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ANOMALOUS X-RAY SCATTERING STUDIES OF SHORT-, INTERMEDIATE- AND EXTENDED-RANGE ORDER IN GLASSES

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ABSTRACT

We present the formalism of anomalous x-ray scattering as applied to partial structure analysis of disordered materials, and give an example of how the technique has been applied, together with that of neutron diffraction, to investigate short-, intermediate- and extended-range order in vitreous germania and rubidium germanate.

INTRODUCTION

While short-range order (SRO) in network glasses is generally well understood, usually in terms of structural units identified thorough comparison with analogous crystalline compounds, the intermediate-range order (IRO) — the manner in which these units are organized to form a large random network — remains a controversial issue.¹ Further, the modification of such glasses by the addition of metal oxides produces an extended-range order (ERO) that is also not well understood.² Part of the problem stems from the fact that a single diffraction measurement of an n -component system yields a weighted average structure factor of the $n(n+1)/2$ separate partial structure factors:

$$S(Q) = \sum_{ab} W_a(Q) W_b(Q) S_{ab}(Q) \quad (1)$$

where $W_a(Q)$ is the weighting factor for element a and $S_{ab}(Q)$ is the partial structure factor for the element pair (a,b) .³ For a multicomponent system it is therefore difficult to extract reliable information about a specific atom pair from a single experiment. However, in recent years improvements in the radiation sources available have led to the exploitation of complementary techniques which allow a more detailed investigation of the structure. Spallation neutron sources and instruments dedicated to amorphous and liquid systems make it possible to carry out neutron diffraction (ND) measurements with sufficient resolution to resolve peaks in the radial distribution function from different atom pairs; in favorable cases, isotope substitution can also be used to vary the weighting factors in Eq. (1).⁴ An especially important advance has been the advent of high-powered synchrotron x-ray sources which make it possible to obtain reliable difference measurements near the absorption edge of a particular element with anomalous x-ray scattering (AXS), varying the weighting factor for that element in Eq. (1) by changing the x-ray energy.⁵

In this review we present the formalism of AXS as applied to partial structure analysis of disordered materials, and then give an example of how the technique has been applied, together with that of neutron diffraction, to the case of vitreous germania and rubidium germanate.

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The scattering cross section in an x-ray diffraction measurement is given by a sum of products of scattering factors and phase factors over all pairs of atoms of the system:

$$\frac{d\sigma'}{d\Omega} = \frac{1}{N} \sum_{ij} f_i(\mathbf{Q}) f_j^*(\mathbf{Q}) \exp[i\mathbf{Q} \cdot (\mathbf{r}_i - \mathbf{r}_j)] \quad (2)$$

where $f_i(\mathbf{Q})$ is the form factor of atom i evaluated at the scattering vector $\mathbf{Q}$ and $\mathbf{r}_i$ its instantaneous position. It is convenient to reformulate Eq. (2) as the sum of three terms which, in the Faber-Ziman formulation⁶, becomes

$$\frac{d\sigma'}{d\Omega} = I(\mathbf{Q}) + [\langle |f(\mathbf{Q})|^2 \rangle - \langle f(\mathbf{Q}) \rangle^2] + N\delta_{\mathbf{Q},0} \langle f(0) \rangle^2 \quad (3)$$

where the angular brackets represent averages over all atoms in the sample. The first term in Eq. (3) is the *interference scattering* which contains the details of the atomic structure, the second is the *Laue diffuse scattering* which varies slowly with $\mathbf{Q}$, and the third is the *forward scattering* which is singular in the small-angle limit for a homogeneous system. Clearly the first term is the important one in the present context. In a multicomponent system, it can be expressed as a weighted sum over element pairs:

$$I(\mathbf{Q}) = \sum_{ab} c_a c_b f_a(\mathbf{Q}) f_b^*(\mathbf{Q}) S_{ab}(\mathbf{Q}) \quad (4)$$

where the *partial structure factor* (psf) $S_{ab}(\mathbf{Q})$ represents the structure associated with a given pair of elements (a, b) and is given by

$$S_{ab}(\mathbf{Q}) = \frac{N}{N_a N_b} \sum_{i \in a, j \in b} \exp[i\mathbf{Q} \cdot (\mathbf{r}_i - \mathbf{r}_j)] - \frac{N}{N_a} \delta_{ab} + 1 - N\delta_{\mathbf{Q},0} \quad (5)$$

It is often convenient to define an *average structure factor* $S(\mathbf{Q})$:

$$\begin{aligned} S(\mathbf{Q}) &= \frac{I(\mathbf{Q})}{\langle |f(\mathbf{Q})|^2 \rangle} \\ &= \sum_{ab} c_a c_b \frac{f_a(\mathbf{Q}) f_b^*(\mathbf{Q})}{\langle |f(\mathbf{Q})|^2 \rangle} S_{ab}(\mathbf{Q}) \end{aligned} \quad (6)$$

In the Faber-Ziman formalism used here, both $S(\mathbf{Q})$ and $S_{ab}(\mathbf{Q}) \rightarrow 1$ as $Q \rightarrow \infty$.

To obtain real-space information, pair correlation functions are obtained by Fourier transformation of the appropriate structure factors, e.g.:

$$g(r) - 1 = \frac{1}{8\pi^3 \rho} \int [S(\mathbf{Q}) - 1] \exp(-i\mathbf{Q} \cdot \mathbf{r}) d\mathbf{Q} \quad (7)$$

where $g(r)dr$ has a direct physical interpretation as the number of atoms in a volume element dr at a distance r from a reference atom at the origin.

We now treat the case of special case of anomalous scattering. Suppose that the incident energy is near an absorption edge for element A and we make a small change ΔE in the incident energy, which we assume has a significant effect on f_A but not on the other f_a . From Eq. (4), the corresponding change in $I(\mathbf{Q})$ is given by

$$\Delta I(\mathbf{Q}) = c_A \Delta f_A \sum_b c_b f_b^*(\mathbf{Q}) S_{Ab}(\mathbf{Q}) + c.c. \quad (8)$$

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In general, and especially near an absorption edge of element A, the form factor contains anomalous energy-dependent terms as well as the regular term:

$$f(Q) = f_0(Q) + f'(E) + if''(E) \quad (9)$$

Substitution of Eq. (9) into Eq. (8) gives, after some rearrangement,

$$\Delta I(Q) = 2c_A \Delta f'_A \sum_b c_b [(f_{b0}(Q) + f'_b) S'_{Ab} + f'_b S''_{Ab}] + 2c_A \Delta f''_A \sum_b c_b [f'_b S'_{Ab} - (f_{b0}(Q) + f'_b) S''_{Ab}] \quad (10)$$

By analogy with Eq. (6), we can define⁷ a *difference structure factor* for element A:

$$S_A(Q) = \frac{\Delta I(Q)}{2c_A \Delta f'_A \langle f_0(Q) + f' \rangle + 2c_A \Delta f''_A \langle f'' \rangle} = \frac{2c_A \Delta f'_A \sum_b c_b [(f_{b0}(Q) + f'_b) S'_{Ab} + f'_b S''_{Ab}] + 2c_A \Delta f''_A \sum_b c_b [f'_b S'_{Ab} - (f_{b0}(Q) + f'_b) S''_{Ab}]}{2c_A \Delta f'_A \langle f_0(Q) + f' \rangle + 2c_A \Delta f''_A \langle f'' \rangle} \quad (11)$$

which also $\rightarrow 1$ as $Q \rightarrow \infty$. Notice that $S_A(Q)$ as defined in Eq. (11) implicitly depends on both E and ΔE . The ΔE dependence could be removed by taking the limit of Eq. (11) as $\Delta E \rightarrow 0$. However, the error in the measured value of $S_A(Q)$ obviously becomes very large as ΔE is made small. In practice we try to choose a compromise value for ΔE that is large enough to reduce this error and small enough so that we can neglect the changes in f_b , $b \neq A$. The values of the f_b in Eq. (11) are then evaluated at $E + 1/2 \Delta E$. The choice of the distance of E from the absorption edge of element A is also a compromise between large values for the anomalous terms in f_A versus an accurate knowledge of Δf_A .

Eq. (11) can be simplified with the help of two approximations. First, a good approximation below the edge of element A is

$$\Delta f_A'' \ll \Delta f_A' \quad (12)$$

E.g., for Ge at $E_{Ge} - 17$ eV, $f_{Ge}' = -6.169$, $f_{Ge}'' = 0.494$; at $E_{Ge} - 200$ eV, $f_{Ge}' = -3.987$, $f_{Ge}'' = 0.504$. With this approximation Eq. (11) simplifies to

$$S_A(Q) = \frac{\sum_b c_b [(f_{b0}(Q) + f'_b) S'_{Ab} + f'_b S''_{Ab}]}{\langle f_0(Q) + f' \rangle} \quad (13)$$

Second, if the incident energies are far removed from the absorption edges of the elements $b \neq A$, a reasonable approximation is

$$f_b'' \ll f_{b0}, b \neq A. \quad (14)$$

In this case, taking account of the fact that S_{AA} has to be real, Eq. (13) simplifies finally to

$$S_A(Q) = \sum_b c_b \frac{(f_{b0}(Q) + f'_b)}{\langle f_0(Q) + f' \rangle} S'_{Ab} \quad (15)$$

To obtain the corresponding real-space information, we can use the equivalent of Eq. (7) to obtain the average real-space environment about an atom of element A.

EXPERIMENTS

We have addressed the issues of the SRO, IRO and ERO in a series of combined AXS and ND experiments on experiments on germania and rubidium germanate glasses.⁸ The AXS measurements were carried out at the X-7A beam line at the National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory, and the ND measurements at the GLAD facility at the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory.

Germania- rather than silica-based glasses, and rubidium as the modifier element, were chosen because the energies of the Ge and Rb edges were suitable for AXS at the X-7A beamline. The glasses were prepared in solid form and polished to give smooth ($\sim 50\mu$ roughness) flat surfaces toward the x-ray beam. A series of AXS measurements were carried out at the Ge and Rb K edges. The Rb edge measurements in the rubidium germanate glass were complicated by the high level of Ge fluorescence. For the neutron measurements the glasses were crushed and loaded into thin-walled vanadium tubes.

RESULTS

1. Neutron Diffraction

Neutron structure factors obtained for germania and rubidium germanate glasses are shown in Fig. 1. The results for the binary are in good agreement with those of Desai *et al.*⁹ The first feature in the structure factor of GeO_2 is the peak at $Q_1 = 1.54 \text{ \AA}^{-1}$, corresponding to a correlation length $L_1 = 2\pi/Q_1 \sim 4.1 \text{ \AA}$ characteristic of IRO in oxide and chalcogenide glasses.¹

On the addition of only 10 % of the Rb_2O modifier to the GeO_2 network, a new peak arises at $Q_0 \sim 0.95 \text{ \AA}^{-1}$, implying ERO on a length scale $L_0 \sim 6.6 \text{ \AA}$. Q_0 is unaffected by changes in Rb_2O concentration x , but the peak height increases up to $x = 0.2$ and then decreases for larger x . This dependence reflects the behavior of the other physical properties such as T_g and viscosity. In the modified glass, the FSDP observed in GeO_2 at $1.54 \text{ \AA}^{-1}$ is replaced by a second peak at larger Q . Both the intensity of this peak and its Q value increase with x . Its position corresponds to a length scale L_1 ranging from 3.1 to 3.6 $\text{\AA}$, slightly smaller than the characteristic scale of IRO in binary glasses.

2. X-Ray Diffraction

X-ray structure factors for GeO_2 and $(\text{Rb}_2\text{O})_{0.2}(\text{GeO}_2)_{0.8}$ are shown in Fig. 2. Comparison of neutron and x-ray diffraction data makes it possible to infer information about the nature and origins of these two features, since the appropriate weighting factors are generally quite different for a given element. The same features are observed as in the neutron structure factors; however, for GeO_2 the FSDP is stronger in $S^x(Q)$ than in $S^N(Q)$, indicating that the cation-cation correlations play an important role in its existence, as is generally the case in oxide and chalcogenide glasses.¹⁰⁻¹³ For $(\text{Rb}_2\text{O})_{0.2}(\text{GeO}_2)_{0.8}$, the first peak is much weaker, and the second much stronger, than in $S^N(Q)$. Since correlations involving two cations (Ge or Rb) are emphasized in S^x relative to S^N , while all correlations involving O are de-emphasized, the relative weakness of the first peak in $S^x(Q)$ shows that correlations involving oxygen atoms are important; cation-cation correlations that make a *negative* contribution to this peak may also be involved. On the other hand, the dominance of the second peak in $S^x(Q)$ shows that the cation-cation correlations play a crucial role in its origin, as in that of the FSDP in the binary.

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Fig. 1. Neutron structure factor $S^N(Q)$ of $(\text{Rb}_2\text{O})_x(\text{GeO}_2)_{1-x}$ glasses; successive curves are displaced upward by one unit for clarity.

3. Anomalous X-Ray Diffraction

Further information about the origin of the IRO and ERO is obtained from the results of AXS measurements made near the Ge and Rb K-shell absorption edges. S_{Ge}^x is shown for GeO_2 and $(Rb_2O)_{0.2}(GeO_2)_{0.8}$ in Fig. 3, and S_{Rb}^x for $(Rb_2O)_{0.2}(GeO_2)_{0.8}$ in Fig. 4. For GeO_2 , S_{Ge}^x has the same features as $S^x(Q)$, the main difference being that the peak at $Q_2 = 2.8 \text{ \AA}^{-1}$, attributed to chemical SRO of Ge and O, is considerably enhanced. In the molecular dynamics (MD) simulations of SiO_2 glass, S_{SiSi} and S_{OO} make positive contributions to this peak while S_{SiO} makes a negative contribution of almost identical magnitude.^{10,13} In all three partials, the peak arises from Coulomb oscillations in the partial pair correlation function $g_{ij}(r)$ with period $L_2 \sim 2.2 \text{ \AA}$, in phase with $\sin(Q_2 r)$ for g_{SiSi} and g_{OO} but out of phase for g_{SiO} . By analogy with SiO_2 and taking into account differences in the appropriate weighing factors, this peak should be scarcely visible in $S^w(Q)$, stronger in $S^x(Q)$ and considerably enhanced in S_{Ge}^x exactly the behavior observed.

For $(Rb_2O)_{0.2}(GeO_2)_{0.8}$, the intensity of the first peak is positively correlated with the scattering amplitude of Ge (Fig. 3), but *negatively* correlated with that of Rb (Fig. 4). This behavior can be explained in terms of a chemical ordering of Rb and Ge with a period $L_0 \sim 6.6 \text{ \AA}$, S_{RbRb} and S_{GeGe} being positive and S_{RbGe} negative at Q_0 . The second peak has a strong positive correlation with both Rb and Ge scattering amplitudes, consistent with the crucial role of the cation-cation correlations discussed above. In molecular dynamics simulations¹⁴ on the analogous silicate system $(Rb_2O)_{0.2}(SiO_2)_{0.8}$, the Rb-Rb partial structure factor shows positive peaks at 1.1 and 1.9 $\text{\AA}^{-1}$, in agreement with this interpretation.

Fig. 2. X-ray structure factor $S^x(Q)$ of GeO_2 and $(Rb_2O)_{0.2}(GeO_2)_{0.8}$ glasses.

Fig. 3. Germanium difference structure factor $S_{\text{Ge}}^x(Q)$ of GeO_2 and $(\text{Rb}_2\text{O})_{0.2}(\text{GeO}_2)_{0.8}$ glasses.

Fig. 4. Rb difference structure factor $S_{\text{Rb}}^x(Q)$ of $(\text{Rb}_2\text{O})_{0.2}(\text{GeO}_2)_{0.8}$ glass.

Writing the ND, XRD and AXS data presented above as a vector $\mathbf{F}(Q) = [S^N(Q), S^X_{13}(Q), S^X_{200}(Q)]$ and the required psf's as a vector $\mathbf{S}(Q) = [S_{\text{GeGe}}(Q), S_{\text{Oo}}(Q), S_{\text{GeO}}(Q)]$, the two vectors have a linear relationship

$$\mathbf{F} = \mathbf{A}\mathbf{S} \quad (16)$$

where the elements of $\mathbf{A}$ are the weighting factors in Eqs. (6) and (15). Inserting the appropriate values for the nuclear and atomic parameters, it is found that degree of conditioning of $\mathbf{A}^{-1}$ is comparable to that obtained in many ND experiments with isotope substitution, so that reliable structure factors should be derived, provided the systematic and statistical errors are small.

The results for the psf's are shown as points in Fig. 5. The FSDP shows up as a positive peak in S_{GeGe} and S_{GeO} and a shallow negative peak in S_{Oo} , supporting the assertion that cation correlations dominate the IRO. The peak at $2.6 \text{ \AA}^{-1}$ is strong and positive, with almost the same height, in S_{GeGe} and S_{Oo} , and strong and negative, with almost the same magnitude, in S_{GeO} , confirming the chemical SRO nature of this peak. The topological SRO peak occurs at $4.4 \text{ \AA}^{-1}$, predominantly in S_{GeGe} .

We are not aware of any published Molecular Dynamics (MD) computer simulations of GeO_2 , but SiO_2 has been studied with classical MD by Vashishta *et al.*¹⁰ and with *ab initio* MD by Sarnthein *et al.*¹³ The *ab initio* MD results,^{13,15} plotted as solid lines in Fig. 5 for comparison, are generally similar to the present ones for GeO_2 but displaced to higher Q due to the smaller length scale in silica. The main qualitative difference is that the FSDP and topological SRO peaks show up as weak positive features in S_{Oo} .

An alternative formulation of the partial structure factors was derived by Bhatia and Thornton¹⁶ in terms of number density (N) and concentration (C) fluctuations, shown in Fig. 6 for the present data and the MD results for SiO_2 . A matter of some interest is the behavior of S_{CC} in the region of the FSDP, in view of the speculation of Elliott that the FSDP arises from concentration fluctuations in oxide and chalcogenide glasses.¹⁷ In the present results for GeO_2 , S_{CC} is in fact slightly negative in this region, so this explanation does not seem to apply here. As expected, the CSRO peak shows up most strongly in S_{CC} and the TSRO peak in S_{NN} . The MD results for SiO_2 are again qualitatively similar with a general shift to higher Q . For these, S_{CC} is close to zero in the region of the FSDP, again at variance with Ref. 17. A similar behavior was found was found in the classical MD result for SiO_2 .¹⁰

DISCUSSION

This work provides results for the SRO and IRO of vitreous GeO_2 in terms of the contributions of correlations involving the three element pairs. The results are consistent with the identification of the first three peaks in the average neutron and x-ray structure factors with IRO, chemical SRO and topological SRO, respectively. Results for the FSDP indicate that in this glass IRO is associated with cation correlations as opposed to concentration-concentration correlations as has been suggested.

In the rubidium -modified glass, the strong oxygen correlations implied by the large weight of the first peak in the neutron diffraction pattern support the picture of Wright *et al.*¹⁸ for network glasses modified with heavy alkali ions. In the binary, the FSDP reflects the periodicity arising from the succession of network cages;¹⁹ in the modified glasses, the Rb^+ ions enter the larger cages and displace the oxygen atoms on their boundaries. This makes the boundaries of the large cages more spherical and leads to a strong diffraction peak at low Q . Clearly, this peak will be associated with strong oxygen correlations and with a chemical order due to the alternation of Ge and Rb ions. The cages without the Rb^+ ions will be correspondingly contracted, causing a shift of the second diffraction peak to Q values higher than the FSDP of the GeO_2 .

BEYOND AXS?

The AXS technique makes use of the changes in the real part of $f_A(Q)$ on going through an absorption edge of element A. The question arises as to whether, if the high absorption for x-ray

Fig. 5. Measured partial structure factors (Faber-Ziman definition) for the three atom pairs in vitreous GeO_2 ($M = \text{Ge}$, points), together with corresponding results from *ab initio* MD computer simulation^{13,15} of vitreous silica ($M = \text{Si}$, lines).

Fig. 6. Measured partial structure factors (Bhatia-Thornton definition¹⁶) in vitreous GeO_2 (points), together with corresponding results from *ab initio* MD computer simulation^{13,15} of vitreous silica (lines).

energies above the edge can be tolerated, changes in the imaginary part of $f_A(Q)$ can be exploited as well. Inserting Eq. (9) into Eq. (6), the result can be written

$$I(Q) = A(Q) + 2f'_A(E)B(Q) + [f'_A(E)^2 + f''_A(E)^2]C(Q) \quad (17)$$

where

$$A(Q) = \sum_{ab} c_a c_b f_{a0}(Q) f_{b0}^*(Q) S_{ab}(Q) \quad (18a)$$

$$B(Q) = c_A \sum_{ab} c_b f_{b0}(Q) S'_{Ab}(Q) \quad (18b)$$

$$C(Q) = c_A^2 S_{AA}(Q) \quad (18c)$$

Since the coefficients in Eq. (17) are linearly independent functions of E , $A(Q)$, $B(Q)$, $C(Q)$ can in principle be determined independently, allowing a full partial analysis on the basis of the x-ray data alone. Furthermore, f'_A and f''_A are also determined in the process for the sample under study, and will be quite different for a condensed sample compared with a free atom, a fact that has of course been exploited for many years in structural studies with absorption spectroscopy. In a diffraction context this leads to the "diffraction anomalous fine-structure" (DAFS) technique which has been widely used in the analysis of mixed or mixed-valence crystalline materials.²⁰ In the context of glass structure, this approach would yield information about structural irregularities in the glass as long as the scale for these is large compared with the range of the photoelectron. Such may well be the case for intermediate or extended range order in inorganic glasses.

CONCLUSION

This example makes it clear that AXS is a powerful technique for obtaining partial structure information in complex disordered materials, and in particular can probe the structure of minority species in a majority host. With the more powerful sources now available, and a corresponding development in experimental methods and analysis techniques, we can expect that before too long full partial structure analysis, with complete derivation of all partial structure factors, will become routine. Further, we may expect that DAFS type information may become available as a probe of structural inhomogeneities in glasses and other disordered materials.

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REFERENCES

1. S. C. Moss and D. L. Price, in *Physics of Disordered Materials*, ed. D. Adler, H. Fritzschke and S. R. Ovshinsky (Plenum, New York, 1985), p. 77; D. L. Price, S. C. Moss, R. Reijers, M.-L. Saboungi and S. Susman, *J. Phys.: Condens. Matter* 1, 1005 (1989); S. R. Elliott, *Nature* 354, 445 (1991).
2. P. Armand, M. Beno, A. J. G. Ellison, G. S. Knapp, D. L. Price and M.-L. Saboungi, *Europhys. Lett.* 29, 549 (1995).
3. Y. Waseda, *The Structure of Non-Crystalline Materials*. (McGraw-Hill, New York, 1980); *Methods in the Determination of Partial Structure Factors of Disordered Matter by Neutron and*

- Anomalous X-Ray Diffraction*, Ed. J. B. Suck, P. Chieux, D. Raoux and C. Riekel (World Scientific, Singapore, 1993).
4. D. L. Price, Ed., *Research Opportunities In Amorphous Solids With Pulsed Neutron Sources*, J. Non-Cryst. Solids 76, No. 1 (1985)
 5. P. H. Fuoss, P. Eisenberger, W. K. Warburton and A. Bienenstock, Phys. Rev. Lett. 46, 1537 (1991).
 6. T. E. Faber and J. M. Ziman, Phil. Mag. 11, 153 (1965).
 7. D. L. Price and M.-L. Saboungi, in Local Structure from Diffraction, Ed. S. J. L. Billinge and M. F. Thorpe (Plenum, NY, 1998), in press.
 8. D. L. Price, A. J. G. Ellison, M.-L. Saboungi, R.-Z. Hu, T. Egami and W. S. Howells, Phys. Rev. B 55, 11249 (1997).
 9. J. A. E. Desa, A. C. Wright and R. Sinclair, J. Non-Cryst. Solids 99, 276-288.
 10. P. Vashishta, R. K. Kalia, J. P. Rino and I. Ebbsjö, Phys. Rev. B 41, 12197-12208 (1990).
 11. I. T. Penfold and P. S. Salmon, Phys. Rev. Lett. 67, 97-100 (1991).
 12. P. Vashishta, R. K. Kalia, G. A. Antonio and I. Ebbsjö, Phys. Rev. Lett. 62, 1651-1654 (1989).
 13. J. Sarnthein, A. Pasquarello and R. Car, Phys. Rev. Lett. 74, 4682 (1995); Phys. Rev. B 52, 12690 (1995).
 14. J. Kieffer and D. Nekhayev, private communication
 15. A. Pasquarello, private communication (1998)
 16. A. Bhatia and D. Thornton, Phys. Rev. B 2, 3004 (1970)
 17. S. R. Elliott, Phys. Rev. Lett. 67, 711 (1991)
 18. A. C. Wright, A. G. Clare, B. Bachra, R. N. Sinclair, A. C. Hannon and B. Vessal, Trans. ACA 27, 239-254 (1991).
 19. A. C. Wright, in *Proc. Symp. on Glass Structure, Amer. Cer. Soc. Mtg. Honolulu, HI, Nov. 1993*, J. Non-Cryst. Solids 179, 84-115 (1994)
 20. I. J. Pickering et al., J. Am. Chem. Soc. 115, 6302 (1993), and references therein.