The development of high energy X-ray total scattering method at Beijing Synchrotron Radiation Facility

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Abstract

As a powerful local structure probe, high energy X-ray total scattering method has been used widely in condensed matter physics, materials science and other fields. With a super-conducting wiggler (SCW) and sagittal focusing monochromator, a high energy X-ray total scattering apparatus has been developed at 3W1 beamline of Beijing Synchrotron Radiation Facility, BSRF, a first-generation synchrotron source. The total scattering apparatus mainly consists of a large two-dimensional flat-panel detector and high-energy X-rays of 50~70 keV, enabling total scattering measurements to be carried out for pair distribution function (PDF) analysis with a Q range of 0.5~25 Å−1. With this setup, a series of in-situ devices were developed, data with good signal noise ratio were obtained and analyzed. Demonstration results for the observation of perovskite oxides with various A-site doping and bioactive glasses upon annealing were introduced. Notably, the PDF method can provide detailed local structural information of amorphous materials, liquids and structural disorder in crystalline materials. This work offers a guideline for people who considers to develop and use total scattering method at the other synchrotron facility.

Full Text

Preamble

Development of a High-Energy X-ray Total Scattering Apparatus at the Beijing Synchrotron Radiation Facility

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As a powerful probe of local structure, the high-energy X-ray total scattering method has been widely applied in condensed matter physics, materials science, and other fields. By employing a superconducting wiggler (SCW) and sagittal focusing monochromator, we have developed a high-energy X-ray total scattering apparatus at the 3W1 beamline of the Beijing Synchrotron Radiation Facility (BSRF), a first-generation synchrotron source. The apparatus primarily consists of a large two-dimensional flat-panel detector and high-energy X-rays in the 50-70 keV range, enabling total scattering measurements for pair distribution function (PDF) analysis across a Q range of 0.5-25 $Å^{-1}$. Based on this apparatus, we have developed a series of in-situ devices, obtained data with good signal-to-noise ratios, and performed detailed analyses. We present demonstration results from studies of perovskite oxides with various A-site dopings and bioactive glasses during annealing. Notably, the PDF method can provide detailed local structural information for amorphous materials, liquids, and structural disorder in crystalline materials. This work offers a guideline for researchers considering the development and application of total scattering methods at other synchrotron facilities.

Keywords: High energy X-ray, Total scattering, Beijing Synchrotron Radiation Facility, Pair distribution function

Introduction

Bragg diffraction enables experimental access to the atomic scale, providing the average long-range order of atomic structures in crystalline materials based on the assumption of lattice periodicity. However, long-range structural information alone is far from sufficient. On one hand, almost all crystals are imperfect, containing various inevitable defects such as point defects, dislocations, and chemical inhomogeneity [?]. Many properties of crystalline materials are strongly dependent on their local disorder structure. On the other hand, materials with amorphous nature preclude Bragg diffraction and necessitate alternative approaches for understanding their local structure [?, ?].

A variety of local structure probes exist, including extended X-ray absorption fine structure (EXAFS) [?], nuclear magnetic resonance (NMR), Raman spectroscopy [?, ?], and others. Among these techniques, pair distribution function (PDF) analysis based on high-energy X-ray total scattering has gained significant attention due to its capability for in-situ measurements and excellent signal-to-noise ratio. Total scattering technique involves measuring the complete diffraction pattern, including both Bragg and diffuse components, across a wide range of momentum transfer (Q) using synchrotron high-energy X-rays. From these measurements, one can obtain the weighted probability of finding atoms at certain distances from other atoms—the PDF.

The resolution of the PDF method is determined by the maximum Q value, Qmax (Δr 2π/Qmax). Qmax is limited by the X-ray wavelength λ, the detector area, and the sample-to-detector distance, since Q = 4π sin /λ, where 2 is the scattering angle. Decreasing λ is essential to achieve larger Qmax. In addition to the incident X-ray wavelength, high flux is needed to detect weak signals at high Q due to the rapid decrease of scattering intensity with increasing Q value. Meanwhile, low background is required, necessitating a well-collimated light source and a thin container with easily removable scattering contributions.

The advent of synchrotron radiation sources provides X-rays with high intensity and short wavelength, enabling accurate and reliable PDF data collection. To date, numerous beamlines with total scattering capabilities have been constructed, including beamline I15-1 at Diamond Light Source (DLS) [**?**], the Powder Diffraction beamline at the Australian Synchrotron [**?**], beamline 28-ID-1 at National Synchrotron Light Source II (NSLS-II) [**?**], beamline BL04B2 at Super Photon ring-8 (SPring-8) [**?**], and ID11 and ID15A at the European Synchrotron Radiation Facility (ESRF) [**?**, **?**].

It is worth mentioning that ESRF completed its Extremely Brilliant Source (EBS) upgrade in 2022, representing a new generation of synchrotron facilities. Over the past few decades, the total scattering method has been widely utilized in structural studies of liquids (including melts) \cite{13-16}, amorphous materials [**?**, **?**], disordered crystalline materials [**?**, **?**], and others. PDF has played a key role in understanding local structure, which is fundamentally important for amorphous and disordered systems.

The Beijing Synchrotron Radiation Facility (BSRF) is a first-generation synchrotron source with 2.5 GeV electrons in the storage ring operating in dedicated mode. BSRF operates in two modes: dedicated mode and parasitic mode. To utilize the parasitic mode, the original biological macromolecule crystallography station was moved to the 1W2 straight section, and 3W1 was transformed into a high-energy beamline station. In 2019, a superconducting wiggler (SCW) was successfully installed in the 3W1 beamline of BSRF, replacing the original permanent magnetic wiggler and offering a wider energy range from 40 keV to 80 keV with sufficiently high flux. These upgrades prompted us to develop a high-energy X-ray total scattering apparatus at the 3W1 beamline.

The aim of this study is to report the optimization process of this apparatus and benchmark total scattering measurements carried out at this beamline, demonstrating its capabilities for the user community.

II. Theoretical Basis

The scattering triangle of wavevectors in a typical total scattering experiment is shown in [Figure 1: see original paper], where **k** and **k**f represent the wavevectors before and after scattering, respectively. The scattering intensity includes both elastic and inelastic contributions, but elastic scattering dominates and determines the diffraction pattern. Here we assume no energy exchange between the incident quantum and the sample, such that $|\mathbf{k}\ | = |\mathbf{k}\text{f}| = 2\pi/\lambda$. The scattering vector is $\mathbf{Q} = \mathbf{k}\ $ - $\mathbf{k}$f, and for isotropic samples, $|\mathbf{Q}| =$ Q. Thus, the scattering vector magnitude is related to the incident wavelength λ and scattering angle 2 via: Q = 2$|\mathbf{k}\ |$ sin = $(4\pi/\lambda)$ sin .

In a scattering experiment, the structure factor S(Q) is related to the measured differential scattering cross-section I(Q) [**?**]:

$$S(Q) - 1 = \frac{I(Q) - \left[\sum_i c_i f_i(Q)\right]^2 - \sum_i c_i f_i^2(Q)}{\left[\sum_i c_i f_i(Q)\right]^2} - C(Q)$$

where c is the atomic fraction of element i, f (Q) is the X-ray atomic form factor, and C(Q) is the inelastic (Compton) scattering contribution. The pair distribution function G(r) can be calculated by Fourier transformation of S(Q) via:

$$G(r) - 1 = \frac{1}{2\pi^2 r \rho_0} \int_{Q_{\min}}^{Q_{\max}} Q[S(Q) - 1] \sin(Qr) dQ$$

where Qmin and Qmax represent the finite range in reciprocal space used during the Fourier transform, which is limited by the instrument' s Q range, and $_0$ is the atomic number density in Å^{-3} [**?**]. In addition to G(r), the total density function D(r) and total correlation function T(r) are often used in publications [**?**], where D(r) = 4π $_0$r[G(r) - 1] and T(r) = 4π $_0$rG(r). Peaks in T(r) indicate the presence of atoms with density exceeding the average number density at distance r, whereas valleys suggest the absence of atoms.

III. Light Source Parameters

The total scattering method was designed and developed at the 3W1 beamline of BSRF. Currently, 3W1 is the only beamline that can provide high-energy (E > 50 keV) X-ray flux at BSRF, equipped with a 2.3 T superconducting wiggler. A sagittal focusing monochromator with Si(111) crystals is positioned 32 meters from the source. With horizontal focusing from the monochromator, the beam size before the beam-defining slit (slit 1 in [Figure 3: see original paper]) is 1.7 (H) mm × 2.9 (V) mm. The photon energy range is 50–70 keV with energy resolution better than 0.65%. The beam is optimized by adjusting the bending radius of the second crystal before total scattering experiments. The calculated photon flux and Q-space resolution of the 3W1 beamline are shown in [Figure 2: see original paper]. From [FIGURE:2(b), subtle differences in ΔQ/Q at three energies are observed, and a value of 0.3% in the high-Q region can meet the requirements of many PDF experiments.

shows the parameters of the BSRF 3W1 beamline for total scattering experiments. The beamline provides photon energies of 50–70 keV with 0.65% energy resolution, using Mercu 1717HS and Mar345 detectors, and a beam size of 0.8 mm × 0.8 mm at the sample position.

IV. Apparatus Optimization Process

A. Experimental Setup

The high-energy X-ray total scattering apparatus was designed and implemented for the 50–70 keV energy range (wavelengths from 0.247–0.178 Å). Obtaining a clean background is crucial for total scattering experiments, as background signals must be reliably subtracted during data processing. A weak background signal is particularly important for weakly scattering samples such as thin films, nanomaterials, and other disordered systems. We utilized several pairs of slits and an evacuated stainless-steel tube with Kapton windows to reduce parasitic scattering and air scattering. Before experiments, all optics were carefully aligned.

The first slit assembly (labeled A in [Figure 3: see original paper]), located close to the incoming beam, defines the beam size and shape. The beam-defining slit consists of four tungsten blades 2 mm thick, whose positions can be adjusted with high-precision slides to define beam sizes of 0.5–0.8 mm. Parasitic scattering from the beam-defining slit is unavoidable, so a clean-up slit (C in [Figure 3: see original paper]) was designed to eliminate parasitic scattering from slit 1. Additionally, two 3 mm-thick lead shields were installed immediately after slit 1 and slit 2 to reduce background radiation. An evacuated stainless-steel tube maintained at 10 Torr was designed to minimize background from air scattering. Another clean-up slit (F in [Figure 3: see original paper]) and a large lead shield were placed just before the sample to remove scattering or diffraction generated by upstream components.

The detector was upgraded to the Mercu 1717HS, a high-speed large two-dimensional flat-panel detector that combines a pixelated detector with a CsI sensor featuring 139 µm pitch pixels in a 3072 × 3048-pixel array, providing a large effective area of 17 × 17 inches. With the Mercu 1717HS, a sample-to-detector distance of 180 mm, and an energy of 60 keV, a Qmax of 25 $Å^{-1}$ can be achieved.

It is worth mentioning that, benefiting from the high frame rate of the Mercu 1717HS (30 fps), it is possible to collect total scattering and diffraction data on subsecond timescales, although data quality may be limited by the relatively low photon flux. XRD patterns of 304 stainless steel during remelting were collected with a temporal resolution of 0.1 s.

B. Determination of Incident X-ray Energy

The most important parameter in PDF experiments, Qmax, is limited by the instrument and affected by the source energy. Higher energy yields larger Qmax values, while the X-ray scattering factor decreases rapidly with increasing Q. To achieve large Qmax and detect weak signals at high Q, both high energy and high flux are required. As shown in [FIGURE:2(a), substantial flux remains available at energies >50 keV, though photon flux decreases with increasing energy. Therefore, we first tested data quality at 50 keV, then gradually increased the energy to 60 keV and 70 keV, selecting the optimal energy by comparing comprehensive data quality.

Energy calibration was achieved using a "two-distance method" : (1) adjusting the sagittal monochromator to the target energy; (2) preliminary energy calibration using a LaB_6 standard sample and the sample-to-detector distance; (3) moving the sample stage by a known distance using a motor; (4) calculating the scattering angle 2 from the geometric relationship of the two LaB_6 positions and determining the exact energy using Bragg' s law.

At the beginning of the apparatus design and construction, a Mar345 detector was utilized and placed 180 mm downstream from the sample. To improve experimental efficiency and achieve larger Qmax values, we upgraded to the Mercu 1717HS detector. Diffraction patterns of CeO_2 at three energy points were collected with 60 s exposure using the Mar345 detector. The X-ray total structure factor S(Q) and pair distribution functions G(r) are displayed in [Figure 4: see original paper]. The maximum Q values of the instrument at 50 keV, 60 keV, and 70 keV are 25, 27, and 32 $Å^{-1}$, respectively. From the perspective of Qmax alone, 70 keV would be ideal. However, in the high-Q region, the S(Q) oscillations at 70 keV are more severe than at 50 keV and 60 keV due to lower flux. Considering both signal-to-noise ratio and Qmax, along with the small fluctuations in the low-r region ([FIGURE:4(b)), 60 keV was selected as the target energy for total scattering experiments. If necessary, the energy can be adjusted within the 50–70 keV range.

V. Benchmark Results

A. Ex-situ Total Scattering of High-Entropy Perovskite Oxides

For ex-situ experiments, samples were collected on Kapton tape and glued to aluminum alloy frames in an aluminum-tungsten-aluminum "sandwich" configuration to suppress stray scattering. Sample station repeatability is controlled to an accuracy of 0.1 mm using a magnetic base for sample exchange. The Mercu 1717HS detector was placed 200 mm downstream from the sample. The setup was calibrated using diffraction patterns from polycrystalline CeO_2 powder. Measurements were controlled by iDetector software with a 20 s exposure time. Background patterns were collected using the same setup and exposure time. Raw diffraction data were reduced from two-dimensional images and corrected for polarization and geometry effects using the Fit2D program [**?**]. Absorption, geometry, detector effects, and normalization were performed using PDFgetX3 [**?**].

$SrTiO_3$, a typical perovskite thermoelectric oxide, exhibits relatively good electrical properties [**?**]. The total scattering method was utilized to interpret the structure-property relationships of $SrTiO_3$-based oxides with different A-site dopings (La, Ba, Ca, and Pb). The X-ray total structure factors S(Q) and pair distribution functions G(r) for these materials are shown in [Figure 5: see original paper]. To extract more detailed structural information, 3D structural models were derived using Reverse Monte Carlo (RMC) simulation [?]. The starting configuration consisted of 5000 atoms. Several constraints were applied: atom-atom approach distances and Ti-O connectivity (all titanium atoms were coordinated to a certain number of oxygen atoms up to 2.5 Å). These constraints were chosen to avoid physically unrealistic structures. Structural information was derived by analyzing the atomic configurations generated by RMC simulations.

From the total scattering and RMC simulation results, A-O and Ti-O bond lengths for $SrTiO_3$-based perovskite oxides (ABO_3) with different entropies engineered at the A-sites were extracted to calculate the tolerance factor t via the equation t = (A-O length)/(B-O length). The tolerance factor is a key parameter in perovskites that reflects the correlation between relative element sizes and structural symmetry, where the deviation of t from 1 describes the extent of symmetry breaking [?]. In this work, as t approached 1, carrier mobility recovered, demonstrating that tolerance factor engineering could enhance weighted mobility and decouple carrier-phonon transport by tuning average element sizes to tailor symmetry and Ti displacement, revealing the structural origin of mobility recovery. This work was published in *Nature Communications* in cooperation with Tsinghua University [?].

B. In-situ Heating High-Energy Total Scattering Experiments

High-temperature experiments are among the most common and important for condensed matter physics and materials structure research. For example, the structural response of glasses to temperature and/or pressure is closely related to ionic transport properties, relaxation, and glass-forming transitions. PDF is one of the most effective methods for characterizing the local order and connectivity in glasses, making in-situ heating PDF experiments valuable for studying how different glass structures respond to temperature. As shown in [FIGURE:6(a), coupled with high-energy synchrotron radiation, a high-temperature and high-pressure device was designed with a large exit angle 2 $>$ 40°, capable of temperatures up to 873 K and pressures to 40 atm (BEIJING OPERANDO TECHNOLOGY CO., LTD).

Bioactive glasses are extensively utilized in orthopedics and dentistry, where structural rearrangement upon heating is a key factor determining processability. In cooperation with Shanghai Jiao Tong University, we tracked structural changes in Hench' s composition glass [?] using in-situ high-energy X-ray total scattering, with results presented in [FIGURE:6(b) and 6(c). As shown in [FIGURE:6(b), the first sharp diffraction peak (FSDP) exhibits minimal variation upon heating, indicating that medium-range order Si-O-Si rings/chains maintain approximate rigidity and cannot be depolymerized by heating below

Tg. This subtle FSDP change is also reflected in real-space structural correlations (as depicted in the G(r) plot in panel (c)). The shoulder peak around 3.2 Å remains almost constant, implying that Si-Si packing is intact and suggesting that these large Si-O-Si rings or chains are rigid. In contrast to the FSDP, the second peak in S(Q) patterns undergoes considerable change with increasing temperature, indicating nonnegligible rearrangement of short-range order (SRO) local structures upon heating. These SRO structural changes are clearly visualized in G(r) patterns, as the intensities of peaks at 2.4 Å and 3.6 Å decrease markedly at high temperatures, elucidating that modifier-oxygen and modifier-cation interactions are significantly weakened upon heating.

C. Total Scattering Coupled with Raman Spectroscopy and Acoustic Levitation

X-ray total scattering enables atomic-scale observation, while Raman spectroscopy is sensitive to chemical species in solution. To obtain chemical species information and local atomic structural evolution of solutions under low-gravity conditions, a multi-modal platform integrating an acoustic levitator, portable Raman spectrometer, and high-energy X-ray total scattering was constructed to study the structure of levitated aqueous solution droplets. The schematic of this platform is shown in [FIGURE:7(a). During experiments, solutions were injected as 0.5-2 mm^3 droplets, with the X-ray beam and Raman spectrometer focused simultaneously on the same droplet. Several colored droplets are shown in the inset of [FIGURE:7(b) for visual clarity, although the detected solutions were colorless. The scattering and Raman spectral data for aqueous M_2SO_4 (M = Li, Na, K) solutions are shown in [FIGURE:7(b) and 7(c), respectively.

This method is expected to be useful for studying precious samples (1.0 L) and for in-situ observation of chemical reactions in solution. It is noteworthy that, combined with laser heating and temperature-controlled systems, the levitation device is applicable for detecting the microstructure of high-temperature melts in a container-free environment, eliminating heterogeneous nucleation at the melt-container interface and increasing the propensity for supercooling. A humidity control system is also available, demonstrating the potential for accomplishing in-situ studies on the evaporation and crystallization processes of aqueous droplets. The levitation platform was established in cooperation with the Qinghai Institute of Salt Lakes, Chinese Academy of Sciences. Further research is described in publications by Yongquan Zhou et al. \cite{30-32}.

D. In-situ Tensile Testing of NiTi and Multi-Principal Element Alloys (MPEAs)

In addition to total scattering experiments, high-energy diffraction experiments coupled with an in-situ tensile frame were conducted with a longer sample-to-detector distance of approximately 800 mm, an energy of 60.05 keV, and a beam size of 0.7 × 0.7 mm. A Gatan microtension tester equipped with a home-built tensile jig was used for tensile testing. In-situ tensile tests were performed with a strain rate of 1×10^{-3} s^{-1}. The schematic of the in-situ tensile testing setup and raw diffraction patterns of SLM-NiTi alloy under different loading conditions are shown in [FIGURE:8(a). Two-dimensional diffraction patterns were converted into one-dimensional high-energy XRD spectra by caking and integrating over a \$±\$5° range along specified azimuthal angles ([FIGURE:8(b)), and diffraction peaks were fitted to Gaussian distributions.

Using these testing and diffraction methods, we investigated the anisotropic phase transition, deformation behavior, and orientation evolution of additively manufactured NiTi alloy during loading, in cooperation with Shanghai University. These results help understand the competition between stress-induced phase transformation and dislocation slip in AM NiTi alloys. We refer readers to relevant publications \cite{33–35}, where Pengyue Gao et al. explored stress-induced phase transformation and lattice correspondence in NiTi shape memory alloy during deformation using in-situ high-energy synchrotron X-ray diffraction. Additionally, the dislocation density of refractory multi-principal element alloys (MPEAs) under tension was determined using the Williamson-Hall method, shedding light on the synergistic combination of ultrahigh strength and large tensile ductility in these advanced materials \cite{36–39}.

VI. Conclusions

This paper describes the high-energy X-ray total scattering setups developed at the 3W1 beamline of BSRF. With an energy of 60 keV and a large-area detector, large quantities of high-quality data have been obtained and utilized in related research. The apparatus is compatible with various environments, including magnetic fields up to 2 T, a self-designed heating furnace with a temperature range of 293–873 K and pressure up to 40 atm, and a tensile frame with maximum tension of 2 kN and temperature of 823 K. This work demonstrates that the total scattering apparatus at 3W1 can collect PDF data with good signal-to-noise ratio for amorphous and disordered crystalline materials. It is worthwhile to mention that beamline scientists at BSRF are currently constructing a new synchrotron light source located in Huairou, Beijing, China.

VII. Perspectives for Total Scattering Techniques at HEPS

The High Energy Photon Source (HEPS), the first fourth-generation synchrotron light source in Asia, will be completed by the end of 2025. HEPS will accelerate electrons to energies of 6 GeV and produce high-energy beams capable of probing samples at nanometer scales. Its time resolution will be 10,000 times better than that achieved by third-generation synchrotrons [?]. Several beamlines at HEPS will provide total scattering capabilities, including the Structural Dynamics, Engineering Materials, and High Pressure beamlines.

The Structural Dynamics Beamline (SDB) of HEPS is dedicated to elucidating the dynamic behavior and structural transformations of materials under varying conditions. The SDB beamline provides dual advantages of high energy (70 keV) and high flux (10^{15} ph/s), enabling both conventional PDF measurements and time-resolved PDF studies with Qmax up to 25 Å^{-1} and 18 Å^{-1}, respectively, using a large-area detector and direct detection at MHz rates. Additionally, aerodynamic and ultrasound levitation techniques will be combined with PDF methods at SDB to study the structure of liquids, melts, and pharmaceutical drugs under containerless conditions.

The Engineering Materials Beamline (EMB) provides high-energy X-ray beams at 100 keV, suitable for conventional PDF experiments with larger Qmax and better spatial resolution. The High Pressure Beamline (HPB) features high-pressure capabilities, where PDF methods can be combined with high-pressure techniques to study system behavior under extreme conditions using monochromatic beams between 20 and 75 keV with a flux of 10^{12} ph/s. Both the Engineering Materials and High Pressure beamlines are equipped with Eiger2 XE 16M detectors, which will enable users to collect diffraction data with better signal-to-noise ratios. At the High Pressure and Structural Dynamics beamlines, X-ray beams will be focused to microscale dimensions, holding great promise for detecting solid-liquid interface structures during crystal growth, electrocatalysis, and other processes. More information about HEPS beamlines can be found online at: https://www.ihep.ac.cn/dkxzz/HEPS/xmgk/HEPSjj/.

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