Temperature dependence of the first sharp diffraction peak in vitreous silica

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The temperature dependence of the first sharp diffraction peak (FSDP) in the neutron structure factor has been measured from room temperature to 1036 °C. With increasing temperature, the FSDP decreases in amplitude in a normal fashion. This is in contrast to the anomalous temperature dependence of the FSDP that is typically observed in covalent oxide and chalcogenide glasses. The change in height of the FSDP with temperature is the result of a competition between an increase in the amplitude of thermal vibration at higher temperature and a diminished frustration at lower density. The normal behavior of the FSDP in vitreous silica is the consequence of an essentially zero thermal-expansion coefficient so that the effect due to thermal vibrations dominates. A structural rearrangement at elevated temperatures is also indicated.

I. INTRODUCTION AND BACKGROUND

Diffraction patterns of covalently bonded oxide and chalcogenide glasses display a characteristic first sharp diffraction peak (FSDP). This peak appears in the structure factor at low momentum transfer, $Q_1 \approx 1.0$–1.5 Å^{-1}, and has an ostensibly narrow half width when plotted in customary units of $Q = (4\pi \sin\theta)/\lambda$. The position of this peak in reciprocal space suggests that Fourier components of period $\approx 2\pi/Q_1 \approx 4$–$6$ Å are involved. The correlation length inferred from the width of the FSDP, $\approx 2\pi/\Delta Q_1$, is of the order of 15–25 Å—a remarkably long range in a solid that by definition has no long-range order. The FSDP is an unmistakable signature of intermediate-range order (IRO) in a glass. However, the nature of the intermediate-range ordering and of the structural entities that give rise to the FSDP remains an unsolved and controversial question.

The x-ray study of Busse and Nagel[1] established the association of the FSDP with anomalous behavior. They reported an unexpected, reversible *increase* in intensity of the first diffraction peak of glassy As_2Se_3 between 40 K and T_g, the glass transition temperature. The intensity of the second peak (and presumably the subsequent peaks) decreased due to the greater thermal-vibration amplitudes at higher temperatures—as expected. They interpreted the anomalous temperature dependence of the FSDP in terms of the existence of crimped, disordered layers in the glass analogous to those that occur in crystalline As_2Se_3. They postulated a flattening of the corrugations with increasing temperature, enhanced registry between the layers, and an increased FSDP intensity arising from interlayer correlations. This gave support to various models for the FSDP in $GeSe_2$, GeS_2, and other binary chalcogenide glasses based on the existence in the glass of structural elements derived from the corresponding crystal structure.[2]

Subsequently, this interpretation has been questioned on the grounds that the FSDP appears in the structure factor without regard to the dimensionality of the glass or the nature of the corresponding crystal structure.[3,4]

Moreover, when plotted on a common Qr_1 scale, where r_1 is the nearest-neighbor atomic separation, the peak positions all fall near the same value, $Qr_1 \approx 2.5$, and the structure factors have a remarkably similar appearance.

The anomalous temperature dependence of the FSDP has been reported for numerous chalcogenide and oxide glasses including B_2O_3,[5] As_2S_3,[6] As_2Se_3,[1] P_2Se_3,[7] GeS_2,[8] $GeSe_2$,[9] $SiSe_2$,[10] and $Ag_4Ge_3Se_9$.[11] In the case of $GeSe_2$, the FSDP persists into the liquid state, and at 1084 K one observes the same intensity and half width as in the glass at 10 K.[12]

Curiously, for vitreous SiO_2, the prototypical network glass, we have found only one published diffraction study of the temperature dependence of the FSDP.[13] Once again, an increase in intensity of the first peak in the x-ray-diffraction pattern with increasing temperature is reported. However, a recent study of vitreous silica at high temperatures using inelastic neutron scattering has yielded an opposite result.[14] By integrating over the dynamic structure factor $S(Q,E)$ and taking the zeroth energy moment, the authors obtain the static structure factor $S(Q)$. They report a decrease in amplitude of the FSDP with increasing temperature within the errors of the inelastic measurement.

II. ORIGIN OF THE FIRST SHARP DIFFRACTION PEAK

A microscopic description of the origin of the FSDP should be (1) independent of the dimensionality of the glass; (2) capable of being generalized to all glasses of a given class (covalent, ionic, etc.); and (3) useful for estimating the position, half width, and intensity of the FSDP and the dependence of these peak parameters upon temperature and pressure. So far, none of the models in the literature satisfies the criteria enumerated above. Ideas based on the random packing of structural units[15] have been instructive. In this case the FSDP arises from a competition between a rapidly falling form factor for a structural unit and a rapidly rising structure factor for the centers of the units. The dimensions of the structural

units are of the order of $2\pi/Q_1$. However, for any given glass with a dimensionality greater than zero, the choice of the structural unit is not unique. For vitreous silica, a covalent network glass of dimensionality three, the choice is not at all obvious.

Alternatively, the molecular-dynamics (MD) technique has been used to obtain a microscopic picture of the correlations that give rise to the first sharp diffraction peak. The treatment of molten and vitreous $GeSe_2$ (Refs. 16 and 17) is typical for covalently bonded oxide and chalcogenide glasses. The MD calculations were performed with effective interparticle potentials consisting of two-body and three-body covalent interactions. Periodic boundary conditions were used and the long-range Coulomb interactions were treated with Ewald's summation. The resulting partial pair-distribution functions, bond-angle distributions, and static structure factors provide a picture of the short-range and intermediate-range order. MD partial static structure factors indicate that the FSDP arises primarily from cation-cation correlations with a smaller contribution from cation-anion correlations. To determine the range of correlations responsible for the FSDP, the partial pair correlations were truncated at various distances and the effect upon the calculated total static structure factor was noted. In $GeSe_2$, intermediate-range correlations between 4 and 8 Å are responsible for the FSDP.[17] A similar result has been obtained for vitreous silica.[18] For covalently bonded glasses, it may be convenient to visualize the structural entities in the 4–8-Å range that give rise to the FSDP as large rings.[19]

The anomalous temperature dependence of the FSDP comes about in the following way. Upon heating a typical covalent glass, thermal-vibration effects broaden the peaks in the radial distribution function and, hence, reduce the amplitudes of peaks in its Fourier transform, the structure factor. However, most covalent glasses expand upon heating; i.e., they decrease in density. In the MD simulation of a binary covalent glass, a decrease in the density of the system decreases the frustration associated with the ordered arrangement of the local structural elements (SiO_4 tetrahedra in the case of fused silica) into a confined volume. This leads to an increase in the intermediate-range order and an enhancement of the structure in the structure factor. Since the length scale for this ordering is greater than 4 Å, the first peak is affected more than the other peaks in the structure factor. Thus thermal-vibration effects seek to decrease the amplitude of the FSDP while the lowered density tends to increase its amplitude. In most covalently bonded glasses the density effect dominates, resulting in an anomalous temperature dependence of the FSDP.

Vitreous silica presents a test case for this microscopic model since the contributions of temperature and density are largely decoupled. The coefficient of thermal expansion of fused silica is one to two orders of magnitude smaller than that of other binary oxide or chalcogenide glasses. Most solids become less dense with increasing temperature. However, the density of fused silica is essentially unchanged over the temperature range of the present neutron-diffraction measurements: the linear

coefficient of thermal expansion is 0.5×10^{-6} per °C.[20] Therefore thermal-vibration effects are expected to dominate, and the amplitude of the FSDP should decrease with increasing temperature, contrary to what has been reported for all other oxide and chalcogenide glasses— including the published x-ray-diffraction results for fused silica.[13]

In this study we report neutron-diffraction measurements on well-characterized samples of fused silica at temperatures from 25 °C to 1036 °C. The results serve as a critical test of the origin of the FSDP and of possible reversible structural transformations in fused silica at elevated temperatures.

III. EXPERIMENTAL DETAILS AND DEFINITIONS

Time-of-flight, neutron-diffraction measurements were carried out on the Special Environment Powder Diffractometer (SEPD) at the Argonne National Laboratory's Intense Pulsed Neutron Source.[21] Data were collected simultaneously in six groups of time-focused detectors placed at $\pm90°$, $\pm60°$, $-30°$, and $+14.9°$ with respect to the incident beam, and subtending solid angles of 0.086, 0.052, 0.017, and 0.017 sr with respect to the sample. The data were combined by weighting with the incident neutron distribution over the range 0.3 to 3.0 Å. Particular care was taken with respect to resolution ($\Delta Q/Q=0.01-0.03$ at $Q\approx2$ Å^{-1}), normalization of the diffraction data, background subtraction,[22] furnace scattering subtraction, and the corrections for sample attenuation, multiple scattering, and inelasticity. Details of the neutron-diffraction measurements and the data reduction procedure are given in previous publications.[23]

The fused silica sample was in the form of a 9-mm-diam rod with a hydroxyl concentration of 289 μg/g. Details regarding the physical and chemical characteristics of typical fused-silica specimens can be found in footnotes 2 and 15 of a previous publication.[24] The cylindrical rod for the neutron-diffraction study and the flat plates for the x-ray-diffraction study were annealed for several hours at 40–160 °C below the glass transition temperature prior to the measurements [$T_g(SiO_2)=1180$ °C]. Neutron-diffraction data sets were collected at room temperature, 682 and 1036 °C with the sample positioned axially in a tubular vanadium-foil furnace.[25] Measurements of the empty furnace were made at the same temperatures. In addition, diffraction measurements were taken at room temperature on the free-standing rod without the confines of the furnace.

We define the average structure factor for the binary (Si,O) fused-silica system as[26]

$$\langle S(Q)\rangle = \sum_{i,j} c_i^{1/2}c_j^{1/2}\overline{b}_i\overline{b}_j$$
$$\times[S_{i,j}(Q)-\delta_{i,j}+c_i^{1/2}c_j^{1/2}]/\langle\overline{b}\rangle^2 ,$$

where c_i and $\overline{b}_i$ are the concentration and mean coherent scattering length of atom i, and $\langle\overline{b}\rangle=(c_{Si}b_{Si}+c_O\overline{b}_O)$. $S_{i,j}(Q)$ is the partial structure factor relating to the pair

of atom types i,j. The Fourier transform of $\langle S(Q) \rangle$ gives

$$D(r) = \frac{2}{\pi} \int_0^{Q_{max}} Q[\langle S(Q) \rangle - 1] \sin Qr M(Q) dQ ,$$

where $M(Q)$ is the Lorch modification function $\sin x /x$, and where $x = Q\pi/Q_{max}$. The modification function is used to reduce termination effects resulting from the finite upper limit on Q, but it has the effect of broadening the peaks in r space by $\approx 5.4/Q_{max}$.

We define the effective pair correlation function $\langle \rho(r) \rangle$:

$$\langle \rho(r) \rangle = D(r)/4\pi r + \rho_0$$

$$= \sum_{i,j} c_i c_j \bar{b}_i \bar{b}_j \rho_{i,j}(r)/\langle \bar{b} \rangle^2 ,$$

which is a weighted average of the partial pair correlation functions $\rho_{ij}(r)$ of atom types. ρ_0 is the mean number density derived from the bulk density of the sample. Additionally, we define the total pair correlation function $T(r)$

$$T(r) = D(r) + 4\pi r \rho_0 = 4\pi r \langle \rho(r) \rangle ,$$

and the effective radial distribution function $n(r)$

$$n(r) = rT(r) = 4\pi r^2 \langle \rho(r) \rangle .$$

The importance of $n(r)$ lies principally in the fact that the area under a given peak yields the effective coordination number for that given shell. The function $T(r)$ is used in obtaining peak fits since it qualitatively yields a better fit than using $n(r)$.

X-ray-diffraction data were collected with a Rigaku system using a sealed, 1.5-kW Cu tube and a NaI scintillation detector. The sample was in the form of a 1.06-mm-thick, fused-silica plate with a hydroxyl concentration of 335 μg/g. It was contained in an evacuated, clamshell furnace and was surrounded by three, concentric, nickel-foil radiation shields 0.006 mm in thickness. The radiation shields also served to remove Cu $K\beta$ radiation. The platinum-wound furnace maintained a uniform sample temperature that was constant to $\pm 3\,°$C. Data sets were collected at a fixed incident x-ray flux. Room-temperature spectra were obtained before and after each high-temperature run to confirm the constant output of the x-ray source.

IV. EXPERIMENTAL RESULTS

The neutron structure factor $S(Q)$ for vitreous silica as a function of temperature is presented in Fig. 1. The full structure factor is the flux-weighted composite of the data collected in six detector banks. An expanded plot of the 30° bank by itself is given in Fig. 2. These data are typical of what is observed in the 14.9° bank (with better counting statistics) and in the 60° banks (with poorer counting statistics) at the maximum of the FSDP. The position, intensity, and full width at half maximum (FWHM) of the FSDP are listed in Table I for each temperature. Within experimental error, the position in re-
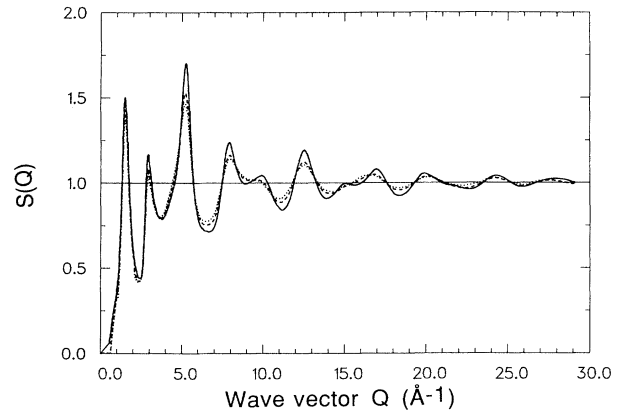

FIG. 1. The neutron structure factor $S(Q)$ of vitreous silica as a function of temperature. ——, 25 °C; – – –, 682 °C; · · · ·, 1036 °C.

ciprocal space and the FWHM do not change between room temperature and 1036 °C. The intensity of the FSDP *decreases* as do all of the other peaks in the structure factor. This is contrary to the anomalous temperature dependence of the intensity of the FSDP reported for all other binary, covalently bonded glasses.

The changes in the structure factor with temperature are small, but they are outside our estimates of experimental error. The differences do not arise from any artifacts of the experimental procedure or the corrections routine. The neutron-diffraction data sets were collected at each temperature in precisely the same way for the sample and the empty furnace in one, continuous, four-day period. Standard vanadium data sets for normalizing against the incident spectrum were collected at the beginning and end of the experiment. The data for each temperature were subsequently analyzed in exactly the same way ensuring that differences between structure factors are meaningful if they exceed statistical error. An expanded $\Delta S(Q)$ difference plot for the 30°-bank data be-
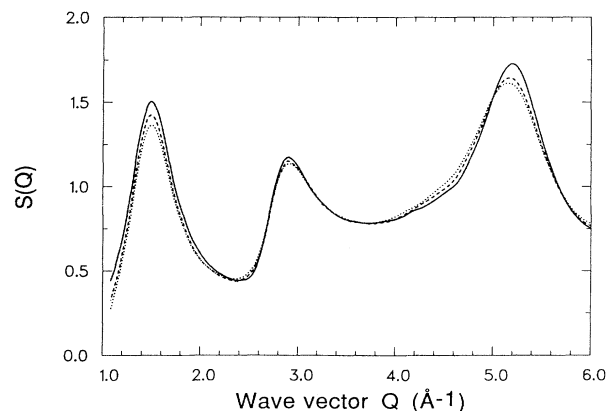

FIG. 2. Expanded plot of $S(Q)$ of vitreous silica for the 30° bank only. ——, 25 °C; – – –, 682 °C; · · · ·, 1036 °C.

TABLE I. Temperature dependence of the first sharp diffraction peak in vitreous silica.

Temperature (°C)	Q (Å^{-1})	$S(Q)$	FWHM (Å^{-1})
25	1.50	1.50	0.66
682	1.49	1.42	0.65
1036	1.49	1.36	0.65

FIG. 4. The total pair-correlation function $T(r)$ of vitreous silica as a function of temperature. ——, 25 °C; — — —, 682 °C; · · · ·, 1036 °C.

tween 1036 and 25 °C is shown in Fig. 3.

Fourier transformations of the $S(Q)$ data in Fig. 1 lead to the real-space, total pair-correlation functions $T(r)$ shown in Fig. 4. The upper limits of the transforms from momentum space were taken at the zero crossings between 28 and 29 Å^{-1}. The experimentally determined areas produced by pairs of atoms can be obtained by integration of Gaussian functions in $T(r)$ fitted to the experimental data in Q. Table II lists the peak-fit parameters where r is the coordination distance, $\langle u^2 \rangle$ is the mean-square displacement, and n is the coordination number for a given peak. [The widths quoted do not include the broadening due to the modification function $M(Q)$.] The parameters for the free-standing fused-silica sample when run outside the furnace are presented for comparison. Recent literature values for a time-of-flight, neutron-diffraction measurement are also given.[27] Systematic errors arising, for example, in the corrections for attenuation and multiple scattering are not expected to be temperature dependent. Absolute values of some of the real-space parameters (such as the coordination number) may not be exact, but relative differences are meaningful when they exceed the statistical error. The $\Delta T(r)$ difference plot for 1035–25 °C is presented in Fig. 5.

V. DISCUSSION

Figure 1 demonstrates that increasing the temperature of vitreous silica from 25 to 1036 °C results in a damping of *all* of the features in the neutron structure factor.
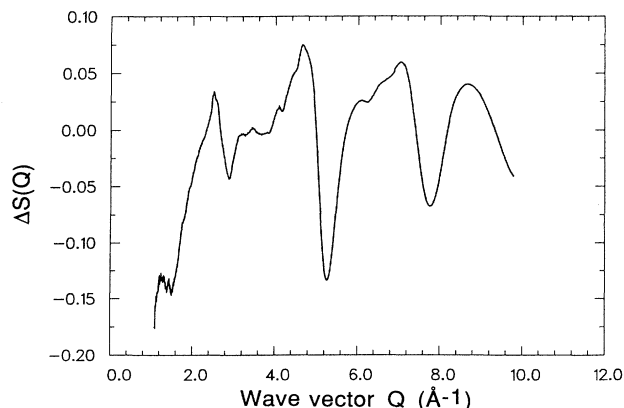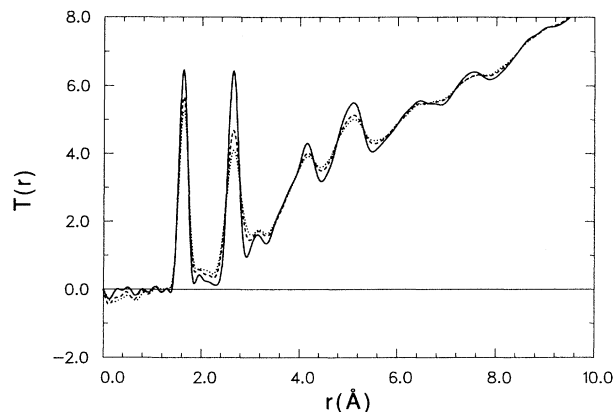
Table I indicates that the intensity of the FSDP decreases by 10%. This decrease in intensity with temperature is not accompanied by any increase in the FWHM or shift in the peak position. The length scale of the ordering giving rise to the FSDP is unchanged. The coherence length reflected in the width of the FSDP is also unaltered at high temperature.

Since the volume of vitreous silica is essentially constant over the temperature range of these measurements, the frustration in the ordered arrangement of the SiO_4 tetrahedra remains nearly the same. The enhancement of the FSDP resulting from diminished frustration with increasing temperature that is present in all other binary covalent oxide and chalcogenide glasses is absent here. The temperature dependence of the FSDP in vitreous silica is dominated by the thermal-vibration effects of its constituent atoms. A quantitative treatment by MD simulation with three-body interactions is in progress.[28]

Table II indicates a small increase in the Si-O and O-O distances and an increase in the mean-square displacements at high temperature as expected. What is not ex-
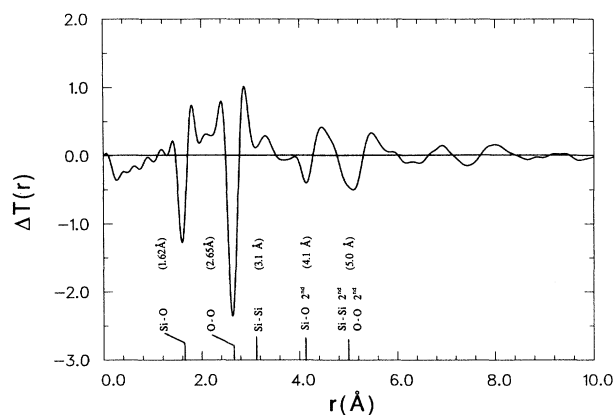

FIG. 3. Expanded difference plot $\Delta S(Q)$ for $S(Q)_{1036\,°C} - S(Q)_{25\,°C}$ for the 30° bank only.

FIG. 5. Difference plot $\Delta T(r)$ for $T(r)_{1036\,°C} - T(r)_{25\,°C}$.

TABLE II. Temperature dependence of the peak parameters for vitreous silica.

Temperature (°C)	r_{Si-O} (Å)	$\langle u^2 \rangle$ (Å^2)	$n_O(Si)$	r_{O-O} (Å)	$\langle u^2 \rangle$ (Å^2)	$n_O(O)$
25	1.62$_6$	0.0022$_7$	3.85	2.65$_0$	0.0072	5.68
682	1.63$_2$	0.0035$_1$	3.67	2.65$_5$	0.0146	5.23
1036	1.63$_7$	0.0045$_2$	3.62	2.65$_3$	0.0181	5.04
25[a]	1.62$_5$	0.0020$_0$	3.93	2.64$_8$	0.0070	5.70
25[b]	1.60$_8$	0.0022	3.85	2.62$_6$	0.0083	5.94

[a]Independent measurement of the free-standing vitreous silica sample outside the furnace.
[b]Reference 27.

pected is the decrease in area of the first and second coordination shells. There are several possible explanations. The fitting to the total pair-correlation function assumes Gaussian peak shapes, i.e., harmonic vibrations. However, the presence of anharmonicity is suggested by the skewing of the high-r tails of the Si-O and O-O coordination peaks at elevated temperature (see Figs. 4 and 5). But this is not supported by the linear temperature dependence of $\langle u^2 \rangle$ of the first coordination shell. The width parameter $\langle u^2 \rangle_{Si-O}$ is measured as 0.0023, 0.035, and 0.0045 at 298 K, 955 K, and 1309 K, respectively. This linear dependence of the mean-square displacement on absolute temperature supports the assumption of harmonic oscillation in vitreous silica to 1036 °C.

The increased intensity at 2.0 and 3.2 Å may arise from a reversible structural change at high temperature. If a simple thermal broadening of the peaks in $T(r)$ were all that were occurring, then the $\Delta S(Q)$ plot would show an oscillatory behavior about zero out of phase with $S(Q)$. The fact that a consistent drop below zero is observed at low Q implies a reversible structural rearrangement of the SiO$_4$ tetrahedra at high temperature (see Fig. 3). Such a rearrangement must be consistent, however, with the nearly zero change in specific volume.
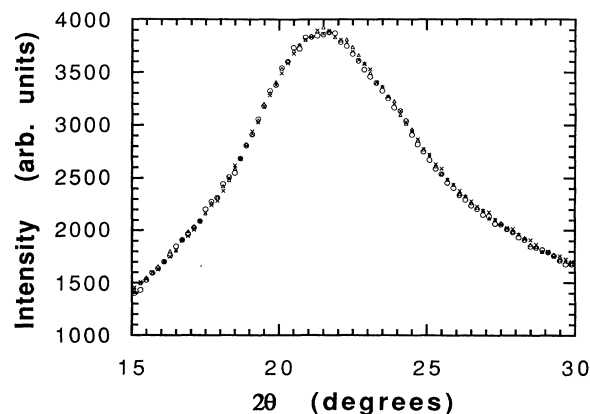
Finally, the decrease in amplitude of the FSDP at

FIG. 6. X-ray-diffraction intensity of vitreous silica as a function of temperature. Circles, 25 °C; triangles, 646 °C; crosses, 1038 °C.

elevated temperature observed in the present neutron-diffraction study stands in contrast to the large increase in amplitude reported in the only published x-ray study.[13] Our x-ray results are presented in Fig. 6. The data were collected in 0.02° increments in 2θ and rebinned in groups of 10. The intensity of the FSDP at 646 °C is unchanged from that observed at 25 °C. This temperature matches the 650 °C temperature of the Russian study and is below the 682 °C temperature of the neutron data presented here. The results of an x-ray-diffraction measurement at 1038 °C are included in the plot of Fig. 6. The x-ray data of Fig. 6 may be compared with the neutron data of Fig. 2. There is no change in the intensity of the FSDP between room temperature and 1038 °C when measured with x rays. The standard deviation over a 0.2° interval centered at the peak of the FSDP is typically ±70 counts out of 4000 for a given run. The standard deviation between the 646 °C run, the 1038 °C run, and three room-temperature runs (bracketing the high-temperature runs) is only ±43 counts.

A possible explanation for observing a decrease in amplitude of the FSDP when diffracting neutrons and no change in amplitude when diffracting x rays is as follows: Neutrons and x rays do not probe the same correlations with the same sensitivity. While cation-cation correlations contribute most to the intensity of the FSDP, there are contributions from the other partial correlations involving the anions. Neutrons are most sensitive to the oxygen correlations since $\bar{b}_O > \bar{b}_{Si}$, while for x rays, $f_{Si} > f_O$. Since oxygen is lighter than silicon, its amplitude of vibration will increase faster than that of silicon as the temperature is raised. In fused silica, only a small reduction in frustration of the ordering can occur due to the change in dihedral angle at elevated temperature. There is no contribution to the FSDP from a drop in density. The thermal-vibration effect of the Si-O partial pair correlation is greater than the small positive contribution of the Si-Si partial for diffraction of neutrons. For x rays, the situation is different. The x-ray probe largely misses the contributions of the oxygen partial pair correlations to the structure factor. The reduction in frustration associated with the structural rearrangement at elevated temperature is exactly offset by the comparatively small increase in vibration of silicon which the x rays sample very well. The final picture is one in which the neutron measurement yields a normal decrease in the intensity of the FSDP with increasing temperature while the x-ray mea-

surement shows no change. The minimal thermal-expansion coefficient of fused silica and the reversible structural rearrangement at high temperature lead to this unusual result. Confirmation of this picture awaits the results of the detailed MD simulation which is in progress.[28]

VI. SUMMARY

The temperature dependence of the first sharp diffraction peak in the neutron structure factor has been measured from room temperature to 1036 °C. With increasing temperature, the FSDP decreases in amplitude in a normal fashion. This is in contrast to the anomalous temperature dependence of the FSDP that is typically observed in covalent oxide and chalcogenide glasses. The height of the FSDP at elevated temperatures is the result of a competition between an intensity decrease from thermal vibrations and an intensity increase arising from diminished frustration in the ordering. The normal behavior of the FSDP in vitreous silica is the consequence of an essentially zero thermal-expansion coefficient so that the thermal vibrations dominate. A structural rearrangement at elevated temperatures is also indicated.

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