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REVIEW ARTICLE



Understanding diffraction from disordered materials: toward the analysis beyond the intermediate-range order

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ABSTRACT

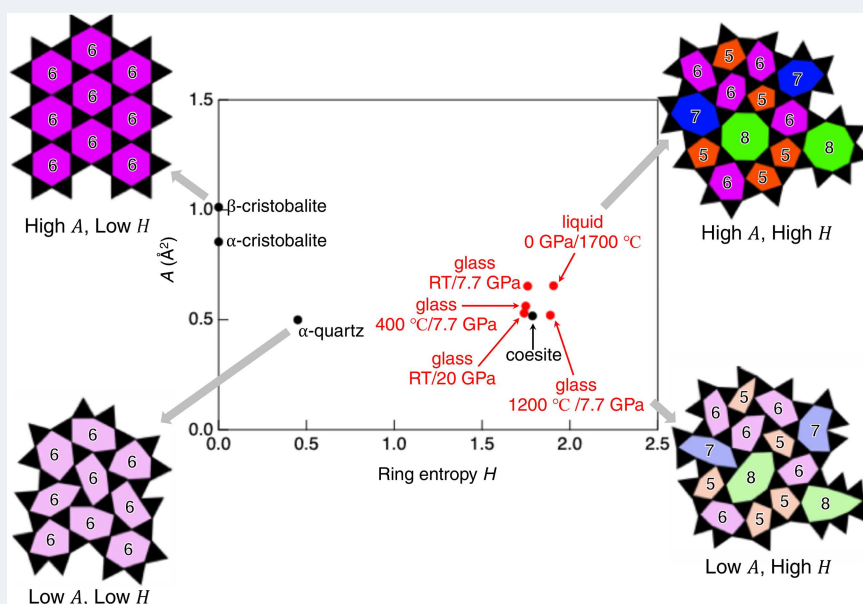
The construction of large quantum beam facilities such as synchrotron radiation and high-intensity proton accelerator facilities has provided access to high-intensity, high-energy quantum beams that are indispensable for the structural analyses of disordered materials via diffraction measurements. By the complementary use of X-rays, which are sensitive to heavy elements, and neutrons, which are sensitive to light elements, along with the advances in computer simulations and topological analysis techniques, we have achieved a deep understanding of the intermediate-range structure of disordered materials. In this review, we introduce the recent results obtained by the complementary use of X-ray and neutron diffraction. In particular, we discuss the differences among tetrahedrally coordinated disordered materials in comparison with dense random packing metallic glass to understand the origin of the three-peak structure, the first sharp diffraction peak (FSDP, Q_1), principal peak (PP, Q_2), and Q_3 observed in quantum beam diffraction data. Moreover, we demonstrate that comparing persistent homology analysis data with the ring size distributions of silica (SiO_2) glass and crystals is important for understanding the low-energy excitations observed in inelastic neutron scattering data.

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1. Introduction

Understanding the structure of non-crystalline materials is still challenging in physics because of the inherent nature of disorder in non-crystalline materials. Diffraction is a powerful experimental method to reveal the atomic structure of crystalline materials because crystallographic theory, which has been developed through the understanding of Bragg peaks observed in diffraction data, is well established. However, the diffraction patterns of non-crystalline materials exhibit the so-called halo patterns, owing to the inherent disorder in their atomic arrangements. The pair distribution function (PDF), obtainable as the Fourier transform of normalised diffraction data, characterises the short-range structure: atomic distances and first coordination numbers. The atomic arrangement beyond the short-range structure, the so-called intermediate (medium)-range order, has been widely studied using X-ray and neutron diffraction from the late 1980s [1]. Furthermore, the intermediate-range order of disordered materials has been discussed in relation to the boson peak observed in Raman scattering, inelastic neutron scattering (INS), and heat capacity measurements, which is a signature of low-energy dynamics [2]. Advanced quantum beam facilities developed in the late twentieth century can provide high-flux synchrotron X-rays and neutrons, enabling diffraction measurements with high throughput and high real-space resolution. Combined with advanced computer simulation techniques that facilitate the interpretation of diffraction peaks, these methods can reveal the intermediate-range order of disordered materials [3–10]. Moreover, intermediate-range order in disordered materials is revealed in terms of topological characteristics: ring size distribution and ring shape [11,12] with the concept of ring persistency [13,14] and ring entropy [14]. Finally, we introduce the atomic structure of non-crystalline materials beyond the intermediate-range order by utilising nanoscale structural information derived from extended-range PDF, which can project the information arising from low- Q (Q is defined in Section 2) diffraction peaks into real space.

2. PDF and total structure factor

The most common function used to describe the structure of disordered materials is the pair distribution function $g(r)$, which is obtained by a Fourier transform of the normalised diffraction data, the Faber–Ziman [15] total structure factor $S(Q)$.

$$g(r) = 1 + \frac{1}{2\pi^2 r \rho} \int_{Q_{\min}}^{Q_{\max}} Q [S(Q) - 1] \sin(Qr) M(Q) dQ \quad (1)$$

Here, ρ represents the atomic number density. $Q = (4\pi/\lambda) \sin\theta$, where 2θ is the angle and λ is the wavelength of the incident X-rays or neutrons) represents the magnitude of the scattering vector. $M(Q)$ is a modification function introduced to reduce the ripples caused by the truncation error of $S(Q)$ in a finite Q range in the Fourier transform of $S(Q)$, for which the Lorch function [16] has been widely used. The relationships among the pair distribution function $g(r)$, the reduced pair distribution function $G(r)$, the total correlation function $T(r)$, and the radial distribution function $RDF(r)$ are expressed as follows.

$$g(r) = \frac{G(r)}{4\pi r \rho} + 1 \quad (2)$$

$$T(r) = G(r) + 4\pi r \rho = 4\pi r \rho g(r) \quad (3)$$

$$RDF(r) = rG(r) + 4\pi r \rho = rT(r) \quad (4)$$

When a disordered material is composed of n types ($n \geq 2$) or more of atoms, the X-ray total structure factor $S(Q)$ is the weighted sum of the partial structural factor $S_{ij}(Q)$ values corresponding to the correlations between atoms of the same or different types.

$$S^X(Q) = \sum_{i=1}^n \sum_{j=1}^n W_{ij}(Q) S_{ij}(Q) \quad (5)$$

$$\langle f(Q) \rangle^2 = (\sum_{i=1}^n c_i f_i(Q))^2 \quad (6)$$

Here, c_i and $f_i(Q)$ are the molar fraction and atomic scattering factor of atom i , respectively, and $W_{ij}(Q)$ is the weighting factor defined by c_i and $f_i(Q)$, where $W_{ij}(Q) = \frac{c_i c_j f_i(Q) f_j(Q)}{(f(Q))^2}$. In the case of SiO_2 glass,

$$S_{\text{SiO}_2}(Q) = W_{\text{Si-Si}}(Q) S_{\text{Si-Si}}(Q) + 2W_{\text{Si-O}}(Q) S_{\text{Si-O}}(Q) + W_{\text{O-O}}(Q) S_{\text{O-O}}(Q). \quad (7)$$

When $f_i(Q)$ is replaced with the atomic number, i.e. $f_i(Q=0)$, the relationships of the X-ray structure factor $S^X(Q)$ and the neutron structure factor $S^N(Q)$ obtained by replacing $f_i(Q)$ with the neutron scattering length b_i with the $S_{ij}(Q)$ are expressed as follows.

$$S^X(Q) = 0.218S_{\text{Si-Si}}(Q) + 0.498S_{\text{Si-O}}(Q) + 0.284S_{\text{O-O}}(Q) \quad (8)$$

$$S^N(Q) = 0.069S_{\text{Si-Si}}(Q) + 0.388S_{\text{Si-O}}(Q) + 0.543S_{\text{O-O}}(Q) \quad (9)$$

Note that the approximation of the atomic form factor by atomic number seems to work in a low- Q region, but Q -dependent changes become more significant in a high- Q region (see Figure S1).

3. Topological analyses

In this section, we present two important topological analysis tools, ring size and persistent homology analyses, and introduce the concepts of topological order-disorder [17], ring persistency [13,14], and ring entropy [14].

3.1 Ring size distribution

The history of ring size distribution analysis is relatively long. The most widely used software was developed by Roux and Jund [18,19]. There are various criteria for a ring: the King [20], Guttman [21], primitive [22,23,24] (or irreducible [25,26]), and strong ring [23,24] criteria. Note that both the King and Guttman criteria are based on the shortest path, whereas the primitive criterion is the basis for detecting large rings because a primitive ring is defined as a ring that cannot be decomposed into two smaller rings. Details are described in ref. [18].

3.2 Persistent homology analysis

Topological data analysis has rapidly progressed and has provided several tools for analysing multiscale data in physical and biological fields [27]. Following the landmark study of Hirata et al. [28], Hiraoka et al. applied persistent homology to disordered materials to understand the homology of rings, whose features cannot be detected by conventional ring size distribution analysis [27]. The input to persistence diagrams (PDs) is given by atomic configurations, and the output is expressed as two-dimensional histograms. Specific distributions such as curves and islands in the PDs are used to identify the meaningful shape characteristics of a cycle on the basis of the given atomic configuration.

The homology of atomic configurations can be calculated on the basis of the PDs obtained by using the HomCloud software package [29]. Given a set of points in space, persistent homology captures the topological multiscale structures, and the structures identified are compactly expressed in a PD. The construction of the PD follows the process schematically shown in Figure 1(a) [27]. We first replace each point by a sphere and increase the radius from zero to a sufficiently large value, which corresponds to the changing resolution of input x, y, z coordinates of atoms. Then, we record the pair of radii (b, d) at which a ring in a specific location appears (birth) and disappears (death), respectively. The PD is a histogram of the birth/death plane with counts of rings at the coordinate (b, d). This construction enables one not only to count the number of rings (cycles), but also to characterise their shapes at a multiscale, by observing the characteristic shapes of the diagrams themselves.

Typical examples of birth/death pairs for typical regular structures are shown in Figure 1(b)–1(d). For a regular hexagonal arrangement of points, in which the distance between points is a , the ring appears at radius $a/2$ and disappears at radius a , as shown in Figure 1(b). For a regular triangular configuration, the ring appears at $a/2$ and disappears at $\sqrt{1/3}a \approx 0.577a$, as shown in Figure 1(c). The one-dimensional PD for regular hexagonal/triangular points is shown in Figure 1(d). In this section, PDs are being used for investigating rings and polyhedral formations in atomic configurations. We also note that the detected

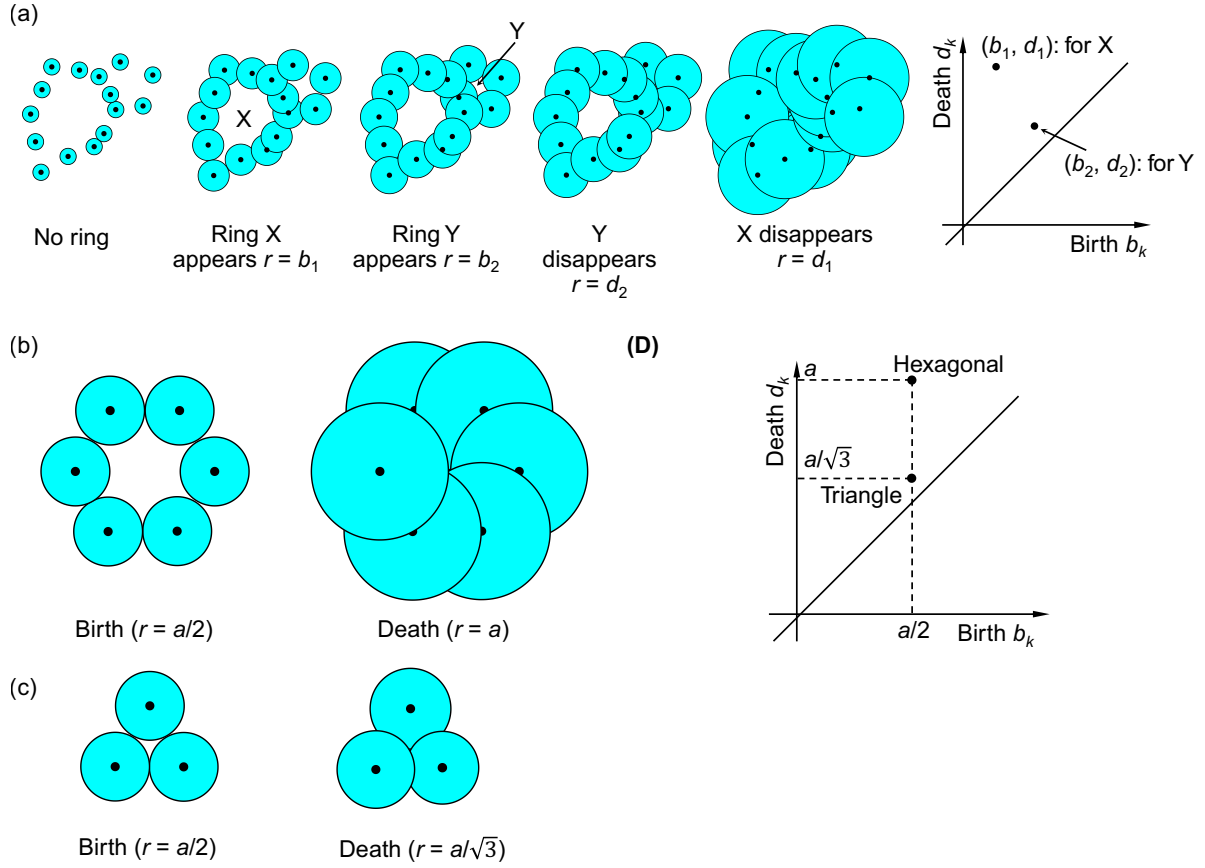


Figure 1. Persistent homology and PD [27]. (a) Sequence of increasing radius of spheres for input data (left). The PD (right) is obtained as a histogram, counting the number of rings on the birth–death plane. (b), (c) Appearance and disappearance of a ring for a regular hexagon/triangle. (d) Pairs of birth and death radii for hexagon and triangle in the one-dimensional PD.

rings are recorded during the computation of the diagrams, and hence, we can explicitly identify their geometric shapes.

3.3 Topological order–disorder

The concept of topological order was proposed by Gupta in 1993 [17]. Figure 2 shows the schematics [17] and primitive ring size distributions in cristobalite (crystalline SiO_2) (a) and SiO_2 glass obtained by molecular dynamics (MD)-reverse Monte Carlo (RMC) modelling (b) [12,30]. The schematics are two-dimensional analogues of the corner-sharing tetrahedral network motif in both materials. A representative crystalline SiO_2 , cristobalite, possesses only sixfold rings (i.e. it consists of six SiO_4 tetrahedra), whereas SiO_2 glass has various ring sizes from threefold to tenfold rings, although sixfold rings are predominant. Gupta characterised that the former is topologically ordered and the latter is topologically disordered [17].

3.4 Ring entropy and ring persistency

Kohara et al. have recently proposed a new analysis method to clarify the ring formation in SiO_2 glasses, liquid, crystals [14]. To quantify the breadth of the ring size distribution, we define the ring entropy H as

$$H = - \sum_n F_n \ln(F_n), \quad (10)$$

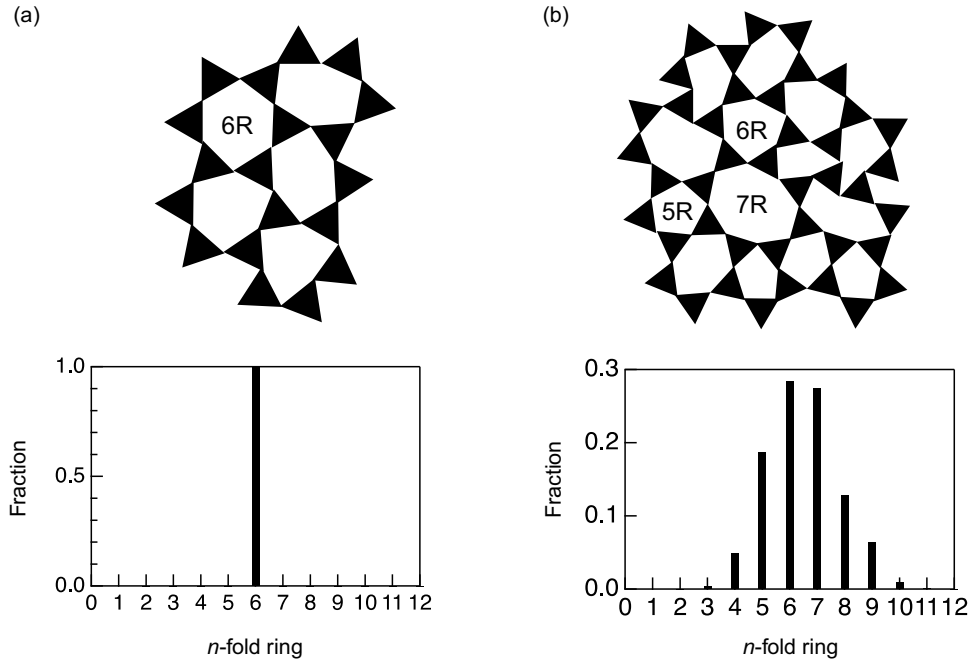


Figure 2. Two-dimensional schematics of (a) cristobalite and (b) SiO₂ glass together with the primitive ring size distributions obtained by MD-RMC modelling [12,30].

where F_n denotes the fraction of Si–O rings with n -fold number of atoms in the primitive ring size distribution of each SiO₂ glass or crystal. H is zero when all rings have the same size, and it increases as the ring size distribution broadens. Thus, H provides a useful measure of the breadth of the ring size distribution. As mentioned in the previous section, the concept of a topologically disordered network was proposed on the basis of ring size distribution. Accordingly, the ring entropy H , which provides the degree of topological disorder, is introduced to quantitatively characterise the breadth of rings.

The average persistence of Si cycles, A , can be defined as

$$A = \sum_n F_n \frac{p_n}{n}, \quad (11)$$

where P_n is the persistence of n -fold Si cycles defined by $d_k - b_k$ derived by persistent homology analysis [27,29]. A characterises the overall degree of symmetry of the rings. Lower and higher A values correspond to more distorted and more symmetrical ring shapes, respectively.

4 High-resolution PDF data obtained by synchrotron X-ray and neutron diffraction measurements

The advent of third-generation synchrotron radiation facilities and spallation neutron sources enables us to access high- Q diffraction data, yielding a high resolution in the Fourier transformed real-space functions, because the resolution depends on $Q_{\max}$ value (see Equation (1)) in the Fourier transform [31]. Figure 3 shows X-ray and neutron total correlation functions, $T(r)$, of SiO₂ glass [8] obtained by a Fourier transform with $Q_{\max} = 30 \text{ \AA}^{-1}$. We can observe well-resolved Si–O, O–O, and Si–Si peaks at 1.61, 2.64, and 3.08 Å, respectively. The Si–O coordination number is found to be 3.9 ± 0.1 .

5 Understanding diffraction peak of various disordered materials

Figure 4 shows the X-ray total structure factor, $S^X(Q)$, of amorphous Si [32,33] and the neutron total structure factors, $S^N(Q)$, of glassy SiO₂ [34] and liquid CCl₄ [35], together with the $S^X(Q)$ of a SiSi₄

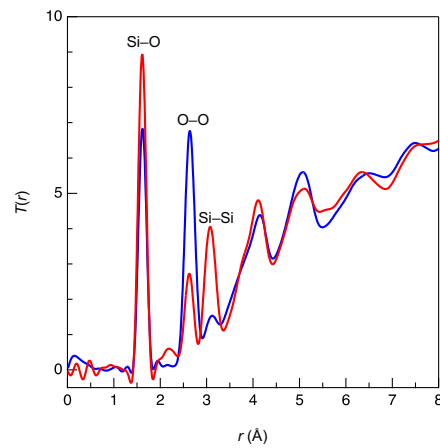


Figure 3. X-ray (red) and neutron (blue) total correlation functions, $T(r)$, of SiO_2 glass [8]. Fourier transforms of the $S(Q)$ were performed with $Q_{\text{max}} = 30 \text{ \AA}^{-1}$. A Lorch [16] function was used as the modification function.

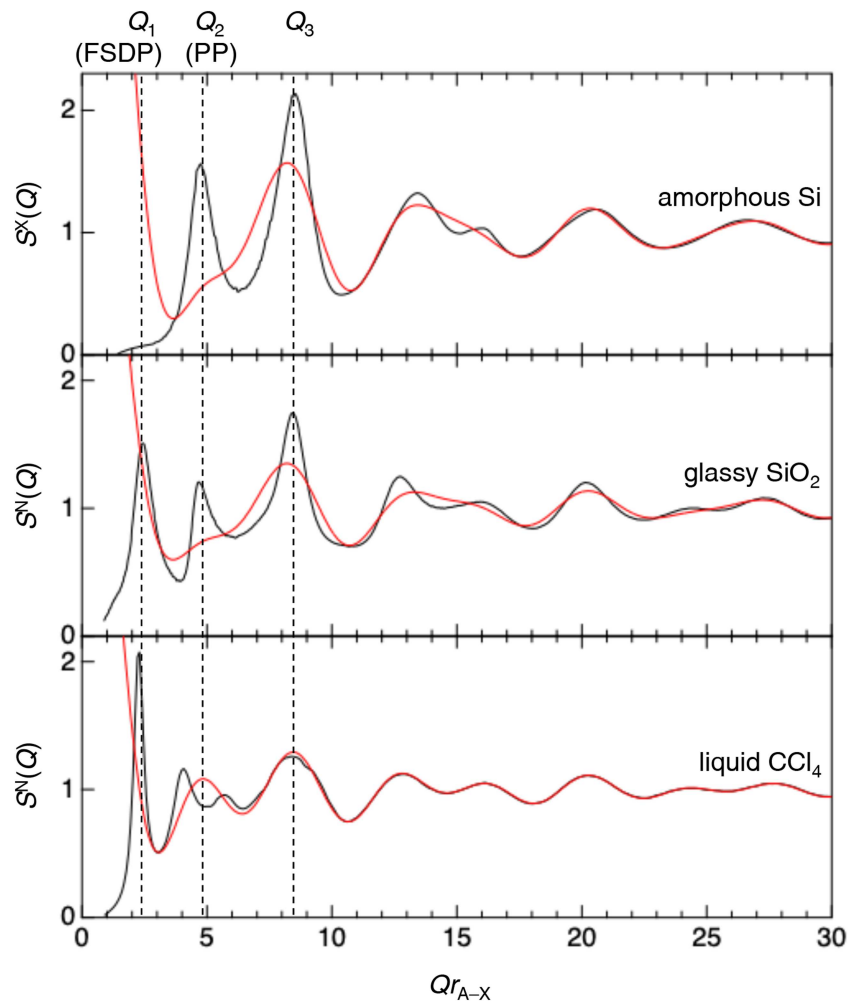


Figure 4. X-ray total structure factor, $S^X(Q)$, of amorphous Si (top) [32,33] and neutron total structure factors, $S^N(Q)$, of glassy SiO_2 (middle) [34] and liquid CCl_4 (bottom) [35], together with the $S^X(Q)$ of a SiSi_4 tetrahedron (top) [14] and the $S^N(Q)$ of a SiO_4 tetrahedron (middle) [14], and a CCl_4 tetrahedron (bottom) [14] calculated using the Debye scattering function [36] (see ref. [14] in details). Black curves, experimental data; red curves, calculated data.

tetrahedron [14] and the $S^N(Q)$ of a SiO_4 tetrahedron [14] and a CCl_4 tetrahedron [14], calculated using the Debye scattering function [36] (see ref. [14] for details). The scattering vector Q is scaled by multiplying it by r_{A-X} (distance between the centre and a corner of a tetrahedron). It is confirmed that the FSDP and PP cannot be reproduced using a tetrahedron in the case of glassy SiO_2 . A CCl_4 model shows a single PP, but experimental data show a doublet peak attributable to the orientational correlation [37,38] of CCl_4 molecules. Since the hard-sphere Monte Carlo (HSMC, RMC without experimental data) [9,38] model gives a single PP [39], only the Q_3 of liquid CCl_4 can be reproduced by a CCl_4 tetrahedron, probably because of the non-network structure in which CCl_4 molecules are isolated. These results suggest that the Q_3 peak can be reproduced by a tetrahedron in the case of a molecular liquid, in which no network is formed. In the case of covalent network tetrahedra in amorphous Si and glassy SiO_2 , the origin of Q_3 is both the inter- and intra-tetrahedral correlations. Note that the Q_3 of liquid Hg can be reproduced by HSMC simulation with a cut-off distance of 2.6 Å [30].

Figure 5(a) shows the X-ray total structure factors, $S^X(Q)$, of glassy $\text{Cu}_{50}\text{Zr}_{50}$ [40] and amorphous Si [32,33], together with the neutron total structure factors, $S^N(Q)$, of glassy SiO_2 [34] and liquid CCl_4 [35]. The scattering vector Q is scaled by multiplying it by d , that is, the first-neighbour distance in real space. It is confirmed that glassy $\text{Cu}_{50}\text{Zr}_{50}$ has no distinct FSDP or PP, and amorphous Si has no FSDP. Figure 5(b) shows atomic configurations together with coordination numbers (N) of glassy $\text{Cu}_{50}\text{Zr}_{50}$ [40], amorphous Si [32,33], glassy SiO_2 [30], and liquid CCl_4 [35]. Glassy $\text{Cu}_{50}\text{Zr}_{50}$ exhibits a dense random packing structure [41] where the coordination number is approximately 12. Amorphous Si has a perfect SiSi_4 tetrahedral network, and glassy SiO_2 has a SiO_4 tetrahedral network in which Si atoms are fourfold coordinated and O atoms are twofold coordinated. The short-range structure of liquid CCl_4 is an isolated CCl_4 tetrahedron, as mentioned above. A comparison of four disordered materials suggests that the packing of atoms governed by chemical bonds, manifested by a well-defined coordination number, has

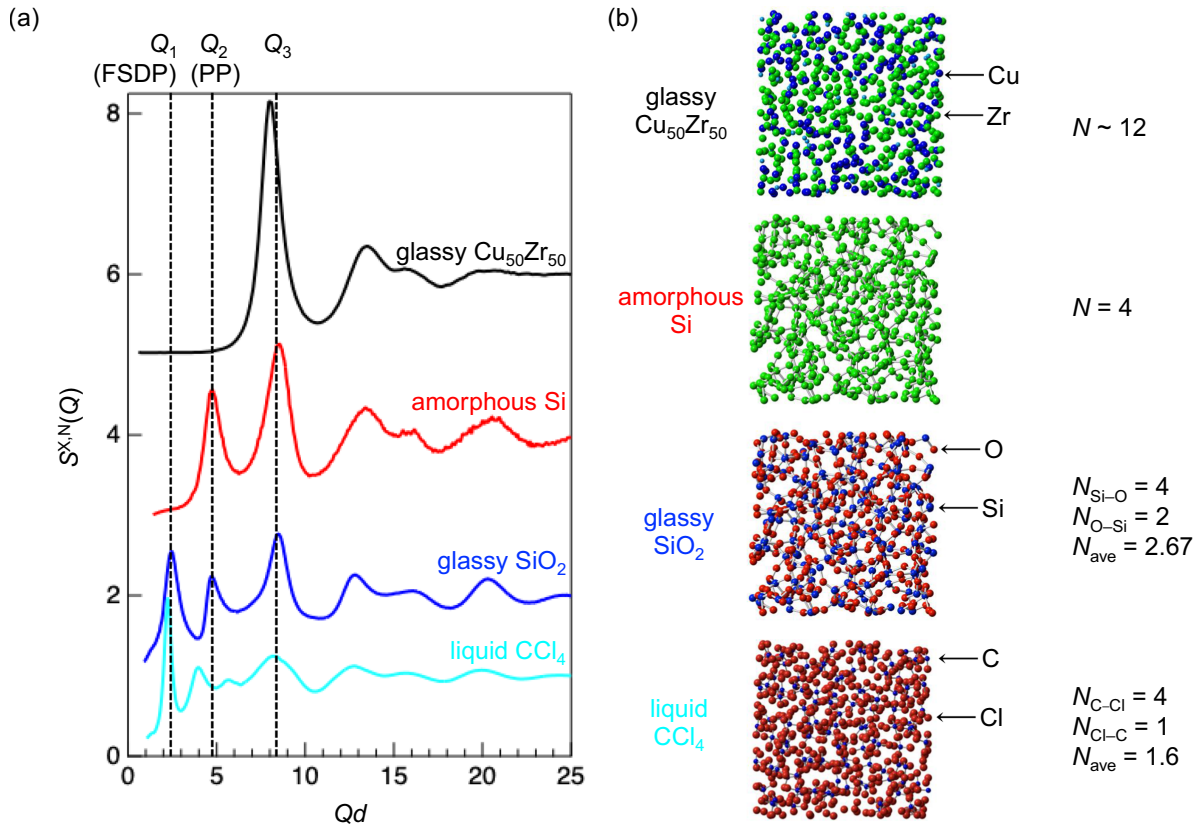


Figure 5. (a) X-ray total structure factors, $S^X(Q)$, of glassy $\text{Cu}_{50}\text{Zr}_{50}$ [40] and amorphous Si [32,33], together with the neutron total structure factors, $S^N(Q)$, of glassy SiO_2 [34] and liquid CCl_4 [35]. $S(Q)$ data are scaled by d (the first-neighbour distance in real space). d is identical to r_{A-X} in Figure 4 in the case of amorphous Si, glassy SiO_2 , and liquid CCl_4 . (b) Atomic configurations of glassy $\text{Cu}_{50}\text{Zr}_{50}$ [40], amorphous Si [32,33], glassy SiO_2 [34], and liquid CCl_4 [35], and coordination numbers N .

significant effects on the appearance of diffraction peaks: in particular, FSDP and PP are signatures of the formation of covalent bonds in disordered materials.

Although the concept of intermediate-range order was already established in the last century, the understanding of diffraction peaks associated with the formation of intermediate-range order is gradually becoming apparent only nowadays. This progress was made possible by the emergence of advanced synchrotron X-ray and neutron facilities in the 1990s, which provide high-quality diffraction data. Such progress was not foreseen in the 1980s: e.g. Egelstaff pointed out the difficulties of obtaining high-quality diffraction data of disordered materials in his article published in 1983 [42]. The progress in computer simulation and the development of data analysis techniques must be noted as well.

6 Understanding quantum beam diffraction data for SiO₂ glass

SiO₂ is the most important glass forming material that can form glass as a single component. SiO₂ glass has a tetrahedral (SiO₄) corner-sharing motif. Figure 6 shows the neutron (a) and X-ray (b) total structure factors, $S^{X,N}(Q)$, of SiO₂ glass together with neutron- and X-ray-weighted partial structure factors, $w_{ij}S_{ij}(Q)$ [30,43]. Neutron diffraction data show a three-peak structure [44,45] consisting of Q_1 (first sharp diffraction peak, FSDP [1,44,45]), Q_2 (principal peak, PP [44,45]), and Q_3 , whereas X-ray diffraction data do not exhibit Q_2 (Q is the scattering vector magnitude). This behaviour can be explained by the neutron- and X-ray-weighted $S_{ij}(Q)$, in which two positive peaks of $S_{\text{Si-Si}}(Q)$ and $S_{\text{O-O}}(Q)$ are counter-balanced by a negative peak of $S_{\text{Si-O}}(Q)$ at the position of Q_2 . This negative peak is observed in A-X-type tetrahedrally coordinated disordered materials [14], and its origin is discussed in ref. [46].

The concept of intermediate (medium)-range order manifested by FSDP has been discussed for a long time, but basic concepts were proposed in 1980s. The most sensible concept was proposed by Price et al. [1]. They analysed the position of FSDP scaled by the first-neighbour distance in real space to clarify the origin of intermediate-range order. The length scale of intermediate-range order can be described by periodicity ($=2\pi/Q_{\text{FSDP}}$) and coherence length ($2\pi/\Delta Q_{\text{FSDP}}$). Salmon and Zeidler extensively studied the length scale of intermediate-range order for various disordered materials [47]. In the case of SiO₂ glass, the lengths estimated on the basis of X-ray diffraction data for periodicity and coherence length are ~ 4 Å and ~ 10 Å, respectively [30,43]. This length scale can be observed in an atomic configuration derived from MD-RMC modelling (Figure 7a [30,43]), in which periodicity and coherence length are highlighted by

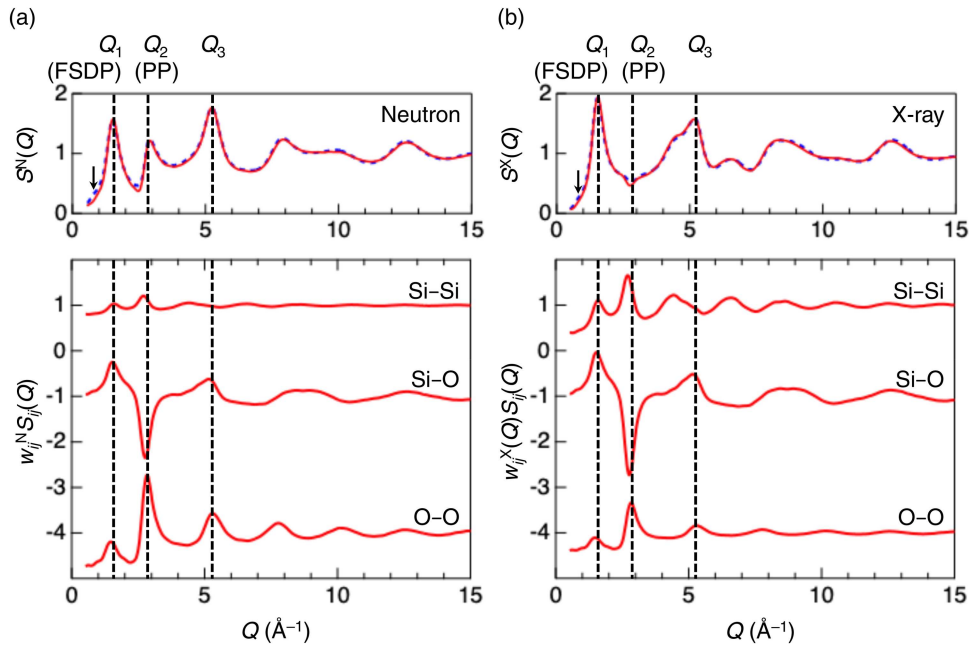


Figure 6. Neutron (a) and X-ray (b) total structure factors, $S^{X,N}(Q)$, of SiO₂ glass together with neutron- and X-ray-weighted partial structure factors, $w_{ij}S_{ij}(Q)$ [30,43]. Dashed lines are a guide to the eyes.

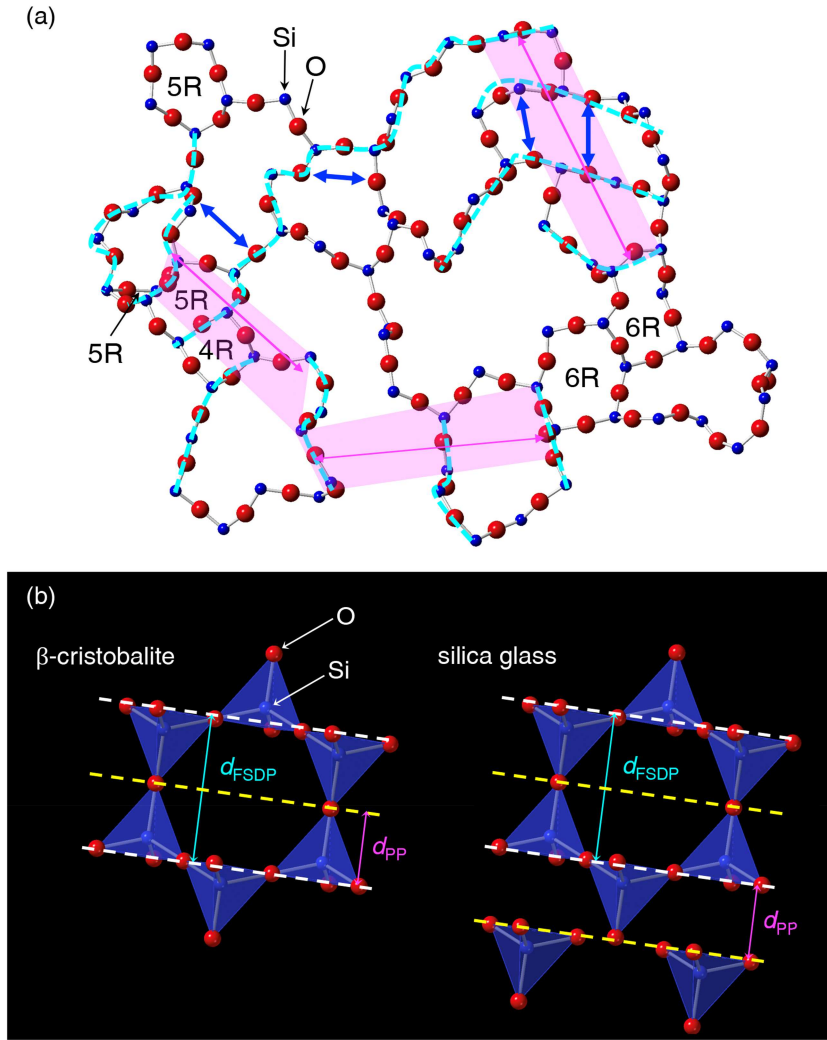


Figure 7. (a) Atomic configuration of SiO₂ glass derived from MD-RMC modelling [30,43]. The thickness of the cell is approximately 9 Å and only the Si and O atoms belonging to the network are shown. (b) Atomic configuration of β -cristobalite (left) [48] and its modified configuration for SiO₂ glass (right) [14].

thick double-headed arrows (blue) and thin double-headed arrows (magenta).

The origin of PP was proposed on the basis of the atomic structure of β -cristobalite. According to Benmore and Wilding, the periodicity of PP corresponds to the distance from the base to the apex of a SiO₄ tetrahedron (d_{PP}), as shown in Figure 7(b, left) [48]. However, this interpretation cannot explain the reason why the PP of SiO₂ glass evolved under high pressures at room temperature [49], where the Si–O coordination number remained four. Recently, we have reported the formation of inter-tetrahedral correlations in SiO₂ glass and modified it as shown in Figure 7(b, right) [14]. It is suggested that the ordering of intertetrahedral oxygen–oxygen correlation under high pressures associated with the reduction in cavity volume is the origin of the pp that evolved under high pressures.

7 Topological order–disorder in SiO₂ glass and crystals

As mentioned in the previous section, the concept of topological order was proposed by Gupta [17]. The primitive ring size distributions in crystalline SiO₂ with the corner-sharing tetrahedral motif, namely, α -cristobalite ($d = 2.327 \text{ g cm}^{-3}$), α -quartz ($d = 2.655 \text{ g cm}^{-3}$), coesite ($d = 2.905 \text{ g cm}^{-3}$), and SiO₂ glass ($d = 2.2 \text{ g cm}^{-3}$) [30], are shown in Figure 8(a)–(d). It is found that high-density SiO₂ crystal phases (α -quartz and coesite) are not topologically ordered, although the fraction of sixfold rings in α -quartz is indistinct.

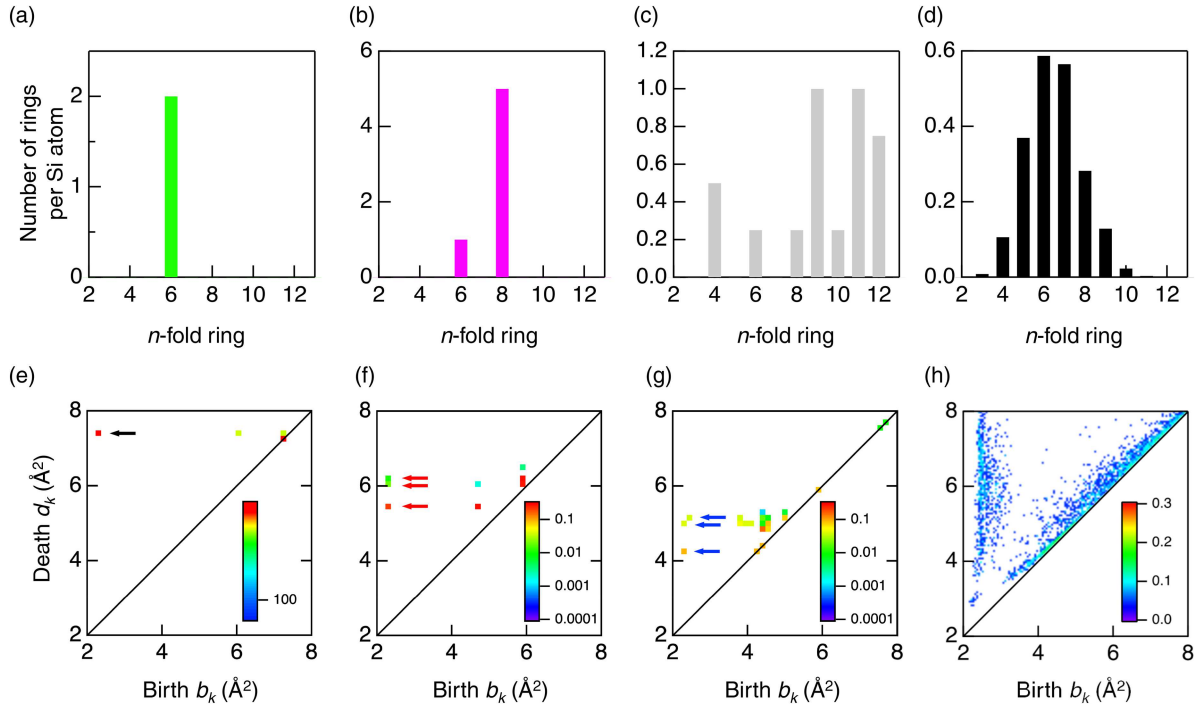


Figure 8. (a–d) Primitive ring size distributions in (a) α -cristobalite, (b) α -quartz, (c) coesite, and (d) SiO₂ glass. (e–h) Si-centric PDs of (e) α -cristobalite, (f) α -quartz, (g) coesite, and (h) SiO₂ glass [30].

Figure 8(e)–8(h) show Si-centric PDs, which provide information on ring shape, of α -cristobalite ($d = 2.327 \text{ g cm}^{-3}$), α -quartz ($d = 2.655 \text{ g cm}^{-3}$), coesite ($d = 2.905 \text{ g cm}^{-3}$), and SiO₂ glass ($d = 2.2 \text{ g cm}^{-3}$) obtained by MD-RMC modelling [30]. The death value at the intense point profile of α -cristobalite (indicated by black arrow) decreases, whereas the number of points increases in the PDs of α -quartz (red arrows) and coesite (blue arrows), suggesting that the rings become increasingly distorted with the density change from α -cristobalite to α -quartz and coesite. On the other hand, the Si-centric PD of SiO₂ glass shows a distinct profile along with the death axis, demonstrating that SiO₂ glass includes the homology of a series of SiO₂ crystals. The combination of ring size distribution analysis and persistent homology analysis provides us with more crucial topological information than was hitherto available.

The variety of ring size distribution in SiO₂ glass and crystals is very important to understand the dynamics of SiO₂ glass in terms of the boson peak, which is observed in the low-frequency region in Raman scattering data and low-energy band in INS data. Onodera et al. have recently reported the INS data of a series of SiO₂ glasses and crystals [50], with which it is possible to discuss the relationship between dynamics and topological order–disorder. Intermediate-range order in SiO₂ glass has been discussed in terms of the low-frequency dynamics of a glass network manifested by the boson peak observed in Raman scattering [51,52] and INS data [53].

Figure 9 shows contour maps of the dynamical structure factors, $S(Q, E)$, of SiO₂ crystals and a series of densified glasses obtained by INS measurements [10,50]. All data show a distinct low-energy band, and one of the most important features in the $S(Q, E)$ is that neither the scattering in the region of the FSDP in the neutron $S(Q)$ data at $Q_{\text{FSDP}} \sim 1.50\text{--}1.87 \text{ Å}^{-1}$ nor the scattering at $Q_{\text{FSDP}/2}$ has a contribution toward the boson peak. Instead, the main contributions towards $S(Q, E)$ arose from Q -values associated with the PP at $Q_{\text{PP}} \sim 2.81\text{--}2.98 \text{ Å}^{-1}$. Note that the densified glasses were synthesised by hot compression (RT/7.7 GPa ($d = 2.24 \text{ g cm}^{-3}$), 400 °C/7.7 GPa ($d = 2.54 \text{ g cm}^{-3}$), and 1200 °C/7.7 GPa ($d = 2.72 \text{ g cm}^{-3}$)) [50]. It is found that the peak position in energy shifts to the higher-energy side with increasing density as reported in refs. [10,50,53]. Among the three spectra for the crystalline phases, only the $S(Q, E)$ of coesite is very broad, and the coesite spectrum appears similar to glass spectra, in which a distinct boson peak is observed at $Q \sim 3 \text{ Å}^{-1}$. This behaviour implies that the dynamics of coesite is rather similar to that of glass even though diffraction data shows prominent Bragg peaks probably

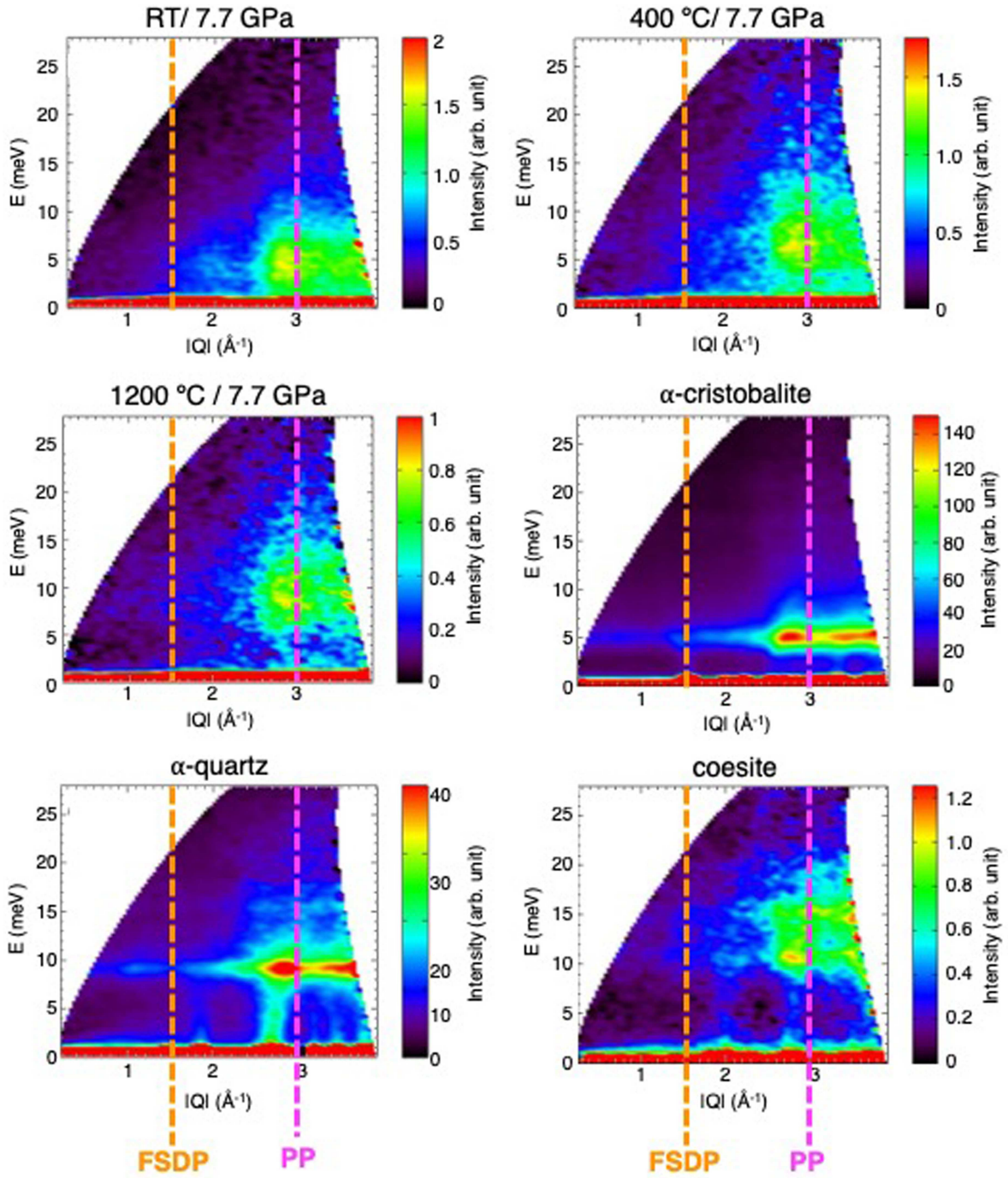


Figure 9. Contour maps of the dynamical structure factors, $S(Q, E)$, of a series of SiO_2 crystals and densified glasses [10,50]. Dashed lines are a guide to the eyes.

owing to high density. Figure 10 shows the dynamical structure factors, $\langle S(Q, E) \rangle$, of a series of SiO_2 crystals and densified glasses [10,50]. RT/7.7 GPa glass shows a prominent boson peak at E of ~ 5 meV, and the peak shifts towards a large value with increasing density. Crystal phases show similar behaviour, but both α -cristobalite and α -quartz show a sharp low-energy peak, whereas coesite shows very broad peaks that stretch towards higher energies. We consider that this behaviour may be related to topological disorder in coesite. Moreover, the variation in ring size is a key feature in understanding the dynamics manifested by the boson peak in SiO_2 glass.

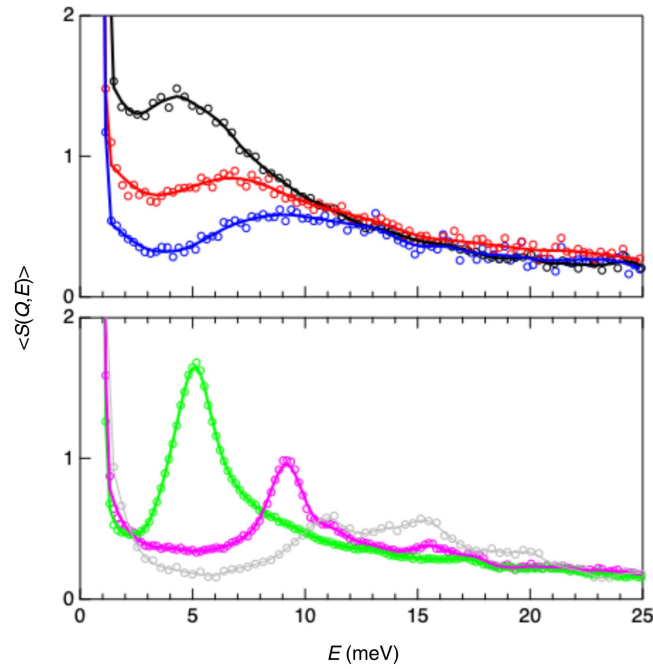


Figure 10. Dynamical structure factor $\langle S(Q,E) \rangle$ of SiO_2 crystals and densified glasses. glasses (RT/7.7 GPa (black), 400 °C/7.7 GPa (red), 1200 °C/7.7 GPa (blue)), α -cristobalite (green), α -quartz (magenta), and coesite (grey) [10,50].

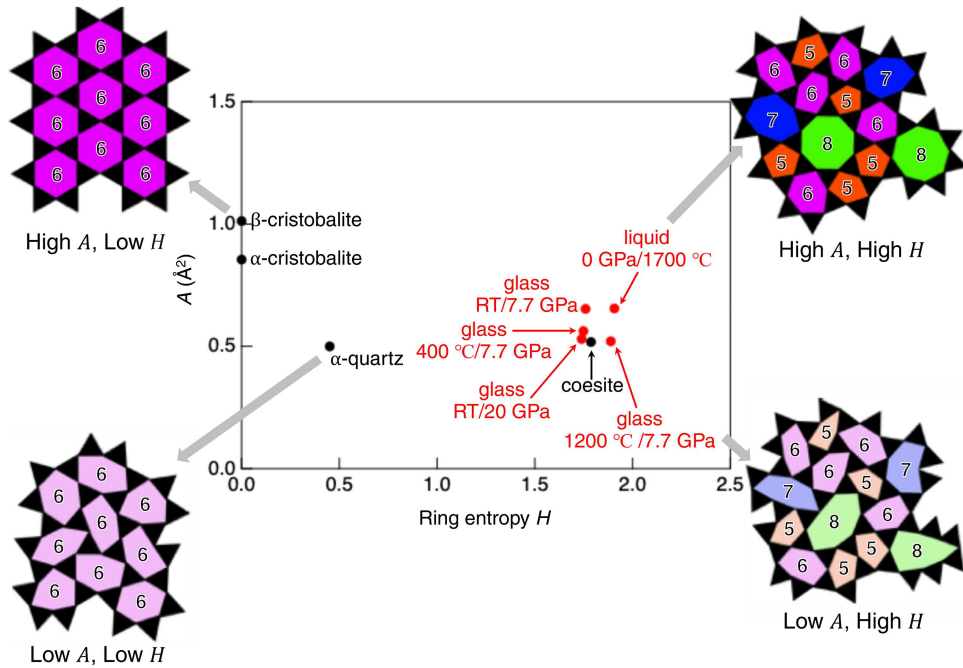


Figure 11. Two-dimensional plot of A versus H for SiO_2 crystals and densified glasses, and liquid [14].

8 Ring entropy and ring persistency in SiO_2 glasses, liquid, and crystals

Figure 11 shows a two-dimensional plot of A versus H for SiO_2 glasses, liquid, and crystals. Since both α - and β -cristobalites consist only of sixfold rings [50], their H values are zero. On the other hand, the ring shapes in β -cristobalite are more symmetric than those in α -cristobalite [50], and therefore, β -cristobalite exhibits a higher A value than α -cristobalite. Unlike cristobalite, α -quartz contains rings of two different sizes, as determined from

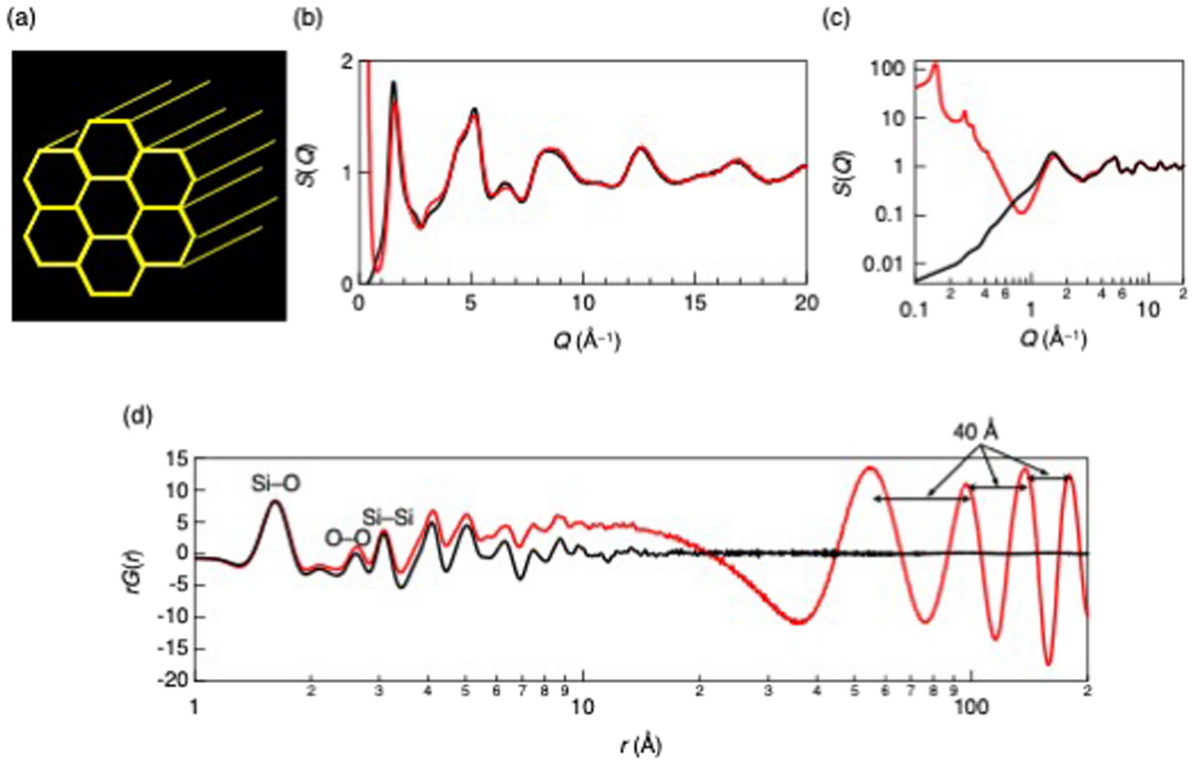


Figure 12. (a) Schematic of the hexagonal pore network of MCM-41. (b) X-ray total structure factors, $S(Q)$, of MCM-41 and SiO_2 glass. (c) Expansion of $S(Q)$ in the low- Q region. (d) r -weighted reduced X-ray pair distributions, $rG(r)$, of MCM-41 and SiO_2 glass. Red curve; MCM-41, black curve; SiO_2 glass.

the primitive criterion [50], i.e. six- and eightfold rings. As a result, α -quartz shows a finite H value of 0.45. In addition, α -quartz exhibits a lower A value than cristobalite, which reasonably reflects the more distorted ring shapes in α -quartz than in cristobalite [50].

Compared with these crystals, the H values of SiO_2 glasses are higher because of the broader ring size distribution of the glasses than of cristobalite and quartz [50]. A decreases in the following order of the glass samples synthesised under different conditions: RT/7.7 GPa, 400 $^\circ\text{C}$ /7.7 GPa, RT/20 GPa, and 1200 $^\circ\text{C}$ /7.7 GPa. This trend is well correlated with the increase in density [50], which is further supported by the fact that the glasses synthesised at RT/20 GPa and 1200 $^\circ\text{C}$ /7.7 GPa exhibit both similar densities [50] and A values. These results support the idea that the densification of SiO_2 glasses induces the distortion of constituent rings, which is already discussed in refs. [13,50].

Note that data the points for the SiO_2 glass cluster around that of coesite in Figure 11. In particular, the similarity in H values between coesite and SiO_2 glasses suggests a similar degree of topological disorder. This similarity may be related to a broad peak observed in the INS data for coesite [50], which is very similar to the feature of the boson peak observed in SiO_2 glasses as mentioned in the previous section.

Although the H and A values of SiO_2 liquid are similar to those of SiO_2 glass, they are higher, indicating that SiO_2 liquid is topologically more disordered than that in the glassy state [50], whereas its rings are overall less distorted.

On the basis of the above discussion, we schematically illustrate two-dimensional corner-sharing networks for different combinations of A and H values [14] in Figure 11. For low A and high H , the network consists of rings with uniform size and symmetric shapes (upper left in Figure 11), which corresponds to β -cristobalite. As H increases, a wide variety of ring sizes appear, whereas the rings generally maintain their symmetric shapes (upper right in Figure 11). In contrast, as A decreases, the rings become distorted, whereas the network still consists of rings with the same size (lower left in Figure 11). When A is low and H is high, the network is composed of distorted rings with different sizes (lower right in Figure 11).

Note that both A and H are insensitive to the distinction between crystalline and non-crystalline states. Both the crystals and glasses can exhibit similar values of A and H , as shown in Figure 11 for SiO_2 glasses and coesite, suggesting that A and H provide a unified framework for evaluating topological features across both crystalline and non-crystalline materials. In addition, the ring entropy H represents a measure of order and disorder different from that inferred from the FSDP. Figure S2 shows the $S(Q)$ in the low- Q region for SiO_2 glasses synthesised at various temperatures and under a pressure of 7.7 GPa. Compared with the $S(Q)$ of the glass synthesised at RT/7.7 GPa, which is almost identical to that of the pristine glass in terms of the position/height of FSDP and density, the sample prepared at 400 °C/7.7 GPa exhibits an FSDP with reduced height and a shift to a higher- Q region. As the temperature further increases, the height of the FSDP is significantly increased, reaching its maximum at 1200 °C/7.7 GPa, indicating that the hot-densified glass synthesised under this condition is the most ordered glass, as discussed above. On the other hand, this hot-densified glass shows the highest H among the glasses synthesised under different temperatures at 7.7 GPa, as shown in Figure 11, suggesting the strong topological disorder of the glass synthesised at 1200 °C/7.7 GPa. These results highlight the conceptual difference between the topological order/disorder proposed by Gupta and that proposed in this section.

9 Extended-range pair distribution functions

As mentioned in the previous sections, the diffraction peaks associated with the formation of intermediate-range order was well understood. The next step in diffraction measurements of disordered materials is to gain a proper understanding of diffraction peaks appearing in the very low Q -region. The measurement of extended-range X-ray pair distribution functions was proposed by Benmore et al. [54], who applied this technique to mesoporous materials. Figure 12(a) shows a schematic of the hexagonal pore network of MCM-41; the pore size of our sample is approximately 40 Å. Figure 12(b) shows the X-ray total structure factor, $S(Q)$, of MCM-41 together with that of SiO_2 glass. The $S(Q)$ of MCM-41 is identical to that of $S(Q)$ except in the low- Q region, suggesting that the atomic-scale structure of MCM-41 is identical to that of SiO_2 glass. Figure 12(c) shows the expansion of $S(Q)$ in the low- Q region, in which Bragg peaks corresponding to the hexagonal structure of MCM-41 are observed. The contribution of Bragg peaks is clearly visible in the r -weighted reduced X-ray pair distribution $rG(r)$ shown in Figure 12(d). Thus, extended-range pair distribution function measurement is a powerful tool for characterising mesoporous materials.

10 Conclusions

In this article, we reviewed the recent advances in the structural analysis of disordered materials by quantum beam diffraction and topological analysis. This progress was made possible by the emergence of advanced synchrotron X-ray and neutron facilities, which provide high quality diffraction data of disordered materials. The progress in computer simulation and the development of topological analysis techniques must be noted as well. We also suggest the importance of extended-range pair distribution function analysis. As indicated by arrows in Figure 6, SiO_2 glass exhibits a very small peak [55] on the low- Q side of FSDP. We confirmed that this peak disappears after hot densification, whereas a small plateau remains after cold densification (see Figure 1B in ref. [56]). Densified amorphous siliceous zeolite has a peak at the same Q position, which is a trace of the zeolite cage [56]. The precise measurement of diffraction from disordered materials over low- Q regions requires low-energy X-rays and neutrons with sufficiently low instrument background. This approach will pave the way toward uncovering order within disorder [57] beyond the intermediate range, which will provide important knowledge in the materials science of disordered materials.

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