Energy-Dispersive X-Ray Diffraction(EDXD) Technique For Structural Studies Of Disordered Materials. Application To SiO_2 , SrP_2O_6, $Pd_{80}Si_{20}$, $Fe_{40}Ni_{40}P_{14}B_6$ Glasses And Liquid Hg

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(Received May 10, 1996; final form July 6, 1996)

ABSTRACT

Structure factors in the wave vector Q region larger than 200 nm^{-1} and corresponding radial distribution functions with improved resolution for SiO_2 and SrP_2O_6 oxide glasses, $Pd_{80}Si_{20}$ and $Fe_{40}Ni_{40}P_{14}B_6$ metallic glasses, and liquid Hg have been obtained by the energy dispersive X-ray diffraction (EDXD) method. Structure data obtained by the present study were compared with the previous wide-angle diffraction experiments of the same disordered materials and the reliability and usefulness of EDXD method were demonstrated.

1. INTRODUCTION

The X-ray diffraction method has been widely used for investigating the atomic scale structure of disordered materials. With the commonly employed wide-angle X-ray diffraction, the X-ray photons scattered from the sample are measured as a function of scattering angle 2θ between the incident and diffracted monochromatic X-rays. By applying proper correction procedures, the coherently scattered X-ray photons are extracted from the measured diffraction spectrum and then reduced to the so-called structure factor $S(Q)$, which actually contains the experimentally accessible information on the atomic scale structure of the material. The details of this experimental approach are well described /1,2/. The reduced atomic distribution function, $G(r)=4\pi r[\rho(r)-\rho_0(r)]$, is readily calculated from the structure factor $S(Q)$ by applying a Fourier transformation,

$$G(r) = \frac{2}{\pi}\int_0^{Q_{max}} Q[S(Q) - 1]\sin(Qr)dQ \qquad (1)$$

where r is the real space distance, $\rho(r)$ and ρ_0 are the number density function and the average atomic number density of the material, respectively, and Q is the wave (scattering) vector defined by the relation $Q = 4\pi\sin\theta/\lambda = 4\pi E\sin\theta/hc$. Here λ and E are the wavelength and the energy of the radiation used, h is the Planck's constant and c the velocity of light. This function and a number of other atomic distribution functions of the pair atomic distribution function $g(r) = \rho(r)/\rho_0$ and the radial distribution function $RDF = 4\pi r^2\rho(r)$, give the probability of finding an atom at a distance r from the original atom by reflecting the various correlations between the positions of the atomic species. Since the atomic distribution functions are associated by a Fourier transformation of the measured quantity of $S(Q)$, it is of crucial importance to obtain the $S(Q)$ function as a region of Q values as extended as possible in order that all atomic correlations in the disordered material are clearly revealed. However, the accessible value of Q is limited by the wavelength (energy) of the monochromatic radiation used since the diffraction angle 2θ cannot exceed 140° by the usual wide-angle diffraction apparatus. Thus, when Mo *Kα* radiation is used, the maximum Q value possible to reach is approximately 150 nm^{-1}. As a result, the atomic distribution functions with resolution no better than 0.02 nm (estimated by $\delta r = \pi/Q_{max}$) could be obtained. If, however, another experimental approach, the so-called "Energy-Dispersive X-ray Diffraction" (EDXD) method is employed, the structure factor over an extended region of Q values can be obtained and the resolution of the corresponding atomic distribution functions is expected to be improved. With the EDXD method the diffraction spectrum is measured as a function of energy E of the X-ray photons scattered at a fixed diffraction angle. Therefore, the sample is irradiated with X-rays of continuous energy (wavelength) spectrum and an energy-sensitive detector coupled with a multi-channel pulse-height analyzer is used to register and store the scattered X-ray photons of different energies. The details of the specific EDXD data correction procedures have been well worked out and can be found in a number of reports /1,3-6/. The advantages for the EDXD analysis on disordered materials are as follows: the experimental time for data collection is shortened and the statistical accuracy of the data is strongly improved since the intensity of the polychromatic radiation produced by a given source is always higher than that of the monochromatic radiation, and fluctuations in X-ray source do not spoil the experimental data quality since all diffracted photons of different energies are recorded simultaneously. The mechanical construction of the experimental set-up is simplified since the scanning of Q space is done electronically by the detector system and this allows us to carry out investigations under non-ambient conditions (high/low temperatures, pressures etc.). The use of higher energy photons up to 50 keV makes it possible to obtain the diffraction spectrum and consequently structure factor over a region Q values extended above 200 nm^{-1}. Several studies have demonstrated these advantages by obtaining structure data for liquids and solid disordered materials by the EDXD method /3,7-11/; this method, however, is still not as widely applied as it could be. It is the purpose of the present paper to promote a wider application of the EDXD method by one more illustration of its potential for obtaining good-quality structure data for a variety of disordered materials.

2. EXPERIMENTALS

2.1 EDXD set-up

The EDXD experiments were carried out on a laboratory set-up consisting of a high-power constant potential generator, a conventional X-ray tube with a W anode and an automated θ–θ vertical type goniometer device. The X-ray source was usually operated at 50 kV and 30 mA. Under these operating conditions the continuous energy spectrum of X-rays delivered by the source extended up to 50 keV. Since sharp W *L* fluorescent lines appeared at energies of approximately 10 keV and the intensity of X-ray photons of energy higher than 40 keV was very low, only the energy interval from about 10 keV to 40 keV was empirically considered in the present studies. The diffracted X-ray photons were detected by a portable Ge solid-state detector firmly attached to one of the arms of the goniometer. The X-ray photons registered by the solid state detector were processed by a multi-channel pulse-height analyzer and stored in a personal computer. A more detailed description of the experimental EDXD set-up can be found in ref. /12/.

2.2 EDXD data collection and processing

The present EDXD experiments on SiO_2, SrP_2O_6 glasses and liquid Hg were carried out using symmetrical reflection geometry, while those on $Pd_{80}Si_{30}$ and $Fe_{40}Ni_{40}P_{14}B_6$ metallic glasses were done using symmetrical transmission geometry. For each of the samples studied, a few EDXD spectra (five to ten) were collected at fixed diffraction angles 2θ, ranging from 2θ = 8 to 80°. The diffraction angles were so selected that any two consecutive Q regions of EDXD data, corresponding to two consecutive diffraction angles, are well overlapped in reciprocal space. Each EDXD spectrum was gathered over a period of time so long that at least 20,000 X-ray photons at an interval of about 50 eV were stored in the corresponding channel of the multi-channel pulse-height analyzer. All EDXD spectra obtained are composed of coherently, incoherently and multiply scattered from the sample and its environment; X-ray intensities follow the shape of the continuous energy spectrum of the X-ray source. The relation between the EDXD intensities registered at a given (fixed) diffraction angle, $I(E,\theta)$, and their individual constituents, specified above, may be expressed as follows /7/:

$$I(E,\theta) = C(E)\varepsilon(E)\{I_{coh}(E,\theta) + I_{ms}(E,\theta) + I_{bgr}(E,\theta) + I_{inc}(E,E',\theta)\} \tag{2}$$

where $C(E)$ is a normalization factor, $\varepsilon(E)$ is the detector efficiency factor, I_{coh} (E, θ), I_{ms} (E, θ) and I_{inc} (E, E', θ) are the intensities of coherently multiply and incoherently scattered from the sample and I_{bgr} (E, θ) is the background intensity arising from the specific sample environment (air, sample holder, etc.). The quantity of interest is the coherently scattered X-raya intensities I_{coh} (E, θ), which are to be extracted from the total EDXD spectrum $I(E, \theta)$ by properly accounting for all other contributions to it. Since the incoherent and multiple scattering from any sample with known physical characteristics (chemical composition, dimensions, etc.) can be calculated with enough accuracy and the background intensity can be experimentally measured with the sample removed from the sample holder, the only problem to be solved with the extraction of I_{coh} (E, θ) from I (E, θ) is the derivation of the unknown spectrum of the incident radiation beam I_p (E) = $C(E)\varepsilon(E)I_o$ (E), where $I_o(E)$ is the intensity profile of incident X-rays. Unfortunately, direct experimental determination of I_p (E) with accuracy sufficient to obtain accurate I_{coh} (E, Q) data is not an easy task as shown in the investigations of Nishikawa and Iijima /13/ and Uno and Ishigaki /14/. Therefore, the spectrum of the incident radiation beam I_p (E) was estimated by using an experimental EDXD spectrum of each sample as suggested by Egami /3/ and Wagner *et al.* /5/. The coherent X-ray intensities I_{coh} (E, θ) scattered by the sample are only a small fraction of the EDXD intensities registered at higher diffraction angles (say $2\theta > 80°$) and tend towards $<f>^2$ as the increment of Q, where $<f>^2$ is the average square of the atomic scattering factors of the interesting sample. Within the framework of this approach, equation (2) can now be solved for the unknown quantity I_p (E) and written in the following two equivalent forms:

$$I_n(E') = I_{coh}(E,\theta) / \{I_{inc}(E,E',\theta) + \frac{I_p(E)[<f>^2 + I_{ms}(E,\theta)]}{I_n(E')}\} \tag{3a}$$

$$I_p(E) = I_{coh}(E,\theta) / [<f>^2 + I_{ms}(E,\theta) + \frac{I_p(E')}{I_p(E)} I_{inc}(E,E',\theta)] \tag{3b}$$

where I_{coh} (E, Q) are the EDXD intensities already appropriate corrected for background and for the escape-peak effect encountered with the solid state detectors /7,12,13/ and the diffraction angle 2θ is high (let us say not less than 80°). When the amorphous material consists of relatively light atomic species and the incoherent scattering from the sample is stronger than the coherent one at high diffraction angle, as is the case with the SiO_2, SrP_2O_6 and $Fe_{40}Ni_{40}P_{16}B_4$ glasses, the spectrum of the incident beam radiation was calculated through an iterative procedure /5/ on the basis of equation 3(b), with an initial assumption I_p $(E')/I_p$ (E) = 1 coupled with the calculated values of I_{coh} (E, θ), I_{ms}, (E, θ) and I_{inc} (E, E', θ). In the case of $Pd_{80}Si_{20}$ glass and liquid Hg, which are presumably composed of heavy atomic species equation (3a) was applied with an initial assumption I_p $(E)/I_p$ (E') = 1. In this way, for each of the samples investigated the unknown quantity I_p (E) was determined. Then each EDXD spectrum from a given sample was normalized against the corresponding incident radiation beam I_p (E), and corrected for the corresponding contributions of the multiple and incoherent scattering according to the scheme of equation (2). Thus processed experimental EDXD data was reduced to the coherent single scattering intensity per atom $I^a_{coh}(Q)$ through the corrections for polarization and absorption. With the $I^a_{coh}(Q)$ data extracted from a particular EDXD spectrum, a segment of the frequently used Faber-Ziman type structure factor $S(Q)$ was calculated as follows:

$$S(Q) = \{I^a_{coh}(Q) - [\sum c_i f_i^2 - (\sum c_i f_i)^2]\} / (\sum c_i f_i)^2 \tag{4}$$

The well overlapping segments of $S(Q)$ data for a given sample were coupled to each other and a corresponding structure factor over an extended region of reciprocal space was constructed. The construction of a total $S(Q)$ started with the segment of $S(Q)$ derived from the EDXD data used for the determination of $I_p(E)$ as well, i.e. with the data corresponding to the highest diffraction angle measured at $2\theta = 80°$. The next $S(Q)$ segment, derived from the EDXD data obtained at the next lowest diffraction angle, was so scaled that the first one and a new master segment, an average value of the first and second segments, was constructed. The procedure was repeated by considering the next segments until the total structure factor was constructed. The part of $S(Q)$ below the lowest Q value covered by the present EDXD experiments (below about $Q = 8\ \mathrm{nm}^{-1}$) was smoothly extrapolated to $Q = 0$. Finally, the reduced atomic distribution functions $G(r)$, for all samples investigated were calculated by Fourier transformation of the corresponding experimental $S(Q)$ data. The structure factors and atomic distribution functions thus obtained for the present samples are shown in Figs. 1 - 4. After processing the raw EDXD experimental data, some careful tests on the quality of the $S(Q)$ and $G(r)$ structure-relevant data were carried out as well. First, the integral

$$\int_0^{Q_{max}} Q^2(S(Q)-1)dQ \qquad (5)$$

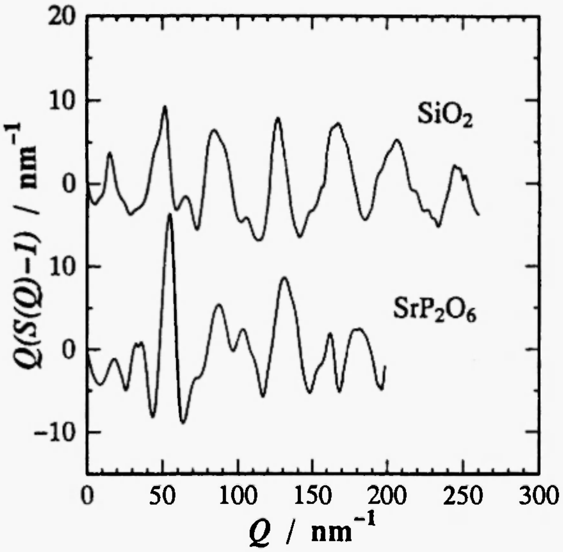

Fig. 1: Structure factors for SiO_2 and SrP_2O_6 glasses obtained by the present EDXD experiments.

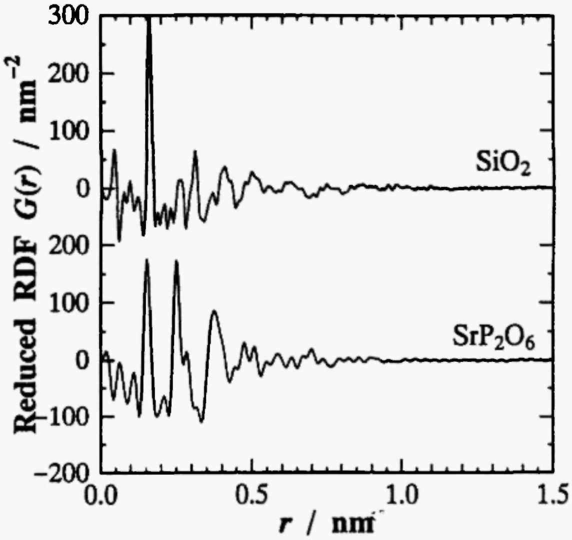

Fig. 2: Reduced radial distribution functions for SiO_2 and SrP_2O_6 glasses obtained by Fourier transformation of $S(Q)$ of Fig. 1.

which should equal $-2\pi^2\rho_0$ /1/ was calculated with the experimental $S(Q)$ data for all the materials presently investigated. Secondly, a straight line to the corresponding reduced atomic distribution functions $G(r)$s was fitted in the range below their first peaks to obtain an estimate for the average atomic number density ρ_0, on the basis of the expression:

$$G(r) = -4\pi r\rho_0 \qquad (6)$$

which holds for small values of r /1/. These tests confirmed the good quality of the structure data obtained by the present EDXD experiments. It may be added that all steps in the experimental EDXD data processing and derivation of the $S(Q)$ data briefly outlined here can be found in more detail in references /7, 12/.

3. RESULTS AND DISCUSSION

3.1 SiO_2 glass

As one can see in Fig. 1, the reduced structure factor for SiO_2 glass obtained by the present EDXD experiments exhibits prominent oscillations up to the maximum Q value of 250 nm^{-1}. The positions, shape and amplitude of the oscillations are rather similar to these of the physically relevant oscillations observed in the results of wide-angle X-ray diffraction experiments /15,16/. This is due to the fundamental features of the atomic arrangement of silicate glasses, indicating a distinct local nearest neighbor correlation such as SiO_4. In order to judge the reliability of the results obtained by the EDXD method, a comparison between the present $Q(S(Q)-1)$ data and other data for SiO_2 glass (available as a table) obtained by wide-angle experiments /16/ has been made. The comparison between both sets of data, made in Fig. 5a, shows that they are in good qualitative agreement but the presence of quantitative difference between the amplitudes of the corresponding peaks is recognized. Since the considered wide-angle diffraction data have been subjected to an additional manipulation so as to remove the oscillating features below the first peak in the corresponding reduced atomic distribution function $G(r)$, we applied a similar manipulation to our experimental data /17/. A comparison between the $S(Q)$ manipulated by us and that obtained by Konnert & Karle /16/ is shown in Fig. 5b. As one can see in the figure, both sets of data now compare in detail except for the amplitude of their first peaks at about 16 nm^{-1}. Since the amplitude of the first sharp peak in the structure factor of SiO_2 glass is suggested to be considerably influenced by the thermal history and the impurity content of the particular sample investigated /18/, the observed discrepancy in Fig.5(b) at the first oscillation peak may be attributed to the fact that two different silica glass samples have been investigated by us and by Konnert & Karle /16/. Therefore, one may conclude that the EDXD and the conventional wide-angle diffraction method, in fact, yield virtually the same structure data for disordered materials provided the data have been processed in a similar way and the particular samples investigated are the same. The EDXD data, however, cover a wider region of Q than those achieved by the wide-angle method with a conventional source of X-ray radiation. As one can see in Fig. 2, the reduced atomic distribution function $G(r)$ exhibits a dominant first peak at 0.161 nm and subsequent rapidly decaying oscillations. The peak reflects the presence of well defined Si-O first-neighbor atomic correlations in SiO_2 glass. By fitting the peak with an analytical function of a gaussian type, it has been found that the first neighbor Si-O separation is 0.1607±0.0005 nm and the number of first neighbor Si-O atoms is 3.9±0.2. These results are quite in line with the widely accepted description of the SiO_2 glass structure as an assembly of SiO_4 units. It may also be

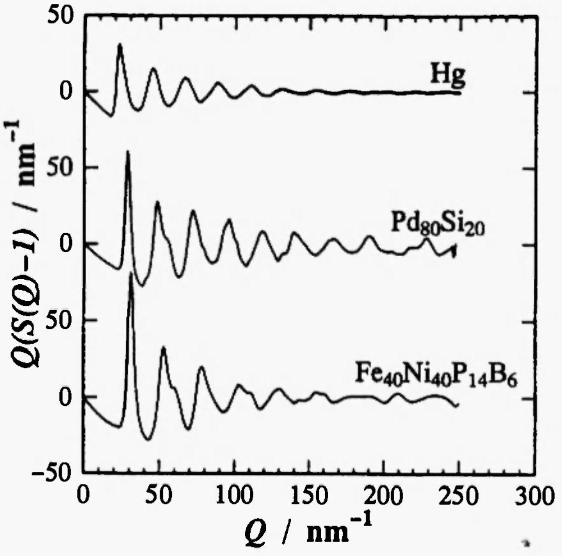

Fig. 3: Structure factors for $Pd_{80}Si_{20}$ and $Fe_{40}Ni_{40}P_{14}B_6$ metallic glasses and liquid Hg obtained by the present EDXD experiments.

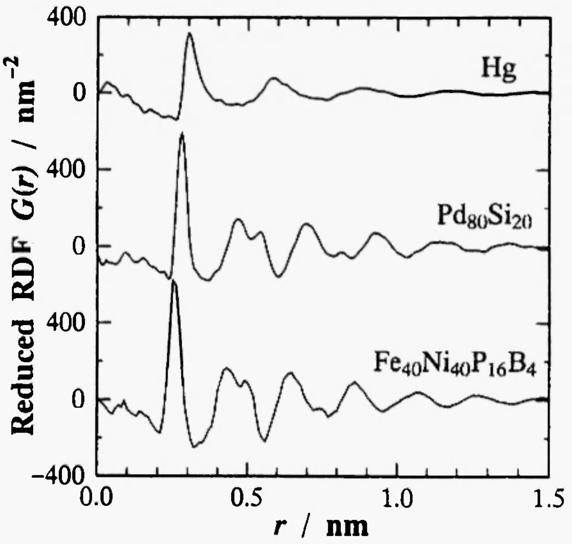

Fig. 4: Reduced radial distribution functions for $Pd_{80}Si_{20}$ and $Fe_{40}Ni_{40}P_{14}B_6$ metallic glasses and liquid Hg obtained by Fourier transformation of $S(Q)$ in Fig. 3.

added that the present results for the first neighbor Si-O distance and coordination number are in fairly good agreement with those (*r*=0.1609±0.0005 nm; *Z*=3.9 (0.2)) obtained by recent time-of-flight neutron diffraction experiments /19/. This agreement we consider as strong evidence in support to the reliability of the present EDXD experiments on SiO_2 glass.

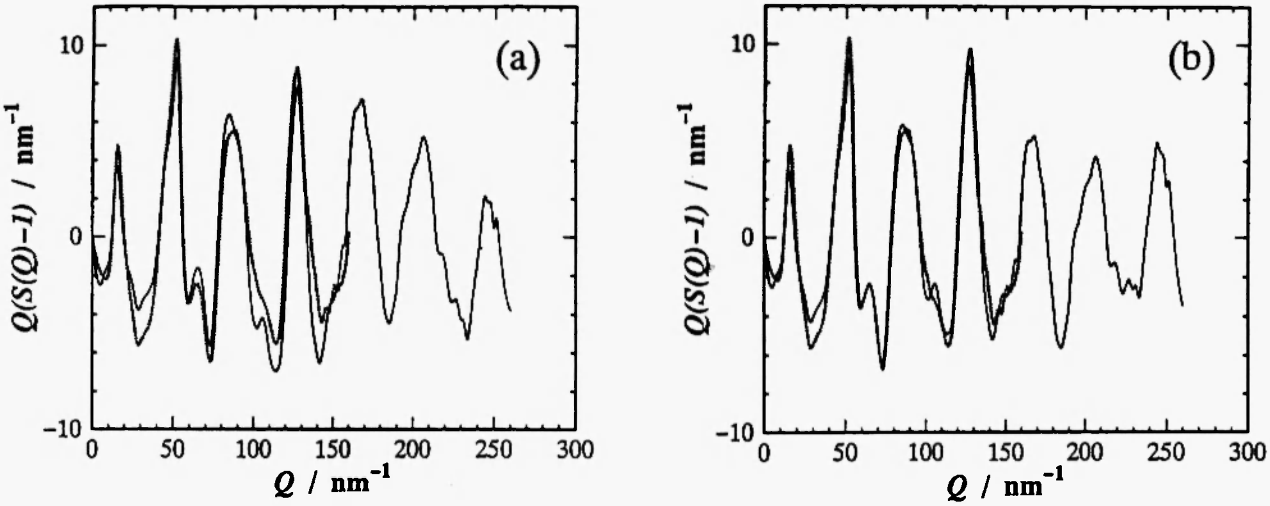

Fig. 5: Structure factor for SiO_2 glass obtained by the conventional X-ray diffraction /16/ (broken line) and by the present EDXD (full line) (a) without any artificial manipulation of the present results and (b) with application of extra manipulation on the present data removing the oscillations in the corresponding reduced atomic distribution function *G*(*r*), at low values of *r*. The wide-angle data have been properly rescaled because of the different sharpening factor used.

3.2 SrP2O6 glass

The reduced structure factor *S*(*Q*) for SrP_2O_6 glass also exhibits prominent oscillations up to the maximum *Q* value of 200 nm^{-1} reached by the present EDXD experiments. The corresponding atomic distribution function shown in Fig. 2 has a well defined first peak at 0.153 nm; a second one split up into two submaxima at 0.252 nm and 0.285 nm, respectively followed by oscillations which were not well pronounced. It may be reasonable to attribute the first peak in the experimental *G*(*r*) to the first neighbor P-O atomic correlations and the coordination number of oxygen around phosphorus has been estimated to be 4.2±0.2 by integrating the area of this peak. Therefore, the PO_4 tetrahedra are quantitatively confirmed as a local ordering unit structure of this glass and this is one of the common results in the alkali- or alkaline-earth phosphate glasses /20/. The second two maxima appear to be the harmony of the Sr-O ,O-O and P-P atomic correlations coupled with the knowledge of the ionic radii of Sr, P and O and present resolution of RDF could not allow to separate each contribution. For the discussion of further structural information on Sr-O, O-O and P-P correlations, another experimental approach such as AXS (anomalous X-ray scattering) /21/ is strongly required which enables us to obtain an environmental radial distribution function around Sr only. In order to demonstrate the importance of obtaining the structure factors over an interval of Q values as extended as possible, an atomic distribution function for SrP_2O_6 glass has been calculated again by including only a reduced part of the corresponding *Q*(*S*(*Q*)-1) data up to 140 nm^{-1}. A comparison between the first peaks in this function with those for *G*(*r*) based on the full set of diffraction data obtained (up to 200 nm^{-1}) is exemplified in Fig. 6. It is worth mentioning that the lack of diffraction data well extended in reciprocal space reduces the resolution of the corresponding atomic distribution function. In the present case, the reduced resolution is manifested by the diminished amplitude of the peaks in $G(r)$ at 0.152 nm and 0.250 nm and especially by the smearing out of the additional peak at 0.285 nm and this makes the differentiation between the atomic correlations much more ambiguous. This example illustrates well the potential of the EDXD method in obtaining structure data with improved resolution.

3.3 $Fe_{40}Ni_{40}P_{14}B_6$ and $Pd_{80}Si_{20}$ metallic glasses

By contrast with $S(Q)$ for SiO_2 and SrP_2O_6 oxide glasses, which show prominent oscillations up to the maximum Q value reached by the present EDXD experiments, $S(Q)$ for $Fe_{40}Ni_{40}P_{14}B_6$ and $Pd_{80}Si_{20}$ metallic glasses given in Fig. 3 show oscillations whose amplitudes rapidly decrease with increasing the value of wave vector Q. As frequently found with metallic glasses of chemical composition close to $Me_{80}T_{20}$ (Me = metal elements, T = nonmetal elements) a characteristic shoulder at the higher Q side of the second oscillation in the structure factors for $Fe_{40}Ni_{40}P_{14}B_6$ and $Pd_{80}Si_{20}$ metallic glasses is observed as well. The atomic distribution functions for both metallic glasses investigated have a well defined first peak, a second one split up into two components, a broad third one and subsequent small amplitude peaks vanishing at approximately 1.8 nm. The well defined first peak in $G(r)$ for $Fe_{40}Ni_{40}P_{14}B_6$ glass is positioned at 0.255 nm while that in $G(r)$ for $Pd_{80}Si_{20}$ at 0.278 nm. On the basis of the facts that atomic diameters of Fe, Ni and Pd are 0.254 nm, 0.250 nm and 0.276 nm and that the contributions of the metallic atoms involving atomic correlations to the experimental $G(r)$ data are predominant for both metallic glasses, one may well assume that the first peak in the experimental $G(r)$ function for $Fe_{40}Ni_{40}P_{14}B_6$ glass predominantly reflects the atomic distribution in the first coordination shells of Fe and Ni atoms while the first peak for $Pd_{80}Si_{20}$ glass reflects the atomic distribution in the first coordination shell of Pd atoms. By fitting the first peaks in these $G(r)$ by gaussians it has been found that the average first coordination number of metallic atoms in $Fe_{40}Ni_{40}P_{14}B_6$ glass is 12.5±0.5 and that of Pd atoms in the $Pd_{80}Si_{20}$ glass is 13.1±0.5 atoms. These results indicate that both metallic glasses investigated by us have quite a close packed structure. Here, it can also be pointed out that the basic structural features of $Fe_{40}Ni_{40}P_{14}B_6$ and $Pd_{80}Si_{20}$ metallic glasses revealed by the present EDXD studies, including the structure factors $S(Q)$, the corresponding atomic distribution functions $G(r)$ and structural parameters for the first neighbors, are close to those resulting from other independent diffraction studies on similar alloy systems /3,22-24/.

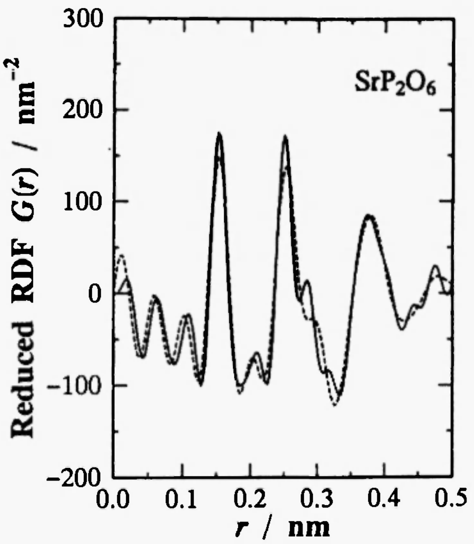

Fig. 6: Segments of the reduced atomic distribution functions $G(r)$, for SrP_2O_6 glass. Broken line indicates $G(r)$ data obtained by the Fourier transformation of only a part of the $Q(S(Q)-1)$ less than Q=140 nm^{-1}; full line indicates $G(r)$ data by the Fourier transformation of fullset of $Q(S(Q)-1)$ data up to 200nm^{-1}.

3.5 Liquid Hg

The reduced structure factor for liquid Hg obtained by the present EDXD experiments is shown in Fig. 3. It consists of rapidly decaying oscillations – a picture typical for most liquid metals. The first oscillation in the present reduced interference function data for liquid Hg is positioned at 23 nm^{-1}. The first peak in the corresponding atomic distribution function is at 0.30 nm as shown in Fig. 4. These values are also close to estimates reported in other structural studies on liquid

Hg /7,25-27/, although the present structure data extend over much wider region of reciprocal space. This experimental information over a wide-Q region is quite important and useful for constructing a three dimensional structural model with least-square variational technique /20,21/ or reverse Monte Carlo simulations /24,28/.

5. CONCLUSION

The results of the present studies show that good quality structure data for liquids and solid amorphous materials can be obtained over a region of reciprocal space more than 200 nm^{-1} by the EDXD method. The EDXD structure data can serve as a reliable basis for analysis of the atomic scale structure of a variety of disordered materials. Further progress in obtaining structure factors extended even up to 300 nm^{-1} can be achieved by employing more powerful laboratory sources of X-ray radiation, such as rotating anode generators, and by streamlining the procedures for raw EDXD data processing. All results reported in this paper were obtained as one of the research projects sponsored by Kimura Metamelt Project, ERATO, JRDC in the period between 1992 and 1993. The authors wish to thank Dr.S.Kimura of the National Institute for Research in Inorganic Materials and Prof. Y.Waseda, Institute for Advanced Materials Processing, Tohoku University, for their useful discussion and support.

REFERENCES

1. C.N.J. Wagner, in: *Liquid Metals, Physics and Chemistry*, S.Z.Beer (ed.), Marcel Dekker, New York, 1972.
2. Y. Waseda, *The Structure of Non-Crystalline Materials* , McGraw Hill, New York, 1980.
3. T. Egami, *J. Mater. Sci.*, **13**, 2587 (1978).
4. S. Aur, PhD thesis, Univ. of Pennsylvania, USA, 1981.
5. C.N.J. Wagner, D. Lee, S. Tai and L. Keller, *Adv. X-ray Analyses*, 24, 245 (1981).
6. W. Hoving, Ph.D. thesis, Univ. Groningen, Netherlands, 1986
7. V. Petkov and Y.Waseda, *J. Appl. Cryst.*, 26, 295 (1993).
8. J. Prober and J. Schultz, *J. Appl. Cryst.*, **8**, 405 (1975).
9. S. Hosokawa, T. Matsuoka and K. Tamura, *J. Phys.: Condens. Matter*, **3**, 4443 (1991).
10. R. Utz, A. Brunsch, P. Lamparter and S. Steeb, *Z. Naturforsch.*, a44, 1201 (1989).
11. V. Petkov, S. Takeda, Y. Waseda and K. Sugiyama, *J. Non-Cryst. Sol.*, **168**, 97 (1994).
12. K. Sugiyama, V. Petkov, S. Takeda and Y. Waseda, *Sci. Rep. Res. Inst. Tohoku Univ.*, **A38**, 1 (1993).
13. K. Nishikawa and T.Iijima, *Bull. Chem. Soc. Jpn.*, 57, 1750 (1984).
14. R. Uno and J. Ishigaki, *J. Appl. Cryst.*, **17**, 154 (1984).
15. R.L. Mozzi and B.E. Warren, *J. Appl.Cryst.*, 2, 164 (1969).
16. J.Konnert and J.Karle, *Acta Cryst.*, **A29**, 702 (1973).
17. R. Kaplow, S. Strong and B. Averbach, *Phys. Rev.*, **A138**, 1336 (1965).
18. Th. Gerber and B. Himmel, *J. Non-Cryst. Sol.*, 92, 408 (1987).
19. D. Grimley, C. Wright and R. Sinclair, *J. Non-Cryst. Sol.*, **119**, 49 (1990).
20. Y. Waseda, E. Matsubara, K. Sugiyama, I.K. Suh, T. Kawazoe, O. Kasu, M. Ashizuka and E. Ishidå, *Sci. Rep. Res. Inst. Tohoku Univ.*, **A35**, 19 (1990).
21. Y. Waseda, *Novel Application of Anomalous X-ray Scattering for Structural Characterization of Disordered Materials*, Springer-Verlag; New York, 1984.
22. G.S. Cargill III, *Solid State Physics*, Academic Press; New York, 1975.
23. Y. Waseda, H. Okazaki, M. Naka, and T. Masumoto, *Sci. Rep. Res. Inst. Tohoku Univ.*, A26, 12 (1976).
24. K. Suzuki, T. Fukunaga, M. Misawa, and T. Masumoto, *Mat.Sci. Engn.*, **23**, 215 (1976).
25. V. Rivlin, R. Waghorne and G. Williams, *Philos. Mag.*, **13**, 1169 (1966).
26. S. Franchetti, *Nuovo Cim.*, **B18**, 247 (1973).
27. L. Bosio, R. Cortes and C. Segaud, *J. Chem. Phys.*, **71**, 3595 (1979).
28. R.L. McGreevy, *Nucl. Instr. and Meth. in Phys. Res.*, **A354**, 1 (1995).