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Dynamical properties of vitreous silica around the first sharp diffraction peak

Mitsutaka Nakamura,* Masatoshi Arai, Yasuhiro Inamura, and Toshiya Otomo
Neutron Science Laboratory, Institute of Materials Structure Science, KEK, 1-1 Oho, Tsukuba 305-0801, Japan

S. M. Bennington
ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 0QX, United Kingdom

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Inelastic neutron scattering measurements on vitreous silica have revealed that the contribution from the acoustic-like mode along the position of the first sharp diffraction peak (FSDP) is prominent below the boson peak energy and at low temperatures. Strong coupling between this acoustic-like mode at the FSDP position and the boson peak has also been confirmed, which implies the existence of Ioffe-Regel limits in glasses. We propose that the origin of the excitations along the FSDP position can be attributed to the harmonic vibrations at the surface of void spaces in glasses. We also consider that the FSDP in glasses arises from a quasi-periodicity, which is caused by the continuity of void spaces, corresponding to the lattice spacing of crystalline counterparts.

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I. INTRODUCTION

Disordered materials have provided a large number of scientific problems, and the universality of their physical properties has fascinated many researchers. Most of the problems remain unsolved, although enormous work has been done on both the theoretical and experimental aspects. Particular efforts have been directed to the energy range below 10 meV in the Raman or inelastic neutron scattering spectra. In this energy range, the Raman and inelastic neutron scattering data are dominated by a strong broad band. This peak, called the boson peak, is not found in crystals. The low-energy broad band in glasses is expected to reflect the cooperative motions of atoms over the correlation length in the nanometer range, and to be related to anomalous thermodynamic properties of glasses such as the thermal conductivity behavior at low temperatures.[1] The existence of low-energy excitation in glasses of different chemical compositions, and the universality of their spectra, would lead to the concept that the nature of anomalous excitation is determined by the more general peculiarities of disordered structure. It has been considered that the inherent inhomogeneity in glasses may be related to the origin of the boson peak.

There is also substantial interest in the character of excitations contributing to the boson peak. In the long-wavelength limit, the phonon wave vector q becomes well defined at small values because glasses behave like elastic continua. Actually, Brillouin scattering measurement has revealed that both transverse acoustic (TA) and longitudinal acoustic (LA) phonons exist in glasses.[2] However, at a large wave vector q (small wavelength λ), glasses are considered to be inhomogeneous media due to the lack of periodicity, where q becomes ill defined and the distinction between the TA and LA modes is obscured. The crossover value of q_{co} or λ_{co} plays an important role in the concept of Ioffe-Regel (IR) criteria.[3] It is considered that the plane wave ceases to propagate in the medium when the wavelength λ is smaller than λ_{co}, which should define the medium-range structural order (MRO). Many recent works using inelastic x-ray scattering (IXS) measurement have been dedicated to the problem of whether the IR crossover actually exists in glasses.[4] The neutron scattering technique is useless for investigating the acoustic excitation around $Q \approx 0$ Å^{-1} in the boson peak region, due to the kinematic restriction of neutrons, which are massive particles.[5]

The MRO in glasses can be attributed to the existence of the first sharp diffraction peak (FSDP) observed in the x-ray and/or neutron structure factor at small scattering vector Q_1. The FSDP may be related to a distance R in real space by the expression, $2\pi/Q_1 = R$, although this simple idea has been criticized by the opinion that the FSDP should arise from the longest period, most slowly decaying, real space Fourier component.[6] Several models have been proposed to account for the origin of the FSDP. The two-dimensional layers of the corresponding crystals was introduced to interpret the FSDP of chalcogenide glasses.[7] However, this idea has been also refuted,[8] because the v-SiO$_2$ and other three dimensional glass systems show distinct FSDP, which has no layered structure. Elliott[8] successfully explained the origin of the FSDP for tetrahedrally coordinated glass systems from correlations between cation-centered clusters and interstitial void spaces. Notwithstanding this criticism, the simple interpretation of FSDP for v-SiO$_2$ in terms of quasi-Bragg planes was recently revived.[9] In this way, the origin of the FSDP has as yet received no satisfactory microscopic explanation. On the assumption that the boson peak originates in the length scale associated with the medium-range order, the boson peak might be related to the FSDP through the structural characteristic length. In fact, attempts to correlate the width of the FSDP with the boson peak have been fairly successful.[10]

It is well known that inelastic neutron scattering (INS) is a very powerful tool for the investigation of excitations in both disordered and ordered materials. In particular, Price and Carpenter reported the dynamic structure factor $[S(Q,E)]$ of vitreous silica (v-SiO$_2$) over a wide momentum-energy space using the pulsed neutron scattering technique.[11] A time-of-flight chopper spectrometer with a pulsed neutron source has provided a new stage for the study of disordered materials with the help of a great improvement in neutron flux and energy resolution.[12] This paper describes our study of the dynamical properties of v-SiO$_2$ from the $S(Q,E)$ maps obtained by time-of-flight INS measurement with the most intense neutron flux in the world. The purpose of this study was to understand the mechanism of the correlated phenomena of glasses as mentioned above. v-SiO$_2$ is a prototypical strong glass former, and the boson peak is clearly observed around 5 meV. The temperature dependences of $S(Q,E)$ at selected energies were precisely investigated. Because both Si and O have negligible incoherent scattering cross sections, we considered only the coherent scattering. In particular, we focused on the dynamical properties in the low E and low Q regions; that is, around the boson peak energy and the FSDP region.

II. EXPERIMENTAL PROCEDURES

The experiments were performed on the MARI spectrometer at the ISIS spallation neutron source in the Rutherford Appleton Laboratory. MARI is a direct geometry chopper spectrometer designed for wide momentum-energy space measurements with high resolution and high flux. The selected incident energy was 25 meV for detailed investigation of $S(Q,E)$ in both the low E and low Q regions with an energy resolution of 0.7 meV. The beam size was 50×50 mm^2. The collected data were normalized by the mass of samples in the region irradiated by the incident neutron beam. The detector calibration was determined by the measurement of a vanadium sample. The sample of v-SiO$_2$ was cut into a hollow cylindrical form in order to reduce multiple scattering and anisotropy of self shielding. Its inner diameter was 25 mm, its thickness was 3 mm, and its length was 120 mm. The environmental temperature was controlled in the range from 20 to 300 K using a top-loading closed-cycle refrigerator.

In Secs. III B and III C, we compare the elastic scattering of v-SiO$_2$ with those of other types of SiO$_2$ in order to elucidate the origin of the FSDP. The additional SiO$_2$ samples were polycrystalline α-cristobalite (17.3 g), polycrystalline α-quartz (25.3 g), and permanently densified SiO$_2$ glass (5.3 g). The prepared polycrystalline α-cristobalite and permanently densified SiO$_2$ glass (d-SiO$_2$) were bulk solids, while the polycrystalline α-quartz was in powder form. The d-SiO$_2$ sample was prepared using the method described by Kitamura *et al.*[13] Measurements of each sample were performed at room temperature, and we narrowed the beam size depending on the sample size.

III. RESULTS AND DISCUSSION

A. Peculiar excitation around the FSDP position

The $S(Q,E)$ map is extremely informative for the investigation of vibrational properties in disordered materials, since from it we can easily extract a series of constant Q or E slices and also the integrations over the arbitrary Q or E ranges. The Q dependences of $S(Q,E)$ for $3 \leqslant E \leqslant 6$ meV at 20 and 300 K are shown in Fig. 1. The integration width was 0.8 meV for each spectrum, which is larger than the energy resolution ($\Delta E = 0.7$ meV). A conspicuous hump around 1.5 Å^{-1} is observed in the lower energy region, which becomes prominent at lower temperatures. It is also obvious that the intensity around 1.5 Å^{-1} is strongly damped beyond the boson peak energy ($E \approx 5$ meV). It should be noticed that $Q \approx 1.5$ Å^{-1} corresponds to the position of the FSDP observed in the static structure factor $[S(Q)]$ of v-SiO$_2$[14]. In order to clarify the peculiar excitation around 1.5 Å^{-1}, we compared the Q dependence of $S(Q,E)$ sliced at 5 meV between 20 and 300 K, as shown in Fig. 2(a), and similarly at 10 meV as shown in Fig. 2(b). For each spectrum in Fig. 2 the constant background was subtracted, and the intensities at 300 K were scaled to the third peak intensity at 20 K. Figure 2 clearly shows that a certain low-energy excitation around the FSDP position appears at low temperatures, or rather except for a feature around 1.5 Å^{-1} there is little difference in the shape of $S(Q,E)$ between 20 and 300 K.

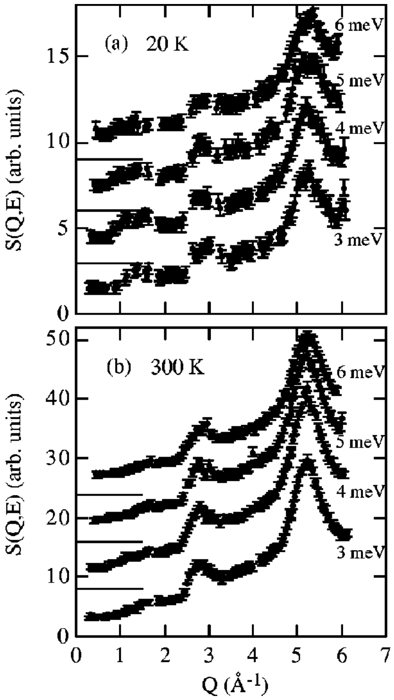

FIG. 1. Constant E slices of $S(Q,E)$ in the range of 3 meV $\leqslant E \leqslant 6$ meV (a) at 20 K and (b) at 300 K. The integration width is 0.8 meV for each spectrum. The base lines are shifted vertically. The dynamical behaviors around 1.5 Å^{-1} shown by vertical arrows were intensively investigated in this study.

To examine the properties of the peculiar hump around 1.5 Å^{-1}, we display magnified plots of $S(Q,E)$ at 20 K for both $E = 3$ and 4 meV in Fig. 3. The solid lines are fitted curves by two Gaussian functions and a linear function as a base line. We can see that the single-like peak profile at 3 meV is greatly split at 4 meV. The estimated peak centers for the $E = 3$ meV spectrum are 1.263 and 1.468 Å^{-1}. In the case of $E = 4$ meV, two peak centers are also determined at $Q = 1.125$ and 1.601 Å^{-1}. The data points around 1.7 Å^{-1} are useless because of the gap between neighboring detector banks. Hence we paid attention to the peak shift of the lower Q peak profile. The separations from 1.364 Å^{-1} (ΔQ) are

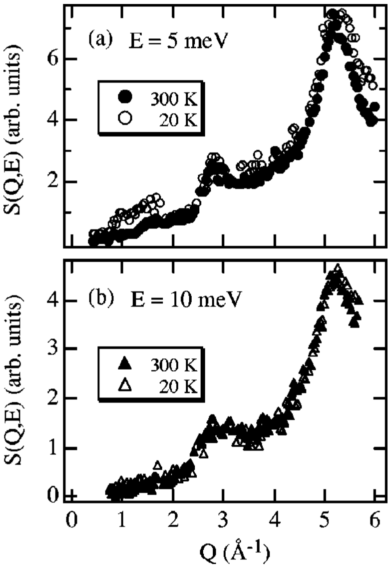

FIG. 2. Comparison of the Q dependence of $S(Q,E)$ between 20 K and 300 K sliced at (a) $E=5$ meV (at the boson peak energy), and (b) $E=10$ meV (above the boson peak energy). For each spectrum the constant background is subtracted. The intensities at 300 K are scaled to the third peak at 20 K.

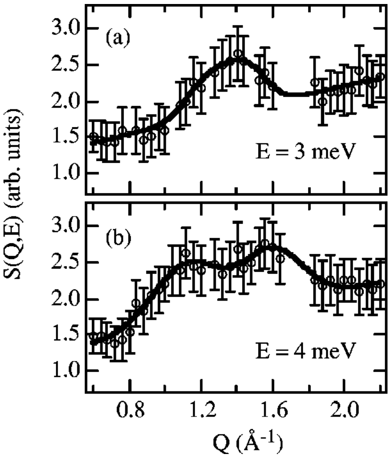

FIG. 3. Magnified plots of $S(Q,E)$ at 20 K for (a) $E=3$ meV and (b) $E=4$ meV. The solid lines are the results of fits by two Gaussian functions and a linear function.

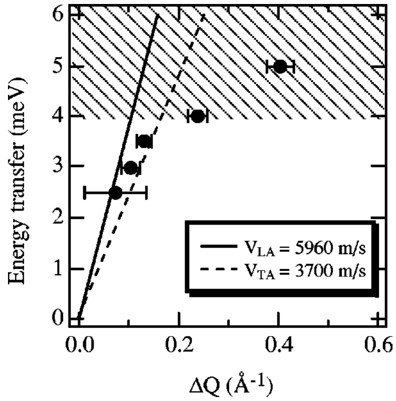

FIG. 4. Energy dependence of the peak separation from 1.364 Å^{-1}. The value of 1.364 Å^{-1} is the midpoint of the double peaks estimated from both the $E=3$ and 4 meV data shown in Fig. 3. The linear relations expected from both the longitudinal and transverse sound velocities at 300 K are also displayed. The shaded area corresponds to the boson peak region.

plotted as a function of energy transfer in Fig. 4. The value of 1.364 Å^{-1} is the midpoint of the two peak centers estimated from both the $E=3$ and 4 meV spectra. Figure 4 also includes the linear relations derived from the longitudinal and transverse sound velocities at 300 K; that is, $V_{LA}=5960$ m/s and $V_{TA}=3700$ m/s.[15] It is clear that the peak shift behavior strongly deviates from a linear relation near the boson peak energy. The properties of the peculiar excitation around the FSDP position are in good agreement with the IR condition,[3] since the boson peak is considered to be a strongly localized mode.[16] Correspondingly, the origin of the hump around the FSDP position is considered to be an acousticlike extended mode. The temperature dependence of the hump characteristics can be explained by that of the mean free path of the acoustic-like mode in v-SiO$_2$. In this paper, we refer to this mode as the FSDP mode. Discernible excitations along the FSDP position were previously also reported in other glass systems, such as B$_2$O$_3$,[17] polybutadiene,[18] and Se.[19] It is considered that there are two different types of contributions to the low-energy region; namely, in-phase modes and random-phase modes. In random-phase modes, the atoms have no phase correlation with neighboring atoms, and so random-phase modes have a localized nature. In-phase modes are identified with sound waves, whose intensities are approximately proportional to $Q^2S(Q,0)$.[20] With regard to v-SiO$_2$, Buchenau *et al.*[21] pointed out that a constant E profile of v-SiO$_2$ is inconsistent with the formula of $Q^2S(Q,0)$. Hence it seems reasonable to suppose that the FSDP mode of v-SiO$_2$ found in our measurements is not a pure sound wave, but could have a propagating character because of its dispersive feature.

B. Microscopic origin of the FSDP mode observed in $S(Q,E)$

We examined the overall vibrational dynamics of v-SiO$_2$ over a wide momentum-energy space. v-SiO$_2$ shows both a dispersive mode up to 55 meV and a nondispersive mode around the boson peak energy.[16] In Fig. 5(a), a series of dispersive modes and the FSDP mode are plotted together. The dashed curve is the dispersion function $E=A|\sin(qc/2)|$, where A is a scale factor, q is the wave vector in one zone, and c is the periodicity of the zone. In addition to these modes, the excitation behaviors around $Q\approx 0$ Å^{-1} derived from the IXS measurement[22] are also included in Fig. 5(a). Pilla *et al.* demonstrated that the propagating acoustic mode extends beyond the boson peak energy and excluded the possibility that the boson peak marks the transition from propagating to localized dynamics in glasses.[22] However, our experimental results clearly indicate that the FSDP mode strongly couples with the boson peak, which suggest the IR crossover limit in glasses. We consider that the calculated result of dynamic structure factor in low Q

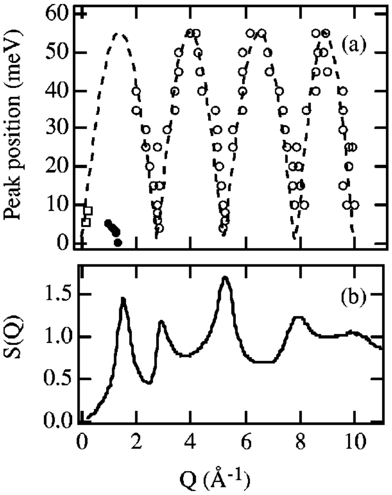

FIG. 5. (a) Peak positions of the FSDP mode plotted against the momentum transfer, shown by the closed circles. The peak positions of the dispersive mode reported in Ref. 16 are also plotted, shown by the open circles. The dashed line represents an estimated dispersion relation fitted by $E = A|\sin(qc/2)|$, where the contents of the parameters are given in the text. The open squares show the data obtained from IXS measurements reported in Ref. 22. (b) Static structure factor of v-SiO$_2$.

region by Nakayama[23] can settle the above discrepancy. The calculated result clearly shows two bands in Q-E space. The higher band is assigned to the dispersive mode above the boson peak energy. On the other hand, the lower band is almost independent of Q at boson peak energy, but the propagating excitation appears at small Q region. This picture can explain both the strong coupling of the FSDP mode with the boson peak and the acoustic behavior at small Q region.

The static structure factor $[S(Q)]$ of v-SiO$_2$ is also presented in Fig. 5(b). Our previous works also reported that the periodicity of the dispersive mode is equivalent to the separation between the neighboring peak positions of $S(Q)$ *except for the FSDP*, which suggests the existence of quasi-Brillouin zones consisting of SiO$_4$ tetrahedral units.[16,24] Investigations of vibrational properties extending to the inelastic region have emphasized the unique characteristics of FSDP. In this sense, understanding of the origin of the FSDP mode should lead to that of the FSDP in glasses, which will be discussed in Sec. III C.

Previously, from a detailed analysis of averaged mean-square displacement for various types of SiO$_2$, we reported that the excess vibrational density of states (VDOS) in v-SiO$_2$ is caused by uniaxially large atomic motions along the buckling direction, whose dynamics are represented by the small barrier height double-well potential.[25] The vibrational character associated with the buckling process should be expressed by a random-phase mode. On the other hand, the in-plane atomic motions perpendicular to the buckling direction were found to be unaffected by densification, and the vibrational amplitudes of the glassy state of SiO$_2$ in the perpendicular direction were found to be almost equivalent to those of crystalline counterparts.[25] We consider that the in-plane atomic motions in v-SiO$_2$ contribute to the harmonic excitations and induce the FSDP mode. If we accept the two-dimensional character of harmonic excitations in glasses, it would be easy to understand the temperature dependence of thermal conductivity at low temperatures, $\kappa \propto T^2$, which is a universal phenomenon observed in all types of glasses.[1] However, there is room for further investigation and consideration on this point.

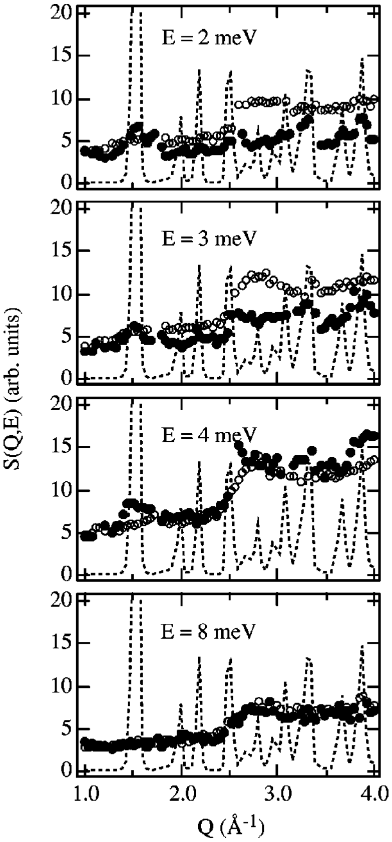

FIG. 6. Q dependences of $S(Q,E)$ at 2, 3, 4, and 8 meV plotted for v-SiO$_2$ (open circles) and polycrystalline α-cristobalite (closed circles). The $S(Q,0)$ of polycrystalline α-cristobalite is also included (dotted lines) for each frame. The integration width was 0.8 meV for constant E slices and 1 meV for elastic scattering. The measurements were performed at room temperature for all spectra.

Next, we discuss the reason why the harmonic excitation appears at the FSDP position. The vibrational properties in both the low Q and low E regions of crystalline SiO$_2$ are compared with those of v-SiO$_2$. Similarities and/or differences of properties between glassy states and crystalline counterparts have greatly contributed to understanding of the anomalous characteristics of glasses. We present a series of constant E slices of $S(Q,E)$ for crystalline SiO$_2$ to elucidate the peculiar features of v-SiO$_2$ in the low-energy region. The constant E slices of $S(Q,E)$ at selected energies for v-SiO$_2$ and polycrystalline α-cristobalite are shown in Fig. 6. The $S(Q,0)$ for α-cristobalite is also included in this figure. Each integration width was 0.8 meV for constant E slices and 1 meV for elastic scattering. Measurements were performed at room temperature. In the constant E slices of α-cristobalite, many spiky features of Bragg peak tails are observed. On the other hand, v-SiO$_2$ shows no spiky features in the low-

TABLE I. Mass densities of prepared SiO_2 samples.

Sample	ρ (g/cm^3)	Type
v-SiO_2	2.20	Glass
α-cristobalite	2.33	Polycrystal
d-SiO_2	2.63	Glass
α-quartz	2.65	Polycrystal

energy region due to the lack of long-range structural order. The spiky features observed in α-cristobalite become blurred with increasing energy. The $S(Q,E)$ of α-cristobalite is almost the same with that of v-SiO_2 at 8 meV, as we reported previously.[24] Looking more carefully into the constant E profiles of α-cristobalite around 1.5 Å^{-1}, we can recognize that the E dependence of this peak quite closely resembles the behavior of the FSDP mode in v-SiO_2 at low temperatures. These acoustic phonon like properties of the FSDP mode are consistent with the numerical analysis by Taraskin and Elliott,[26] who exhibited a dispersion relation for v-SiO_2 using the molecular dynamics calculation. According to their numerical study, the flattening of the TA branch occurs at the length scale defined by the position of the FSDP, $R_1 = 2\pi/1.5$ Å^{-1} ≈ 4.2 Å, and the pseudoperiod of R_1 coincides with the height of oxygen decorated $SiSi_4$ tetrahedra, which is correspondent to a certain lattice spacing in crystalline SiO_2.[26]

C. Microscopic origin of the FSDP observed in $S(Q)$

Judging from the above arguments, the FSDP observed in $S(Q)$ of v-SiO_2 is considered to be a trace of periodicity in crystalline SiO_2. In this picture, the FSDP is considered to be an isolated peak in reciprocal space Q_1 caused by the diffraction from a quasi-periodic array of atoms in real space. This reminds us of the interpretation of the FSDP introduced by Gaskell and Wallis.[9] They proposed that the FSDP may originate in the quasi-Bragg planes of the corresponding crystalline phase.

In order to confirm the validity of the above idea, we investigated the density dependence of the first peak observed in elastic scattering measurement of both glassy SiO_2 and polycrystalline SiO_2. The mass densities of the prepared SiO_2 samples are listed in Table I. Plots of $S(Q,0)$ for the different forms of SiO_2 are shown in Fig. 7. For each spectrum, the integration width was 1 meV and the measurement was carried out at room temperature. It is well known that the position of the most intense diffraction peak of polycrystalline α-cristobalite, which is assigned to the $\{101\}$ plane, is the same as the FSDP of v-SiO_2,[27] as shown in Fig. 7(a). An interesting finding is given in Fig. 7(b), which indicates that the most intense peak position of polycrystalline α-quartz at $Q \approx 1.8$ Å^{-1} assigned to the $\{10\overline{1}\}$ plane is coincident with the position of the FSDP of d-SiO_2. This correspondence can be directly explained by the mass density of the sample, where the ratio Q_1/ρ has a constant value. Inamura *et al.* elucidated that the appearance of the FSDP must have a close relationship to the void spaces in a glass network from the relation between Q_1 and ρ.[28] The successive void spaces could form a quasi-periodicity corresponding to the lattice spacing for the first peak of crystalline SiO_2, where the mass densities of glassy and crystalline SiO_2 are almost the same.

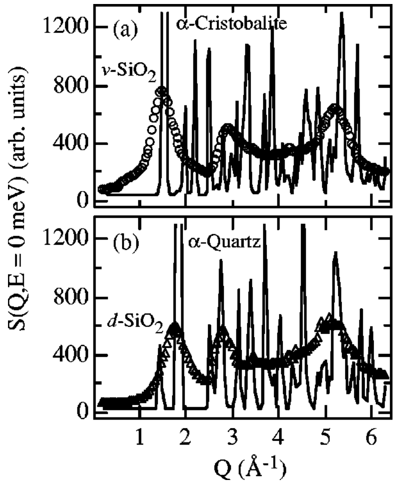

FIG. 7. Elastic part of the dynamical structure factor $S(Q,0)$ for four types of SiO_2: (a) v-SiO_2 and polycrystalline α-cristobalite, and (b) d-SiO_2 and polycrystalline α-quartz. The integration width was 1 meV. The measurements were performed at room temperature for all spectra.

As for the origin of the FSDP, Elliott proposed a plausible model that the FSDP is a prepeak in the concentration-concentration structure factor due to the chemical ordering of interstitial voids around the cation-centered clusters in the structure.[8] In particular, this model beautifully explains why the FSDP intensity is drastically changed by packing the Li or Na atoms into the void spaces of v-SiO_2.[29] However, we have to interpret the acoustic behavior of the FSDP mode and the density dependence of the first peak of both glassy and crystalline SiO_2. Hence we regard the height of the $SiSi_4$ tetrahedra as the characteristic length of glassy SiO_2, which is correspondent to a certain lattice spacing of crystalline counterpart, and simultaneously the average distance of void spaces, instead of nearest-neighbor Si-Si separation or Si-void separation.[8] It should be noted that this modification does not invalidate the Elliott model.

We consider that the void spaces in the glass network may be a key to understanding the anomalous low-energy properties of glasses.[28] We propose a scenario for the relationship between the atomic motions and low-energy dynamics in glasses as follows: the harmonic vibrations occurring at the surface of void spaces and the correlation of atomic motions between neighboring surfaces produce the FSDP mode, while on the other hand, the flexible atomic motions toward void spaces induce the excess VDOS in the low-energy region, which are sensitive to the density[25].

IV. CONCLUSION

In conclusion, we have performed an inelastic neutron scattering measurement of v-SiO_2 and found that peculiar excitation around the FSDP position (FSDP mode) appears below the boson peak energy and at low temperatures. The intensity of the FSDP mode is strongly damped beyond the boson peak energy. In addition, the peak shift behavior of the FSDP mode strongly deviates from a linear relation near the boson peak energy. These results suggest that the Ioffe-Regel condition is applicable to the FSDP mode.

Concerning the FSDP observed in $S(Q)$, we found that the position of the FSDP in glassy SiO_2 coincides with that of the most intense first peak in the crystalline counterpart whose mass density is almost equivalent to that of the glassy SiO_2. We consider that the FSDP of glass is characterized by successive void spaces that induce a quasiperiodicity corresponding to the lattice spacing in crystalline counterparts.

Finally, we have presented a scenario for the coexistence of strongly localized modes and harmonic extended modes in terms of void spaces associated with anisotropic atomic motions. The atomic motions at the surface of void spaces correlate with those at the adjacent surface, and consequently produce harmonic excitations, which may be the origin of the FSDP mode. We should note that the extended range of harmonic excitation is restricted by the finite correlation length. In contrast, the atomic motions toward void spaces are induced by buckling or rocking processes, which produce strongly localized modes in glasses.

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[*] Present address: Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan.

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