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A Three-Crystal Spectrometer for 150 KeV Synchrotron Radiation*

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ABSTRACT

A triple axis spectrometer for high energy (≥ 150 keV) synchrotron radiation has been constructed and characterized. The reciprocal space resolution function of this instrument has been measured using perfect silicon crystals as monochromator, sample, and analyzer. It was found to have dimensions of $1.0 \times 10^{-5} \text{ \AA}^{-1}$ and $2.3 \times 10^{-4} \text{ \AA}^{-1}$ in the directions perpendicular and parallel to the momentum transfer $\vec{Q}$ respectively, comparable to those achievable using standard 8 keV triple axis instruments. The large penetration depths of high energy x-ray photons, combined with the high instrumental resolution, opens new possibilities in the study of truly bulk condensed matter systems.

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MASTER

INTRODUCTION

X-ray scattering has been one of the primary tools for the determination of the structure of matter ever since the first observation of a diffraction pattern in the early part of this century. In the 1950's inelastic neutron scattering was first used to study thermal excitations in solids (phonons) directly. Since then the realm of structural analysis has been dominated by x-rays, while inelastic scattering from excitations in the meV regime and below has been the mainstay of neutron scattering. With the widespread availability of synchrotron radiation sources in the last decade, the x-ray scattering techniques have provided reciprocal space resolutions corresponding to coherence lengths of microns, as well as insights into uncharted frontiers. These techniques have utilized, for the most part, photon wavelengths of about 1.5 Å (8 KeV energy), where the absorption and extinction lengths for strong scattering are not dissimilar, and are typically a few tens of microns for most solids. Neutrons, on the other hand, suffer almost no absorption and thus allow the study of bulk materials. With this in mind we have set out to study the feasibility of using large penetration depths, typically associated with neutrons, combined with the high resolution possible with x-rays from synchrotron sources, by using high energy photons (≥ 150 keV).

This study is an extension of the gamma ray diffraction techniques that are lab-based using radioactive sources.¹ The orders-of-magnitude-enhanced brightness of synchrotron sources allows the capability of achieving the resolution of conventional (1.5 Å) experiments with high intensities at 150 KeV and beyond, not possible with radioactive sources. To understand the choice of energy, figure 1 shows plots of the total absorption cross section as a function of photon energy for silicon and iron, along with the various contributions to the cross section, in particular the photoeffect, coherent and incoherent (Compton) scattering.² As can be seen, at energies of order one to

several hundred KeV, the dominant contribution comes from incoherent scattering, and this is almost independent of energy. Thus, the best choice of energy is probably the lowest in this region, since the synchrotron radiation spectrum falls exponentially and the Bragg angles fall linearly with increasing energy.

A second advantage of using high energies is that the extinction length, which is a linear function of energy for a given reflection, is now small compared to the absorption length. This translates into an increase in the ratio of weak scattering that can be described by the kinematic approximation to the strong Bragg scattering that is limited by extinction. It thus becomes possible to probe the wavevector dependence of incommensurate phases, magnetic scattering, and Huang scattering from samples that are also useful for neutron studies of inelastic scattering. Furthermore, as we will show below, using the same triple axis geometry as is used at 1.5 Å, the reciprocal space resolution achievable is the same at 0.08 Å (150 KeV).

INSTRUMENT DESIGN

All of the considerations in designing a three axis spectrometer for this application, $\lambda=0.08\text{\AA}$, are the same as for a 'conventional' instrument operating at 1.5 Å, but scaled by the ratio of the photon wavelengths. Thus the required angular range and angular resolution are reduced by a factor of approximately twenty. For example the Bragg angle for the Si(220) reflection is now about 1.3 degrees. This reduction in scale makes the use of high resolution tangent arms an ideal choice for the monochromator, sample, and analyzer axes. In the prototype instrument constructed for the initial studies, the tangent arm length of 864 mm. combined with a precision micrometer drive gave an angular step size of 0.04 arcsec. and an angular range of 3 deg.

A schematic of the instrument is shown in fig. 2, and its realization in fig. 3. As can be seen from the schematic, collimators were placed before and after each axis to filter unwanted scattered beams. These collimators were constructed from mild steel to make the dominant fluorescence background appear at 6.4 KeV from iron, rather than at 74 KeV (much closer to the energy of the photons in use) from lead. Also shown in the schematic are two pre-filters, one of steel that serves to remove the heat load on the monochromating crystal and one of lead to attenuate the incident beam to keep the count rate in the solid-state detector below about 10,000 per second. The use of the solid state detector for this as well as any scattering experiment proves to be invaluable, both for diagnostics and reducing the background levels. In this experiment, for example, the background rates were much less than 1 cps from a perfect Si(220) sample away from the Bragg condition.

For the prototype studies, the equipment was chosen to fit into the A3 experimental enclosure at CHESS and no provision was made for sample environments other than ambient. The very small Bragg angles, however, lend themselves to a simple arrangement for environmental chambers and the problem of vacuum compatible, x-ray transparent windows is easily solved. Aluminum, for example, would satisfy.

Referring back to the earlier comment about the pre-filter, it should be pointed out that these experiments were carried out at wavelengths far shorter than the critical wavelengths, even from high field wigglers on high energy machines (for this experiment, $\lambda_c = 0.4\text{\AA}$ from the CHESS six-pole, 1.6T wiggler, with CESR operating at 5.4 GeV). Therefore, the thermal problems that are of concern for present high-power wiggler sources, and even more so for the proposed new sources, can be solved easily at these wavelengths. Experiments such as the present one are thus immune to this serious difficulty in performing high resolution, high intensity experiments at storage ring sources.

RESULTS AND DISCUSSIONS

The aim of the preliminary studies was to evaluate the feasibility of synchrotron based x-ray scattering studies at very short wavelengths. The major unanswered question was one of resolution: could one achieve reciprocal space resolution at $0.08\text{\AA}$ comparable to that of the now standard $1.5\text{\AA}$ triple axis instruments. That is, longitudinal resolutions of $\sim 10^{-4}\text{\AA}^{-1}$ FWHM³ which correspond to coherence lengths of several microns in real space.

To perform those studies, measurements using float zone Si(220) Laue case monochromator and analyzer crystals were performed. The 'sample' was a third float zone Si(220) Laue wafer. These crystals were placed in the conventional (+, -, +) or focusing arrangement. Since the reflections were the same, wavelength dispersion need not be considered.

An additional simplification occurs at these short wavelengths. With $k_i = 73\text{\AA}^{-1}$, scans in reciprocal space along and perpendicular to the momentum transfer are well approximated by simple angular scans of analyzer and sample respectively. Typical transverse and longitudinal scans are shown in figure 4. The rocking curve widths correspond to $\Delta Q_{\perp} = 9.2 \times 10^{-6}\text{\AA}^{-1}$ and $\Delta Q_{\parallel} = 1.7 \times 10^{-4}\text{\AA}^{-1}$. Similar rocking curves to these were measured for different combinations of sample and analyzer crystal orientations to map the resolution function for this focusing setting. The resulting contour map is shown in figure 5.

The features of the resolution function can be understood qualitatively.⁴ The Laue case monochromator and analyzer have Lorentzian diffraction profiles with $1/q^2$ tails. These profiles produce streaks tilted at $\pm\theta$ with respect to the momentum transfer $\vec{Q}$. As seen in figure 5, two streaks extend a considerable range nearly parallel to $\vec{Q}$ split by an angle equal to 2θ . Also seen in the figure is a "bulge" transverse to $\vec{Q}$. This stems from the

Lorentzian profile of the perfect crystal sample, also in the Laue case. When the sample is away from the exact Bragg condition, the entrance and exit points on the entrance and exit media (vacuum) Ewald spheres differ by a reciprocal lattice vector which is always perpendicular to the crystal surface; this is due to the conservation of the tangential component of the wavevector across the surface.⁵ Thus in the pure Laue case, the $1/q^2$ tails (i.e., the deviations from the exact Bragg condition) produce streaks perpendicular to the reciprocal lattice vector $\vec{H}$,⁶ and therefore to the momentum transfer, as in figure 5. In the pure Bragg case, the $1/q^2$ tails would produce streaks parallel to $\vec{H}$.⁶

From the figure the FWHM for $\Delta Q_{//}$ and $\Delta Q_{\perp}$ are $2.3 \times 10^{-4} \text{ \AA}^{-1}$ and $1.0 \times 10^{-5} \text{ \AA}^{-1}$ respectively. These values correspond to longitudinal and transverse correlation lengths of 5.5 and 126 microns, defined through $\text{HWHM} = 2\pi/\xi$, where ξ is the correlation length.

Results at 7 KeV, at the focusing condition³, are similar. The slightly smaller values for the resolution function dimensions result from the choice of Si(220), compared with the 7 KeV measurements using Si(111).³ It is clear from these preliminary studies, however, that high q-resolution elastic scattering is not solely the domain of the 1 Å region of the spectrum. The degradation of the resolution as one moves off the focusing condition, $d_{\text{sample}} \neq d_{\text{mono}} = d_{\text{analyzer}}$, still requires investigation.

With use of deformed crystals for monochromator and analyzer, one can realize more intensity at the expense of resolution. Experiments using annealed Czochralski Si(220) wafers are described in another paper.⁷

SUMMARY

Storage rings operating at high energies (5-6 GeV) and strong magnetic fields (~ few T) provide copious fluxes of 150 KeV photons. Using a 'conventional' three axis scattering instrument, the penetration depth of these short wavelength photons can be combined with high q-resolution to open new possibilities in the study of truly bulk condensed matter systems. The short wavelengths also seem to simplify, rather than complicate, the experimental conditions because of the small angular ranges and x-ray transparency of easily fabricated windows for standard environmental chambers.

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DISCLAIMER

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FIGURE CAPTIONS

1. The various contributions to the absorption cross section are plotted vs. photon energy for silicon (top) and iron (bottom). At high energies, incoherent scattering is the dominant contribution. Taken from reference 2.
2. Schematic drawing of the three axis spectrometer. The three silicon wafers (thickness T_0) diffract in the Laue geometry. The indicated dimensions are in mm.
3. Side views of the spectrometer. The photograph at top, in which the incident beam enters from the left, shows the diffracting crystals and steel collimators. The photograph at bottom, in which the incident beam enters from the right, highlights the tangent arms and precision stepping motor/micrometer drives.
4. Transverse and longitudinal scans for three Laue case Si(220) diffracting crystals, of thickness $T_0 = 10$ mm.
5. Isointensity contours derived from transverse scans for a series of orientations of the analyzer crystal. The vertical direction is parallel to $\vec{Q}$. The streaks which run nearly parallel to $\vec{Q}$ are misaligned by 2θ . The fact that the contours are not symmetric with respect to the coordinates is merely an artifact of the data transformation used.

Figure 1

Figure 2

figure 3

Figure 4

(perf - perf - perf) resolution function

Figure 5.