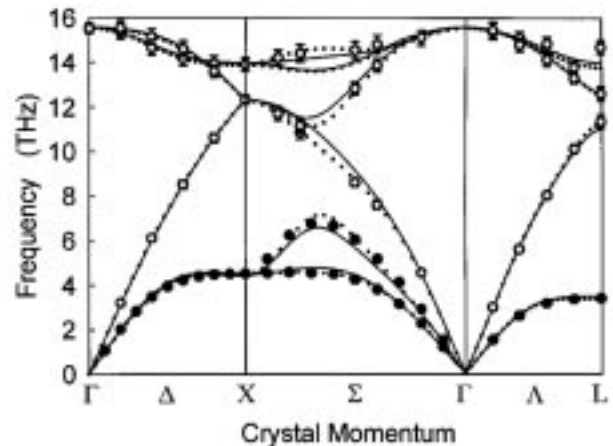


FIG. 2 Phonon dispersion curves of Si. The both of open circles and closed circles are neutron scattering data. Solid Curves are derived from the best fit to the x-ray TDS patterns. Dotted curves are an interpolation of neutron data.

first diffracting asymmetric crystal is accepting wide divergence and squeezes the divergence to match the angular acceptance of the higher indexed crystal. [4] The resultant angular acceptance can be increased close to the vertical source divergence of a typical undulator (about 10  $\mu$ rad). There are also variants of these two methods. A photon flux of  $9 \times 10^8$  ph/sec in 3.6 meV energy resolution has been reported. [5]

### V. Ultra Small Angle X-ray Scattering (USAXS)

The small angle scattering leads to the determination of the important microstructural parameters such as radius gyration, volume, shape, and total surface area. Particularly Bonse-Hart's method offers extended Q-space coverage and better resolution. Bonse-Hart USAXS instruments use two pairs of crystals. The first crystal pair works as a collimator and the second pair works as an analyzer. With the smaller source divergence of the high-brilliance sources, USAXS can produce 1000 times the throughput of earlier instruments. The available range became  $0.00015 \text{ \AA}^{-1}$  to  $0.5 \text{ \AA}^{-1}$ . A PIN diode with a linear operation over a 10-decade range allows uninterrupted measurements from the direct beam to the tail of small angle scattering curves without using filters. [6]

### VI. Time resolved surface x-ray diffraction

One of the most benefited area from high brilliance is surface x-ray scattering. Improvement resulted from the sheer increase of photon flux and the smaller emittance of the sources. In a grazing angle geometry, where the incoming x-rays are almost skimming the sample surface, the effective sample size becomes about tens of microns. Signal-to-noise ratio improves as one can focus the beam smaller without increasing angular divergence too much. The improvement is so much that now one can measure RHEED oscillation with a time resolution of 5 msec and

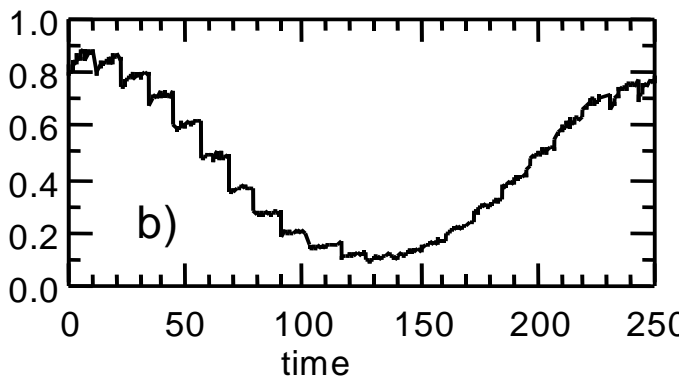


FIG. 3 Time-resolved diffraction from SrTiO<sub>3</sub> at (0 0 1/2) anti-Bragg position. Grown in 5 mTorr of O<sub>2</sub> at 780° C. An expanded view of first layer deposition is shown.

spot profiles with 3.5 sec. A milli-second time resolution allowed studies of kinetics of surface aggregation after each single laser pulse deposition. [7] Their results in Fig. 3 clearly show the abrupt drop in specula reflectivity at each laser deposited material lands on the substrate. Also, the angular distribution of the diffraction intensities from the growing film was collected in real time. [8] This was done using a CCD without interrupting deposition. A profound alteration of the detailed balances between the competing surface processes could be avoided. Amazingly, many so-called time-resolved structural studies during film growth were performed with repeated interruption in growth.

### VII. Reflection Surface X-ray Diffraction (RSXD)

RSXD is a method to observe reflection high energy electron diffraction (RHEED)-like surface diffraction patterns with x-rays. RHEED techniques have long been used to probe surface structures. Since the late 1970's surface diffraction has been the preferred methods for reliable structural determination of surfaces and interfaces. This is because the x-ray scattering can be treated with a simple kinematic calculation due to the absence of strong dynamical effects in RHEED. However, x-ray diffraction from surfaces is very weak and the intensity is often measured by point detectors. The point detection methods requires many scans on various possible diffraction spots to reveal the surface structures. RHEED or other electron diffraction techniques were developed to see the whole two dimensional diffraction pattern at once. RSXD utilize the full high-brilliance at higher energy (20KeV and beyond) and a highly sensitive CCD detector. Fig. 4 shows a typical RSXD pattern obtained with 20 KeV x-rays. [9]

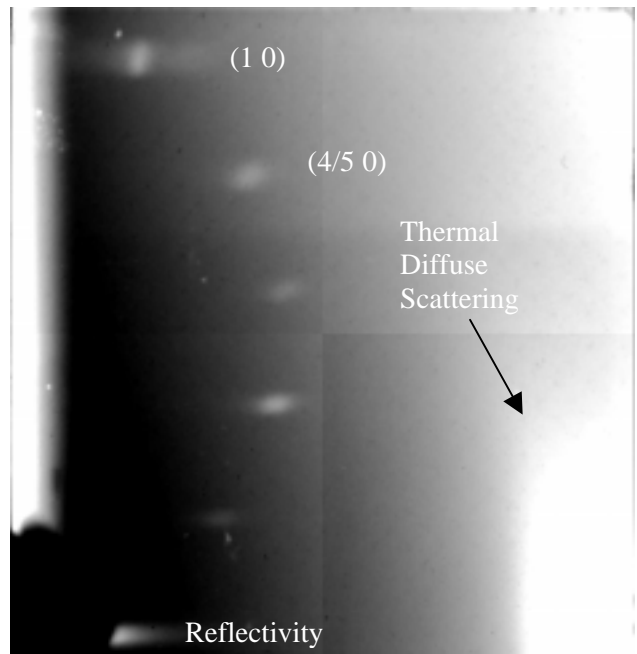


FIG. 4 A typical RSXD pattern of a Ge/Si(111)-(5x5)

## VIII. Conclusion

Beyond the examples presented here, there are many extremely useful techniques utilizing high brilliance synchrotron sources and more will emerge. All of them are expected to produce profound progresses in various areas in science, such as, condensed matter physics, material science, medicine and biology.

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