

## A study of the vibrational spectrum of amorphous and crystalline SeTe samples by inelastic neutron scattering

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1993 J. Phys.: Condens. Matter 5 637

(<http://iopscience.iop.org/0953-8984/5/6/001>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.96

The article was downloaded on 11/05/2010 at 01:06

Please note that [terms and conditions apply](#).

## A study of the vibrational spectrum of amorphous and crystalline SeTe samples by inelastic neutron scattering

L M Needham†‡, M Cutroni§, A J Dianoux† and H M Rosenberg||

† Institut Laue-Langevin, 38042 Grenoble Cédex, France

‡ Department of Physics, University of Warwick, Coventry CV4 7AL, UK

§ Department of Physics, University of Messina, 98166, Italy

|| The Clarendon Laboratory, Oxford, OX1 3PU, UK

Received 15 July 1992, in final form 5 October 1992

**Abstract.** The vibrational density of states of both crystalline (trigonal) and amorphous samples of  $\text{Se}_{96.85}\text{Te}_{3.15}$ ,  $\text{Se}_{93.6}\text{Te}_{6.4}$ ,  $\text{Se}_{86.6}\text{Te}_{13.4}$ ,  $\text{Se}_{79}\text{Te}_{21}$  and of crystalline Se have been determined by inelastic neutron scattering. In each case the spectrum was found to be divided into three bands which for the amorphous specimens are at 5.6, 14 and 30.5 meV and for the crystalline specimens are at 6.5, 14 and 28.5 meV. At the lowest energies the amorphous samples, in common with other amorphous materials, show an  $\omega^2$ -dependence which then increases over a short range to a higher power of the energy. The crystalline specimens have an  $\omega^{1.5}$ -dependence at low energies. The temperature dependence of the specific heat of crystalline Se calculated from these data is in good agreement with that quoted in the literature.  $S(Q)$  for the amorphous samples was also measured and this shows that the first- and second-neighbour distances are similar to those for crystalline Se. This study suggests that crystalline SeTe resembles a chain polymer of helical chains with weak inter-chain bonding. In the amorphous samples the structure is similar but with inter-chain entanglements.

### 1. Introduction

This paper presents neutron scattering measurements of the density of states  $g(\omega)$  as a function of the energy  $\hbar\omega$  for amorphous and crystalline samples of SeTe alloys. The reason why such an investigation is of interest is that recent measurements have shown that  $g(\omega)$  does not follow a simple Debye  $\omega^2$  spectrum. Experiments indicate that in amorphous materials additional excitations are present which modify this behaviour. For example in the case of amorphous  $\text{SiO}_2$  Buchenau *et al* [1] have shown that the extra excitations might be due to coupled rotations of  $\text{SiO}_2$  tetrahedra, whilst for epoxy-resins [2] a rise above the  $\omega^2$  dependence may be accounted for if one assumes that the complex nature of the amorphous network is fractal. It has been shown by Aharony *et al* [3], that such a system can have a frequency dependence which, over a certain range, can rise faster than  $\omega^2$ .

In order to further our understanding of amorphous systems we felt it would be useful to make a comparison between  $g(\omega)$  for crystalline and for amorphous samples of the same material. This would then give us some confidence that any 'anomaly' which might be found in  $g(\omega)$  for the amorphous system might be really due to its glassy nature if it was not also found for the crystalline sample. In fact our results

show that the crystalline samples also exhibit interesting features and their density of states does not follow three-dimensional Debye behaviour at low energies.

We chose to study SeTe alloys, since they have not been investigated before by neutron scattering and both crystalline and amorphous samples can be easily prepared. A preliminary account of the work on amorphous samples has already been given [4]. (It should be noted that the specimen compositions in that paper are given in weight percent and not atomic percent.) In addition to the density of states, we also investigated the structure of the amorphous material by neutron diffraction.

## 2. Structure of Se-Te

Se has six allotropes [5]. Three are amorphous—glassy, black and red. Three are crystalline—one trigonal and two monoclinic. Our amorphous specimens were of the glassy form and the crystalline specimens were trigonal—this is the most stable of the structures—with  $a = 0.4366$  nm and  $c = 0.4954$  nm. It is composed of either left- or right-handed helical chains (see figure 1). The shortest distance between atoms on a chain is 0.2373 nm with a bond angle of  $101^\circ$  [5]. The shortest distance of two atoms on neighbouring chains is 0.346 nm. There is strong covalent bonding within the chains and weak bonding (possibly of the van der Waals type) between them. Te substitutes randomly for the Se up to a concentration of about 25 at. %.

## 3. Samples and experiments

Samples of both amorphous and crystalline Se-Te containing 3.15, 6.4, 13.4 and 21 at. % Te, and also a pure Se crystalline (trigonal) sample, were prepared at the Department of Physics, University of Messina, Italy. The ingots were powdered and specimens each weighing about 30 g were investigated using the time-of-flight neutron spectrometer IN6 at the Institut Laue-Langevin (ILL), Grenoble, France. The scattering of neutrons with incident energy 3.12 meV was studied in the up-scattering region over an energy range 0–50 meV.

The samples were contained in an aluminium sample holder of effective diameter 50 mm and thickness 5 mm. The neutrons were incident at  $135^\circ$  to the sample plane. Data were taken over a range of scattering angles from  $10^\circ$  to  $115^\circ$  when the sample was held at 100 and 200 K and in a few cases at 300 K. However, only the 100 K data were used in the analysis presented in this paper, in order to limit the multiphonon contribution.

Similar measurements at 10 K on some crystalline samples were also taken at higher incident neutron energies, 17.12 meV and 115 meV, using the time-of-flight spectrometer IN4 at the ILL. In this case the down-scattered neutrons were detected from  $-48^\circ$  to  $+101^\circ$ . These measurements do not have the precision nor the resolution of the data taken on IN6, but as will be discussed below, they provided complementary results which confirmed the validity of the assumption made in the analysis of the IN6 data.

Measurements of the structure factor  $S(Q)$  were taken at room temperature on the amorphous samples containing 3.15, 13.5 and 21 at. % Te using the diffractometer LAD, at the ISIS facility, Rutherford Appleton Laboratory, Chilton, UK. The samples were contained in a cylindrical Zr-Ti holder (to reduce coherent background scattering) which was 10 mm diameter and 50 mm high.

#### 4. Analysis of the experimental data

The neutron scattering experiments performed both on IN4 and IN6 provide information about the number of neutrons scattered per unit energy range into unit solid angle. This quantity, the double differential cross section,  $d^2\sigma/d\omega d\Omega$  is defined as

$$\frac{d^2\sigma}{d\omega d\Omega} = \frac{k_f}{k_i} \exp\left(\frac{-\hbar\omega}{2kT}\right) \frac{\sigma}{4\pi} \tilde{S}(Q, \omega) \quad (1)$$

where  $k_f$  and  $k_i$  are the emergent and incident neutron wave vectors respectively,  $\sigma$  is the average neutron scattering cross section and  $\tilde{S}(Q, \omega)$  is the symmetrized scattering function. There are two methods [6] which can be used for extracting the density of states  $g(\omega)$  from the data. The first, the so-called incoherent approximation, sets  $\tilde{S}(Q, \omega)$  equal to  $S_{\text{inc}}$  where

$$S_{\text{inc}} = \sum_i \frac{b_i^2}{\langle b^2 \rangle} \exp(-2W_i) \frac{Q_i^2}{4M_i} \frac{g_i(\omega)}{\omega \sinh(\hbar\omega/2kT)} \quad (2)$$

where  $W_i$  is the Debye-Waller factor,  $Q_i$  is the modulus of the change in neutron wave vector,  $\omega$  is the energy transfer,  $g_i(\omega)$  is the density of states,  $b_i$  is the scattering length of the  $i$ th nucleus and  $\langle \rangle$  indicates an average over all  $b_i^2$ .  $M_i$  is the atomic mass. This approximation, which is sometimes referred to as the self-scattering approximation, neglects distinct contributions to the scattering and analyses it as though it were only self-scattering.

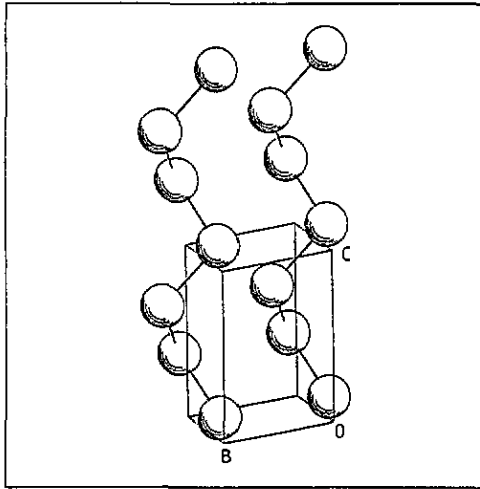
The data presented in this paper have been treated using this approximation and yield plots of the generalized density of states. But since both Se and Te are mainly coherent scatterers the validity of the incoherent analysis (which should only use data for large  $Q$ ) is suspect because data for small  $Q$  values could not be excluded. However a second more rigorous treatment, using a coherent analysis following the method of Buchenau [7], was found by us [13] to be no more satisfactory for our samples since it then has to be assumed that the cross section can be written as

$$\frac{d^2\sigma}{d\omega d\Omega} = \frac{k_f}{k_i} \frac{3\hbar N}{4\pi} \frac{g(\omega)}{\omega} n_B I(Q, \omega) \quad (3)$$

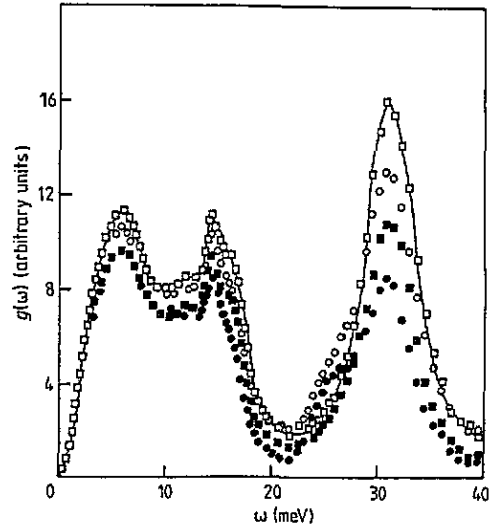
where  $n_B$  is  $[(\exp(\hbar\omega/kT) - 1)]^{-1}$ ,  $N$  is the number of atoms, and  $I(Q, \omega)$  is assumed to have the same  $Q$ -dependence for all values of  $\omega$ —it is not at all obvious that such an assumption is justified. A full analysis and discussion of the  $Q$ -dependence of our samples will be published in a later paper [14].

#### 5. General form of the results

Figures 2 and 3 show  $g(\omega)$  as a function of  $\omega$  for the amorphous and crystalline samples using data taken on IN6. The curves were derived using the incoherent analysis. They show a significant  $g(\omega)$  distribution up to about 35 meV in which there are three well defined peaks. Above a few meV both the amorphous and the



**Figure 1.** Two neighbouring helices of crystalline Se showing their relationship to the trigonal unit cell (courtesy of Dr A M Glazer).



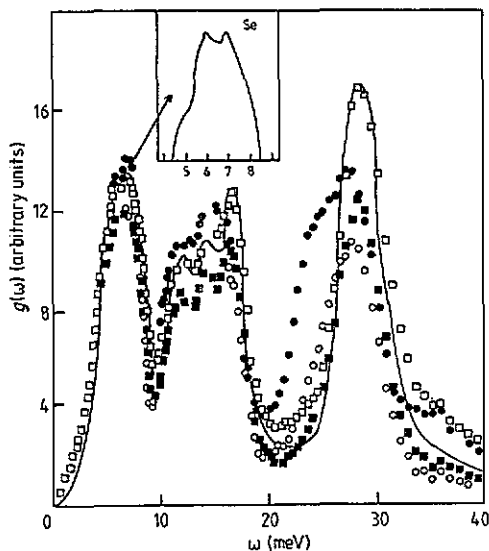
**Figure 2.** The density of states  $g(\omega)$  plotted against the energy,  $\omega$ , for the four amorphous Se–Te samples. For clarity some points at low energy are omitted. Note the wing on the low-energy side of the 30 meV peak for Se<sub>79</sub>. □: Se<sub>96.85</sub>. ■: Se<sub>93.6</sub>. ○: Se<sub>86.6</sub>. ●: Se<sub>79</sub>.

crystalline specimens have similar features except that the two lower peaks are shifted to lower energies for the amorphous specimens.

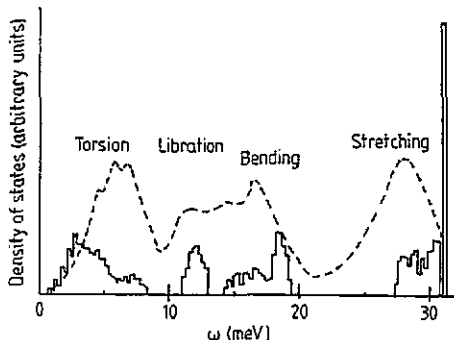
Starting from zero energy there is first of all a continuous spectrum which peaks at about 5.6 and 6.5 meV for the amorphous and crystalline samples respectively. In the latter this can be resolved into three sub-peaks—see inset to figure 3. This is followed by a more complex peak (also split in the crystalline samples) at 14 meV. At much higher energies there is a quite separate well defined peak at 28.5 meV (crystalline) and 30.5 meV (amorphous). As more Te is added to the samples the peak energies remain almost unchanged until we reach both amorphous and crystalline Se<sub>79</sub> samples, where a shoulder appears on the low-energy side of the highest peak.

This triple-peaked spectrum is similar to that calculated for Se by Etchepare *et al* [8] (see figure 4). It was also found experimentally in an amorphous sample of Se by Gompf [9] and in the SeGe and SeAsGe systems by Kamitakahara *et al* [10].

A prediction of the theory [8] is that the number of stretching modes should be one third of the total number of modes. Our experimental findings for the relative numbers of modes in each energy range for each of the crystalline samples are shown in table 1 and it will be seen that this prediction is confirmed for pure Se. It should be noted that these values are only correct if the system has no higher-energy modes. It was not possible to access this region using IN6 without heating the sample which, since the melting point is about 400 K, would have destroyed it. We therefore measured the scattering from the crystalline samples at 10 K on IN4 in the down-scattering mode, using an incident energy of 115 meV. No excitations beyond 35 meV were detected. These data involve much higher values of  $Q$  than those contained in the IN6 data and they therefore lie closer to the conditions which are necessary for



**Figure 3.** The density of states  $g(\omega)$  plotted against the energy,  $\omega$ , for the four crystalline Se-Te samples and for the Se<sub>100</sub> sample. Some points are omitted for clarity. The inset shows on a larger scale the low-energy peak for Se<sub>100</sub> which is divided into three sub-peaks. This was also observed for the other specimens. Note also the wing on the low-energy side of the 30 meV peak for Se<sub>79</sub>. — : Se. □: Se<sub>96.85</sub>. ■: Se<sub>93.6</sub>. ○: Se<sub>86.6</sub>. ●: Se<sub>79</sub>.



**Figure 4.** The density of states of crystalline trigonal Se as calculated by Etchepare *et al* [8]. The dashed line shows our experimental results for crystalline Se<sub>100</sub> (from figure 3).

the incoherent approximation. In this more accurate limit the envelope of excitations which we had obtained from the IN6 data was confirmed although the statistics are much poorer on IN4.

**Table 1.** The ratios of the density of states in the crystalline Se-Te alloys after a scaling to the total density of states between 0 and 50 meV for each alloy.

	Torsional	Librational and bending	Stretching
Se	0.245	0.321	0.323
Se <sub>96.85</sub> Te <sub>3.15</sub>	0.223	0.297	0.355
Se <sub>93.6</sub> Te <sub>6.4</sub>	0.253	0.313	0.302
Se <sub>86.6</sub> Te <sub>13.4</sub>	0.251	0.309	0.306
Se <sub>79</sub> Te <sub>21</sub>	0.207	0.286	0.379

## 6. Interpretation of the results

The trigonal structure of Se is shown in figure 1 and the density of states, as calculated by Etchepare *et al* [8], is compared in figure 4 with the experimental curve which we obtained for crystalline Se. The density of states which we have derived from the IN6 data for the crystalline Se<sub>x</sub>Te<sub>1-x</sub> samples all have the same form as that of figure 4.

In the amorphous samples the overall envelope of  $g(\omega)$  is very similar to that of crystalline Se, as was observed by Gompf [9]. Thus the basic structural units in the amorphous and crystalline materials must be very similar. The lowest-energy section is thought to be due to torsional oscillations in the helical chain structure, whilst the (double) peak at 14 meV is caused by librational/bending modes. The isolated high-energy peak at 28.5 meV is associated with localized stretching modes.

### 7. The high-energy peak

Our measurements show that the basic Se spectrum is unchanged by the addition of Te atoms, but as the Te content of the samples is increased the high-energy peak broadens to lower energies due to the growth of another peak at about 24.7 meV. This is clearly shown for crystalline  $\text{Se}_{79}$ . The new peak is presumably due to the influence of the more massive Te ions which (if the force constants are unchanged) would tend to reduce the vibrational frequency. If we assume that the 28.5 meV peak is due to Se-Se interactions and that the lower one is mainly due to Te-Se interactions then their energies should be in the ratio (average mass)<sup>-1/2</sup>, i.e. 0.87. This is in good agreement with the ratio of the energies which is 0.87.

In the amorphous samples the high-energy peak is pushed to a higher energy, 30.5 meV, presumably because the localized excitations are more inhibited (i.e. a larger force constant) in the amorphous environment. The separation into two peaks is again clearly seen in the  $\text{Se}_{79}$  sample, the lower peak being at 27.5 meV. This gives a ratio for the two energies of 0.9 which is very similar to that for the crystalline samples (0.87).

### 8. The low-energy density of states

The low-energy collective excitations, which are thought to be torsional modes, have a continuous density of states. However, due to the finite instrumental resolution and the presence of the large elastic peak, the lowest energy for which we can have confidence is about 0.5 meV. The distribution rises to a peak centred at about 5.6 and 6.5 meV for the amorphous and crystalline specimens respectively. This peak has a FWHM of about 5 meV. It has about the same height for all samples. For the crystalline specimens the peak is resolved into three small sub-peaks which are spaced about 1.5 meV apart—see inset to figure 3.

A log-log plot of the low-energy data for all crystalline samples shows that between 0.6 and 3 meV it has an energy dependence of  $\omega^n$  with  $n = 1.5 \pm 0.05$ . This should be compared with the  $\omega^{2 \pm 0.05}$  dependence which we find at the lowest energies for the amorphous samples. This difference in behaviour is illustrated in figure 5 for both crystalline and amorphous  $\text{Se}_{79}$ . Note that for the amorphous sample there is also a short region where the energy dependence is greater than quadratic.

We have calculated the specific heat for trigonal Se using the density of states derived from the neutron data from 0.5 to 37.5 meV and assuming a  $\omega^2$  dependence for  $g(\omega)$  below 0.5 meV. The results of this calculation are shown in figure 6 together with a set of recommended experimental values [11]. For ease of comparison our results are normalized with the experimental value of the specific heat at 50 K. The agreement over most of the experimental range is very good indeed and it gives us

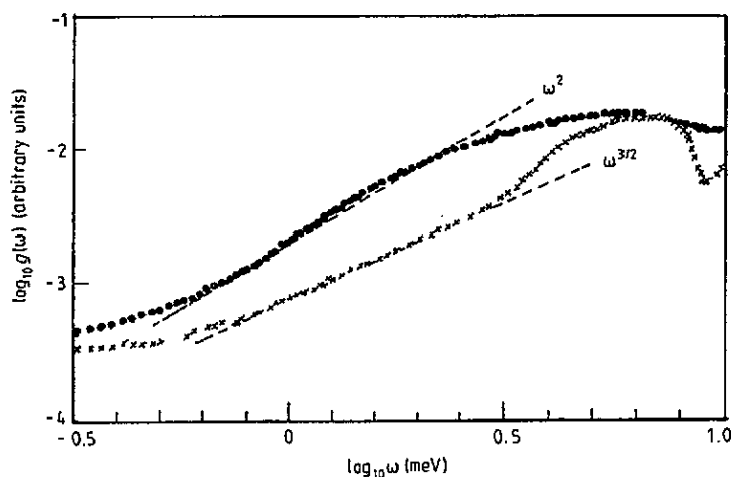


Figure 5. The density of states versus energy curves plotted on a logarithmic scale for crystalline (x) and amorphous (●) Se<sub>79</sub>. Note (i) that the crystalline material has an extensive region where the energy dependence is  $\omega^{1.5}$  and (ii) the curve for the amorphous sample rises above  $\omega^2$  over part of the range. The points at low energies reflect the influence of the elastic peak and they should be discounted.

confidence that, in spite of the uncertainty for the validity of the incoherent analysis, the form of the density of states must be more or less correct.

Genensky and Newell [12] calculated the density of states using a chain polymer crystal model. They predicted that the density of states from the lowest energies should pass through regions varying, in order of increasing energy, as  $\omega^2$ ,  $\omega^{1.5}$ ,  $\omega^{0.5}$  and  $\omega^{-0.5}$ , although it is doubtful whether these last two regions exist, except perhaps over a very short energy range. Gompf [9] found for trigonal Se an  $\omega^2$ -dependence from his neutron scattering measurements. There is however a trend at the lower end (around 1.5 meV) of his published data towards an energy dependence which has a power less than 2. The  $\omega^{1.5}$  dependence of  $g(\omega)$  found in our experiments for the crystalline samples and the known structural form of trigonal Se (figure 1) leads us to believe that the Se and the SeTe samples which we have studied can be considered to be behaving as chain polymers.

This leads to the conclusion that the intra-chain bonding strengths are more important than those for inter-chain bonding in this low-energy regime. For the amorphous samples it can be envisaged that the disorder in the system produces entanglement of the chains and hence a cross-linking, which will result in some three-dimensional character and hence to a  $g(\omega)$  which varies as  $\omega^2$  over a more extended energy range.

As noted previously, above about 1.2 meV the amorphous samples show a breakaway to an energy dependence which is stronger than  $\omega^2$  over a limited energy region. This seems to be a general characteristic of the density of states of amorphous materials [1, 2] and would appear to occur irrespective of the behaviour of the crystalline material, although this has not been observed in some recent experiments on B<sub>2</sub>O<sub>3</sub> [15].

## 9. Experiments to measure $S(Q)$

The spectrometer LAD is a time-of-flight spectrometer specifically designed for diffraction measurements of liquid and amorphous materials [17]. The experimental  $S(Q)$  is obtained by assuming that all the neutrons in a 'white beam' incident on the sample are elastically scattered at the sample. The summation over all energies of



the neutrons arriving at a particular detector bank then effectively measures

$$\int S(Q, \omega) d\omega \simeq S(Q). \quad (4)$$

This can be derived theoretically from correlation function considerations and is termed the static approximation. Corrections need to be made to take account of the fact that (i) the scattering is not perfectly elastic (Placzek corrections) and (ii) that the sample scattering cross section is energy-dependent.

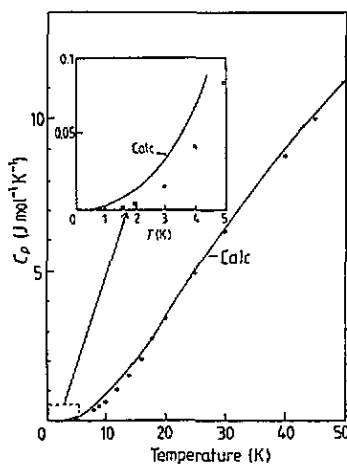


Figure 6. The specific heat of crystalline Se as a function of temperature. The solid line shows the values as calculated from the density of states which was derived from the neutron scattering experiments. The circles show the experimental values as given by [9]. The two sets of data are normalized at 50 K.

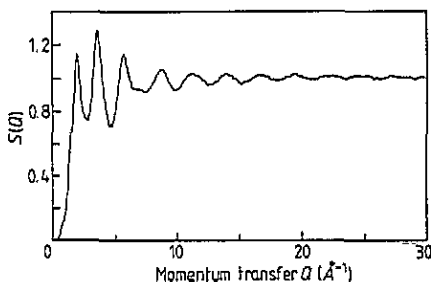


Figure 7. The scattering function  $S(Q)$  as a function of momentum transfer for amorphous  $\text{Se}_{86.6}$  as derived from data obtained on LAD. The results for the other amorphous samples are very similar.

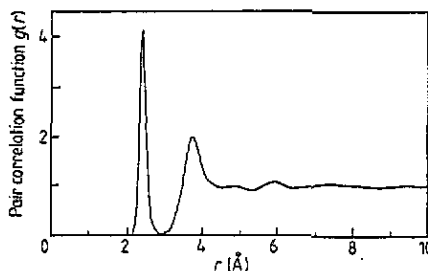


Figure 8. The pair correlation function,  $g(r)$ , as a function of spacing for amorphous  $\text{Se}_{86.6}$  derived from the data shown in figure 7. The results for the other amorphous samples are very similar. The data processing program [16] forces  $g(r)$  to be zero for small values of  $r$ .

LAD enables measurements to be made out to  $Q = 50 \text{ \AA}^{-1}$  which is a great advantage when performing a Fourier transform of the data in  $Q$ -space to obtain the pair-distribution function  $g(r)$ . Figures 7 and 8 show plots of  $S(Q)$  versus  $Q$  and of  $g(r)$  versus  $r$  for amorphous  $\text{Se}_{86.6}$ . (Those for  $\text{Se}_{96.85}$  and  $\text{Se}_{79}$  are very similar). From the  $g(r)$  data it is possible to extract the following information. The nearest-neighbour distance,  $r_1$ , occurs at  $2.4 \text{ \AA}$ . The sharpness of the peaks in  $g(r)$  is an

indication of the amount of order present. It can be seen, therefore, that the system does not have much disorder associated with the nearest-neighbour distance. The second peak at  $r_2$ , however, is broader and hence it appears, not surprisingly, that the system is more disordered beyond nearest neighbours. Its position is  $r_2 = 3.7 \text{ \AA}$ . The bond angle is given by  $2\sin^{-1}(r_1/2r_2)$  and the commencement of the disorder observed in the second-nearest-neighbour regime can be interpreted in terms of disorder in the bond angle. The average bond angle that was obtained was  $100.86^\circ$  which agrees well with that for crystalline Se. At distances greater than  $3.7 \text{ \AA}$  the particle correlation density reveals peaks which are just distinguishable at about  $4.9 \text{ \AA}$  and  $5.9 \text{ \AA}$ . However, at these distances it can be seen that these are very broad and they merge into one another indicating that there is a large amount of disorder in the system for  $r > 4 \text{ \AA}$ .

The bond angle and the first- and second-neighbour distances are the same as for trigonal Se. Hence, these results add further confirmation to our conclusions from the vibrational density of states measurements, that the amorphous samples have a local structure very similar to that of trigonal Se (and that the addition up to 21 at.% Te does not alter the basic properties of this local order).

Another interesting feature which has arisen from the  $S(Q)$  study is that the co-ordination number obtained is  $1.7 \pm 0.1$  for both  $\text{Se}_{96.85}$  and  $\text{Se}_{86.6}$ . Since the co-ordination number of both Se and Te is 2, this would suggest that in these amorphous systems there are voids and also 'dangling bonds'.

## 10. Summary of results

(i) The crystalline SeTe samples showed chain-polymer behaviour in their vibrational properties at low energies.

(ii) The observed density of states in the crystalline samples is very similar to the model predictions for trigonal Se for all concentrations of Te except  $\text{Se}_{79}\text{Te}_{21}$  and the ratio of the number of modes in each energy region is also in agreement with this model.

(iii) The density of states in the amorphous samples has a basic envelope very similar to that observed in the corresponding crystalline sample. Hence it would be expected that the local order is the same as for the crystalline samples. However,  $g(\omega)$  is proportional to  $\omega^2$  (for  $\omega < 1.2 \text{ meV}$ ), in agreement with the observations in other amorphous materials, although the crystalline counterpart has a density of states which suggests chain-polymer behaviour in this energy regime.

(iv) The values of the nearest-neighbour and next-nearest-neighbour distances and the value of the dihedral angle obtained from  $S(Q)$  measurements in the amorphous samples confirm the conclusions in (iii) that the local order is very similar to that of crystalline Se.

(v) The measured co-ordination number of 1.7 for the amorphous samples is lower than would be expected from calculations based on the fact that Se and Te are both doubly co-ordinated atoms.

## 11. Conclusions

The field of study of amorphous materials is becoming more and more 'consistent' in that all amorphous materials show a short low-energy range where  $g(\omega)$  varies as

$\omega^2$  (where  $\omega$  is, typically, about 1 meV) irrespective of the nature of the crystalline counterpart. Beyond this is a region where the energy dependence increases in the same way as was observed in epoxy resins [2] and which was there ascribed to fractal behaviour. We have obtained a very complete set of results for the Se-Te systems which are in good agreement with previous measurements on Se, of specific heat capacity measurements and chain polymer behaviour for the crystalline samples.

### Acknowledgments

We should like to thank A M Glazer, A C Hannon, W S Howells, R L McGreevy, A Murani, A Soper and J D Wicks for their valuable advice and assistance.

### References

- [1] Buchenau U, Prager M, Nücker N, Dianoux A J, Ahmad N and Phillips W A 1986 *Phys. Rev. B* **34** 5665
- [2] Rosenberg H M 1984 *Phys. Rev. Lett.* **54** 704
- [3] Aharony A, Alexander S, Entin-Wohlman O and Orbach R 1985 *Phys. Rev. B* **31** 2565
- [4] Carini G, Cutroni M, Dianoux A J, Galli G, Needham L M and Rosenberg H M 1990 *J. Non-cryst. Solids* **117/118** 140
- [5] Zingaro R A and Cooper W C 1974 *Selenium* (New York: Van Nostrand) p 109
- [6] Dianoux A J 1989 *Phil. Mag.* **59** 17
- [7] Buchenau U 1985 *Z. Phys. B* **58** 181
- [8] Etchepare J, Kaplan P and Merian M 1975 *Int. Conf. on Lattice Dynamics (Paris)* 60
- [9] Gompf F 1981 *J. Phys. Chem. Solids* **42** 539
- [10] Kamitakahara W A, Capelletti R L, Boolchand P, Halpap B, Gompf F, Neumann D A and Mutka H 1991 *Phys. Rev. B* **44** 94
- [11] Gaur U, Shu H-C, Mehta A and Wunderlich B 1981 *J. Phys. Chem. Ref. Data* **10** 89
- [12] Genensky S M and Newell G F 1957 *J. Chem. Phys.* **26** 486
- [13] Needham L M 1989 *D. Phil. Thesis* Oxford University p 70
- [14] Needham L M to be published
- [15] Hannon A C, Sinclair R N and Wright A C 1992 *The Physics of Non-crystalline Solids* ed L D Pye, W C La Course and H J Stevens (London: Taylor and Francis) p 67
- [16] Soper A K 1990 *Neutron Scattering Data Analysis (Inst. Phys. Conf. Series 107)* p 57
- [17] Howells W S 1980 *Rutherford-Appleton Laboratory Report* RAL-80-017