

Structural Inhomogeneity in Semiconducting Melts((A)Amorphous Alloys)

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Structural Inhomogeneity in Semiconducting Melts*

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Synopsis

The structural features of liquid metals and alloys have been reviewed using the concept of structural inhomogeneity in melts. Some physico-chemical properties of semiconducting melts such as viscosity, electrical conductivity, magnetic susceptibility and specific heats appear to indicate a certain chemical ordering, different from the usual short range ordering found in various liquid metals and alloys. Characteristic properties of chalcogen bearing alloy melts or alkali-polyvalent alloy melts have also been presented in terms of the structural information.

1. Introduction

Many single crystals such as Si, GaAs and oxides are known to be synthesized in the industrial scale using several techniques such as Czochralski method. This Czochralski process is founded on the crystal growth of these materials from the respective pure molten substances.

Experimental efforts have been devoted, in the past, to the mechanisms of these crystal growth from the melts and the results are discussed mostly from the view point of the relevance with physico-chemical properties of melts as well as the resultant crystals [1-3]. There seems to be few investigations on the structure of melts in this field. It is evident, however, that the fundamental information of the

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melt structures is necessary for up-grading the quality and scaling up the single crystal size with respect to the crystal growth process.

On the other hand, it has been reported that there exists a certain local ordering whose size is larger than the usual short range ordering in some liquid alloys, especially systems including chalcogens or alkaline and polyvalent metals [4,5]. These systems indicate a peculiar behavior in their electric and thermodynamic properties at a certain concentration near the melting point [6]. Therefore, it is reasonable to anticipate that the short range ordering in the liquid state is related to the crystallization process. However, this relatively new field has not been explored yet.

The purpose of this paper is to review the structural inhomogeneity in melts with special reference to the crystal growth process.

2. Principles of structural analysis of disordered systems

The structure of disordered systems such as liquid and glasses can only be quantitatively described in terms of the radial distribution function (RDF), indicating the average probability of finding another atom in a specified volume from an origin atom as a function of radial distance. The RDF provides spherically averaged information of the atomic correlations. The RDF can be determined from diffraction data with X-rays and neutrons and the relation between measured diffraction data and the RDF has been described in detail [5,7].

The essential equations are as follows for convenience of discussion in this article.

$$\rho(r) = \sum_{i=1}^n \sum_{j=1}^n c_i f_i f_j \rho_{ij}(r) / \langle f \rangle^2 \quad (1)$$

$$\langle f^2 \rangle = \sum_{i=1}^n c_i f_i^2, \quad \langle f \rangle^2 = \left(\sum_{i=1}^n c_i f_i \right)^2 \quad (2)$$

$$\begin{aligned} S(Q) &= [I_a^{\text{coh}}(Q) - (\langle f^2 \rangle - \langle f \rangle^2)] / \langle f \rangle^2 \\ &= \sum_i \sum_j c_i c_j \frac{f_i f_j}{\langle f \rangle^2} S_{ij}(Q) \end{aligned} \quad (3)$$

$$S_{ij}(Q) = 1 + \int_0^{\infty} 4\pi r^2 \rho \cdot [g_{ij}(r) - 1] \frac{\sin(Q \cdot r)}{Qr} dr \quad (4)$$

$$G(r) = 4\pi r [\rho \cdot g(r) - \rho_0] \\ = \frac{2}{\pi} \int_0^\infty Q [S(Q) - 1] \sin(Q \cdot r) dQ \quad (5)$$

where $\rho(r)$ is the average radial density function of multicomponent disordered system, $\rho_{ij}(r)$ the partial radial density function, c_i the atomic fraction of i-type atom, f_i the atomic scattering factor for atom i, $S(Q)$ the total structure factor, $I_a(Q)$ the coherent X-ray scattering intensity per atom, $S_{ij}(Q)$ the partial structure factor, $G(r)$ the reduced radial distribution function and $g_{ij}(r) = \rho_{ij}(r) / c_i \rho_0$ the partial pair distribution function.

Equation (1) implies that the average radial density function $\rho(r)$ of multicomponent disordered systems can be given by the summation of the partial radial density function of $\rho_{ij}(r)$ with a weighting factor expressed by c_i and f_i . Eqs. (3) and (5) provide the relation between the atomic scale structure and measured intensity data for disordered system including more than two kinds of atoms. The conventional diffraction experiments for multicomponent disordered systems only allow to obtain the total structure factor. However, the knowledge of the structural functions of individual pairs such as $\rho_{ij}(r)$ is required for discussing various characteristic properties of multicomponent disordered systems of interest at a microscopic level. The individual partial structure factors may be estimated by making available three independent scattering experiments for which the weighting factors are varied. Some methods to obtain the individual partial structure factors have already been summarized in ref. [8].

3. A Brief background of the features suggesting the structural inhomogeneity in the melts.

The basic profile of the experimental structure factor of most liquid metals is represented by the hard sphere solution of the Percus-Yevick equation so long as the hard-sphere diameter or the packing density (η) is suitably selected [9]. The hard sphere structure factor shows the following features:

- 1) The first peak of the structure factor is symmetrical.
- 2) At temperature just above the melting point, the best agreement with experimental data is found with a packing density $\eta = 0.45$.
- 3) The ratio (Q_2/Q_1) of the position of the second peak (Q_2) to that of the first peak (Q_1) is about 1.86.

Thus, the deviation from the simple mixture approximated by the

hard-sphere structure factor indicates one of the useful informations on the structural inhomogeneity in liquid metals. Based on the systematic structural investigations for liquid metals [5], all of the metallic elements are classified into one of the following three categories.

- a) The structure factor satisfies the features of the hard-sphere structure factor above-mentioned (Al, et. al.).
- b) The structure factor indicates an asymmetry of the first peak (Zn, Cd, Hg).
- c) The structure factor indicates a small hump on the high angle side of the first peak (Sn, Ga, Sb, Si, Ge).

It may be worth mentioning that more than 85 % of the metallic elements, including transition and rare earth metals have the structure factors similar to those of class (a). This implies that a random distribution of the hard-spheres of appropriate diameter is a basis for atomic distribution in all liquid metals. The increase in the freedom of atomic configuration contributes to the construction of universal short-range order.

The elements which anisotropic bonding in the crystalline state show a deviation from the hard sphere structure factors and this is related, in a crude approximations, to a crystal structure. Deviation of this type has been found to decrease with an elevation of temperature. The liquid structures of (c) type show such behavior and Ga, Si, Ge, Sb and Bi is included in this group. For convenience, the structure factors of Si and Ge are given in Fig.1 [10].

The deviation from the hard-sphere structure factor in the (b) type liquids has been discussed in terms of the electron theory of metals [11]. This is out of scope in this article and not repeated here.

In contrast to these structural features of the metallic elements, particular profiles are obtained in semiconducting elements such as Se and Te. The structure factor for liquid Se by X-ray diffraction is shown in Fig.2 together with the structure factor of liquid Na for comparison [12]. The following observations may be stressed:

- 1) The heights of the peaks in the structure factor of liquid Se are almost the same with the first peak considerably lower than the second one.
- 2) The height of the first peak decreases with an increase in temperature, while the second and third peaks are relatively insensitive to temperature.
- 3) The oscillation in the structure factor for liquid Se is observed

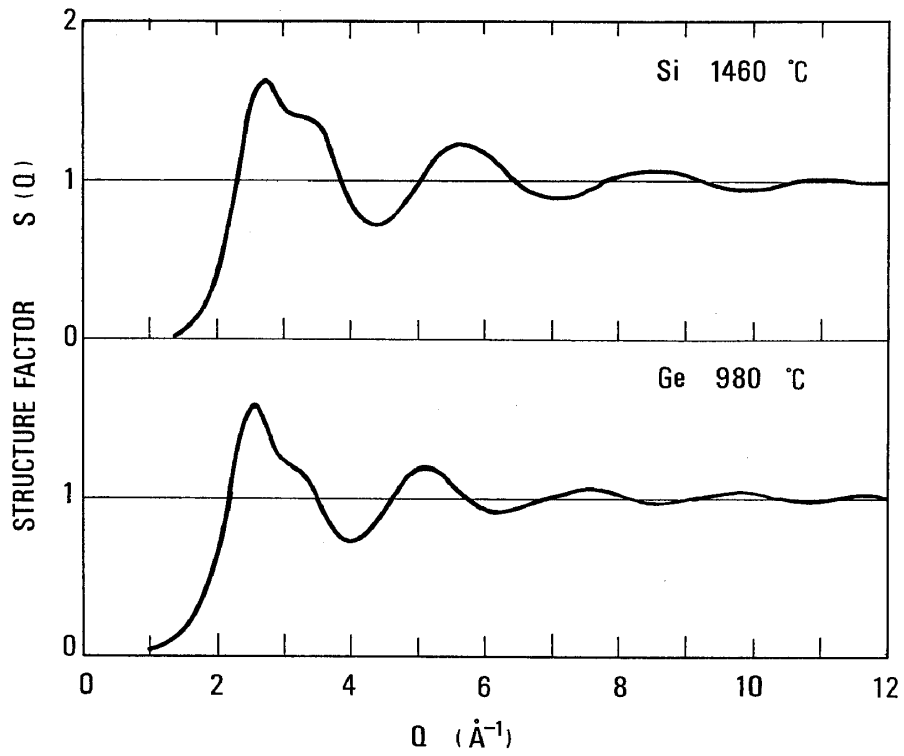


Fig. 1 Structure factors of liquid Si and Ge near the melting point [10].

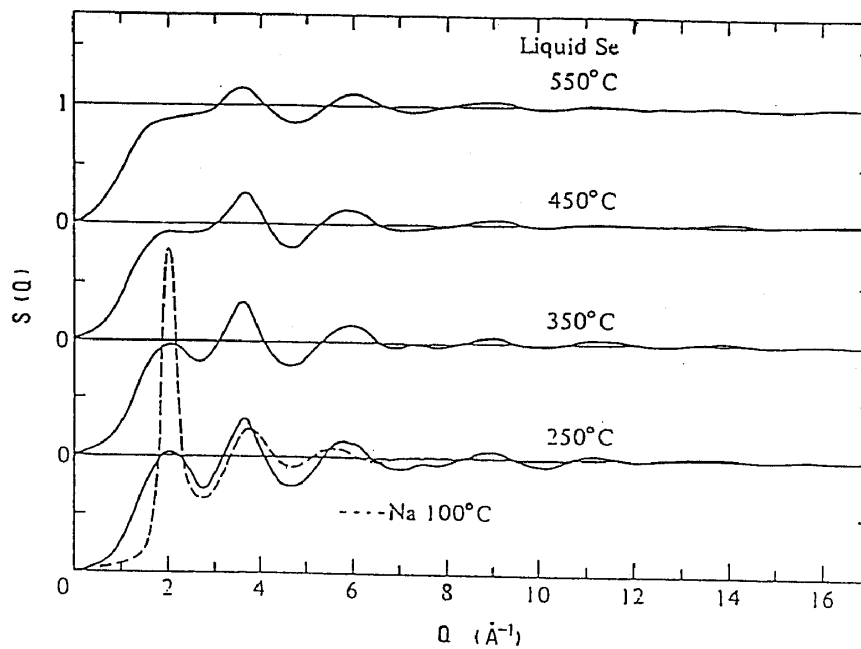


Fig. 2 Temperature dependence of the structure factor of liquid Se together with that of liquid Na [12].

in the higher Q region than for liquid Na.

These particular structural features imply that a considerable fraction of a certain local ordering having definite binding and angles related to the crystal structure remains in the liquid state [12]. Similar behaviors of the structure factor for liquid Se are also recognized in liquid Te, although there are differences in detail [13]. Although the basic features of most metallic melts could be explained, more or less, by the structural homogeneity, we also frequently find the experimental results suggesting the structural inhomogeneity exemplified by the results of liquid Se. Such behavior, different from simple mixture, is quite likely in the melts including chalcogens, because an anisotropic bonding in these semiconducting elements contributes to their melt structure. This may also be true, when the melts consist of more than two kinds of atoms having different physico-chemical properties such as size, charge number and electronegativity.

4. Anomalous behavior of physico-chemical properties suggesting the structural inhomogeneity in the melts.

Viscosity, electrical conductivity, magnetic susceptibility and specific heats of semiconducting melts have been widely investigated because these physico-chemical properties are sensitive to change in the structure and in the nature of the chemical binding.

Of these, the viscosity is known to be one of the most structure-sensitive properties. Figure 3 shows temperature dependence of the kinematic viscosity of liquid Si and Ge [14]. At higher temperature, the logarithmic kinematic viscosity linearly depends on the inverse of the absolute temperature in both cases. At lower temperature, however, such linearity is not held and the deviation from the linearity is quite obvious at temperature 50-100 K above the melting point of the respective elements. This deviation has been explained by the trace of a certain local ordering of the crystal structure, which disappears completely at the high temperature.

Figure 4 shows the temperature dependence of the viscosity of molten $\text{Ga}_{1-x}\text{As}_x$ ($0 \leq x \leq 0.53$) [15]. One of the most notable observation in Fig.5 is the drastic increase in the viscosity of molten $\text{Ga}_{0.5}\text{As}_{0.5}$ near the melting point. If the viscous flow of the melt is a single activation energy process, the viscosity of the melt should vary as Arrhenius' equation as a function of the temperature, as observed for simple metallic melts. However, the temperature dependence of the

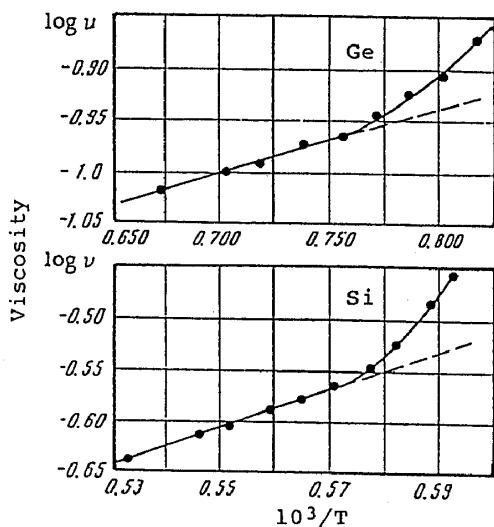


Fig. 3 Temperature dependence of the kinematic viscosity of molten Si and Ge [14].

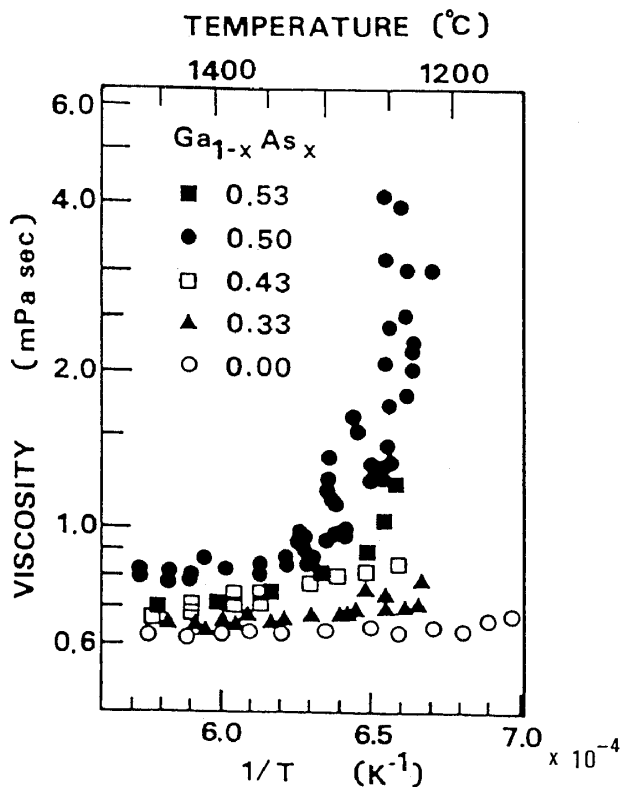


Fig. 4 Temperature dependence of the viscosity for molten $\text{Ga}_{1-x}\text{As}_x$ ($0.0 \leq x \leq 0.53$) [15].

viscosity of $\text{Ga}_{1-x}\text{As}_x$ ($x=0.5$) indicates a considerable deviation from the Arrhenius relation, as shown in Fig.4. This data again suggests that $\text{Ga}_{0.5}\text{As}_{0.5}$ melt likely involves a certain local ordering whose size is larger than that of short range ordering in the usual melts.

The electrical conductivity is governed by the product of the carrier density and the carrier mobility. Structural changes have an influence on the carrier mobility and thus also contribute to the change in the electrical conductivity. The electrical conductivity of Si and Ge increases gradually with an elevation of temperature, then indicating a sudden increase at the melting point as shown in Fig.5 [16]. The electrical conductivity of Si and Ge in the liquid state is of the order of the conductivity of liquid metals such as Sn or Hg ($10^4 \Omega^{-1} \text{cm}^{-1}$), which indicates that these substances become metallic on melting. It is rather the present intention to suggest the wide variation of the experimental points near the melting point, compared to those at higher temperature. The present authors maintain the view that the structural inhomogeneity is quite feasible in the close vicinity of the melting point in both Si and Ge melts.

The logarithmic electrical conductivity of Ga_2Te_3 is given in Fig.6 [17]. The electrical conductivity of this compound is known to

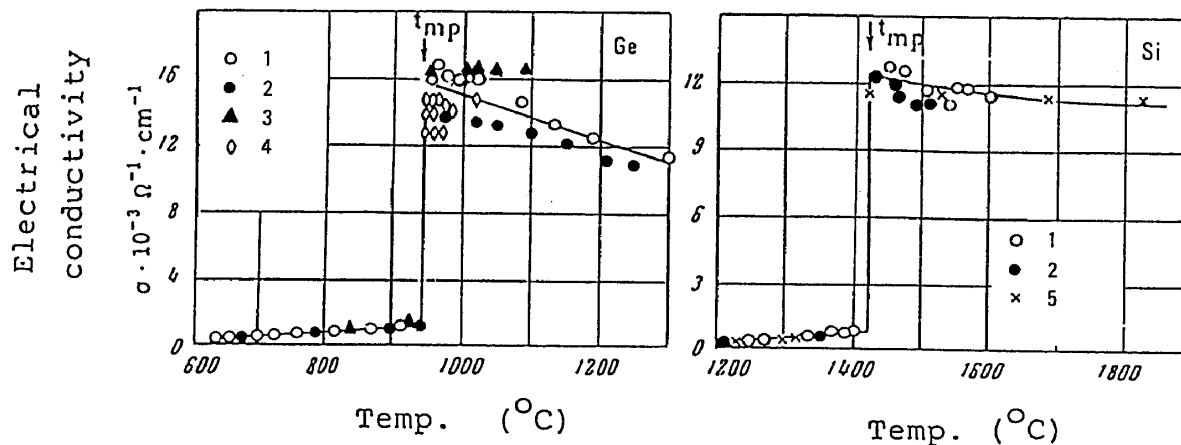


Fig. 5 Temperature dependence of the electrical conductivity of Si and Ge [16]. The numbers of the notation denote the respective results obtained from each investigators.

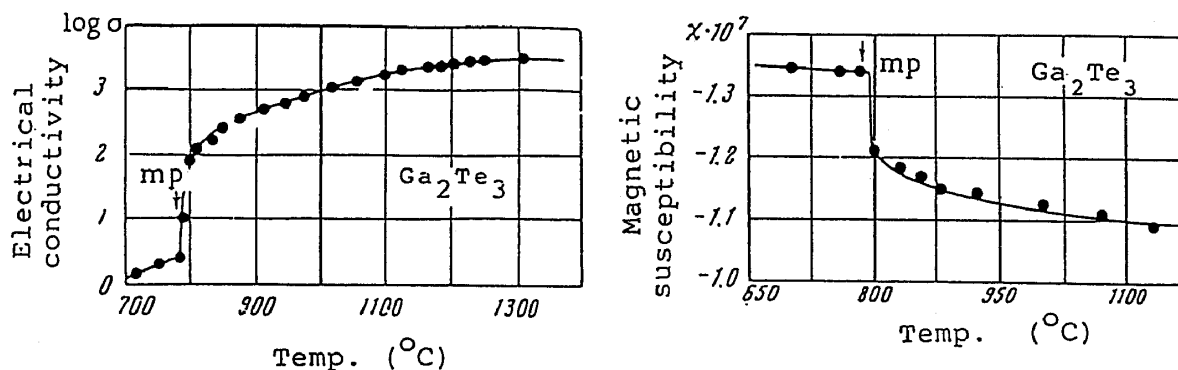


Fig. 6 Temperature dependence of the electrical conductivity and the magnetic susceptibility of Ga_2Te_3 [17].

increase exponentially with temperature in the solid state. Whereas it rises suddenly at the melting point and continues to increase rapidly in a certain temperature range above the melting point, although further heating produces the gradual increase of the electrical conductivity of Ga_2Te_3 in the molten state. A similar behavior has been reported in other A B compounds such as In_2Te_3 in the molten state [6].

The temperature dependence of the electrical conductivity of these compounds indicates the semiconductor-semiconductor transition occurs on melting. The large jump in the electrical conductivity of Ga_2Te_3 compound at the melting point and the strong rise immediately above it are due to an increase in the carrier density and a probable increase in the mobility. However, the covalent bonding nature of the

crystalline Ga_2Te_3 compound still remains in the molten state and it is gradually destroyed by further heating.

The temperature dependence of magnetic susceptibility of Ga_2Te_3 is also given in Fig.6 [17]. It is easily seen from this figure that the magnetic susceptibility drops suddenly at the melting point and continues to show the relatively large decrease in a certain temperature range close to the melting point. At much higher temperature, the susceptibility ceases to depend on temperature. This is one of the clear observations suggesting the existence of structural inhomogeneity in the melts. This is also consistent with the measurements of thermodynamic properties. For example Fig. 7 shows the temperature dependence of measured specific heats of Ga-Te alloys for various Te concentration [18]. As shown in the figure a large temperature variation is found in the alloys with 50 and 60 at.% Te and in these concentration region, a large deviation from the ideal linear interpolation has been clearly detected. From these experimental data coupled with thermodynamic consideration, the intermediate state in the dissociation process from Ga_2Te_3 to $(\text{Ga}_2\text{Te} + 2\text{Te})$ is suggested in this semiconducting melt. It has also been ascertained that the large specific heat around the composition of Ga_2Te_3 and its temperature

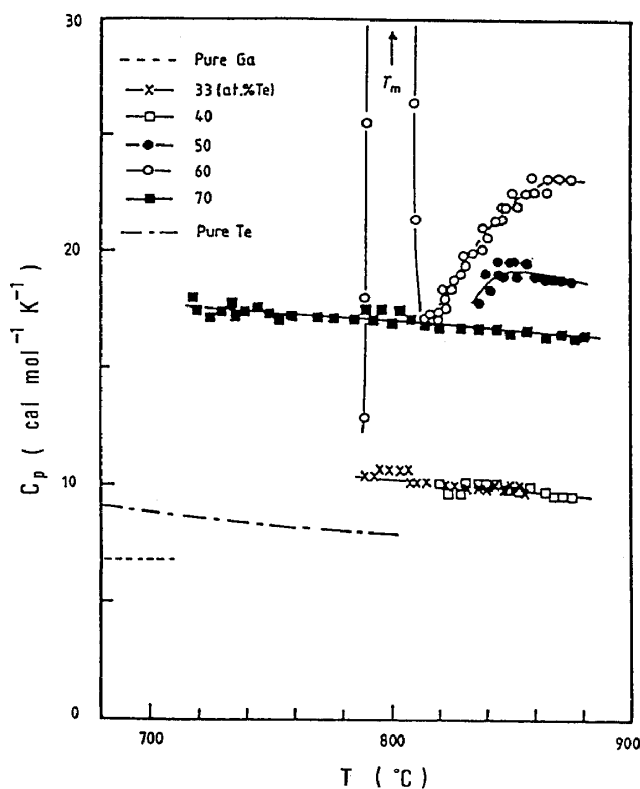


Fig. 7 The specific heats of liquid Ga-Te alloys as a function of temperature [18].

dependence are explained in terms of the partial dissociation of the compound such as local ordering with increasing temperature. Some more details about a local ordering will be given in the next chapter.

5. Some selected topics on the structural homogeneity in the melts by diffraction method

Liquid structure may be characterized by the structural homogeneity where the distribution of constituent elements around the respective component element shows no deviation from the average value. In fact, we can frequently find the experimental results suggesting that a completely random mixing of the hard spheres with different size is not obtained in the melts. No unique definition is available for describing the structural inhomogeneity in the melts. Nevertheless, the concentration-concentration structure factor $S_{CC}(0)$ in the long wavelength limit ($Q \rightarrow 0$) in the Bhatia-Thornton form is the most useful method to estimate the deviation from the random mixing in disordered systems [19].

$S_{CC}(0)$ give a physical significance indicating the mean square fluctuation in the concentration in liquids of interest. According to Bhatia-Thornton [19] the concentration-concentration structure factor in the long wave length limit, $S_{CC}(0)$, is given by the following equation providing a direct link with the thermodynamic activity data of the desired disordered systems.

$$S_{CC}(0) = (1 - c_j) \left[\left(\frac{\partial \ln a_j}{\partial c_j} \right) \right]_{T,P}^{-1} \quad (6)$$

where a_j and c_j are the thermodynamic activity and the atomic fraction of j -component, respectively.

$$S_{CC}(0) = c_j(1 - c_j) \quad (7)$$

Figure 8 shows the function of $S_{CC}(0)$ for some liquid alloys as a function of concentration estimated from measured thermodynamic activity data [6]. A negative deviation from ideal mixing is pronounced at a certain composition such as A_4B in liquid Na-Sn [20], Na-Pb [21] alloys and AB_2 in liquid Bi-Mg [22] alloys. This indicates qualitatively the formation of chemical complexes in the melts near a certain composition. This is consistent with suggestions from the measurements of physico-chemical properties such as heat of mixing, electrical resistivity and magnetic susceptibility of several liquid alloys. It

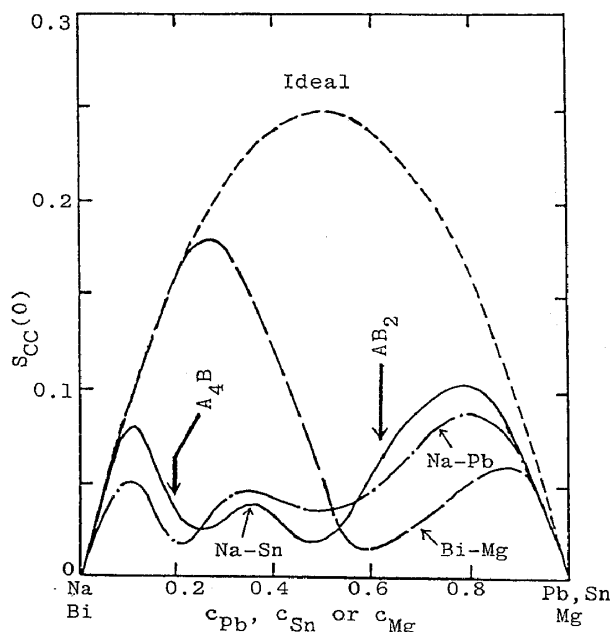


Fig. 8 Concentration-concentration fluctuation function of some liquid binary alloys suggesting compound forming [8].

may be added that a large positive deviation from ideal mixing indicating phase separation is also detected at the composition near AB_3 in liquid Ga-Na [23] and Bi-Zn [24] alloys.

In general, the presence of a prepeak in the structure factor of disordered systems is qualitatively interpreted as indicating compound formation, because, if species having definite size exist in disordered system the inter-relation between such species could be observed in addition to the usual correlations of the components [5]. As for the theoretical approach of these compound forming melts, Copestake et.al.[25,26] proposed an ionic model with hard-sphere Yukawa interaction for the ordering potential and applied it to the liquid Li_4Pb alloy. Hafner et.al.[27] has extended this approach to liquid Na-Pb alloys.

An example of such compound forming liquids is given below using the results of liquid Na-Pb alloys. Figure 9 shows the total structure factors of four alkaline metal-Pb alloys with the equi-atomic composition measured at temperatures above their melting points [28]. All structure factors of these alkaline metal-Pb alloys indicate a characteristic sharp prepeak at low Q region of the first peak except for NaPb. It has also been reported that a shift in stoichiometry occurs in NaPb alloys, where the departure from the equi-atomic composition leads to an increase in the height of the prepeak with the maximum

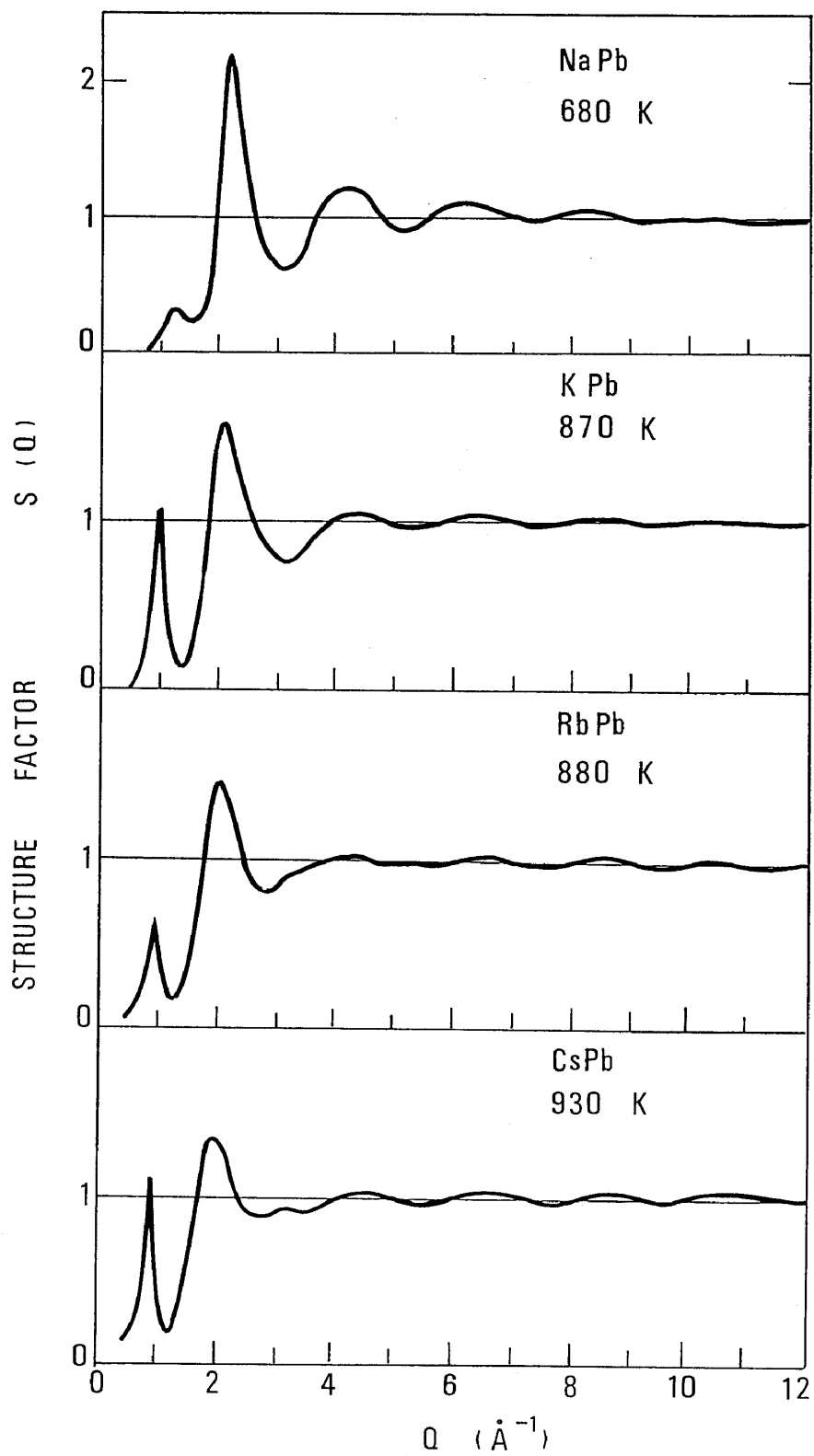


Fig. 9 Total structure factors of four liquid alkaline metal-Pb alloys measured by neutron diffraction [28].

reached at Na_4Pb [29].

Based on the measurements of thermodynamic and electric properties of liquid Na-Pb alloys, the most probable configuration of the compound forming unit structure may be a tetrahedron with Pb at the center as schematically inserted in Fig.10 and the distance between Pb and Na atoms is estimated to be 0.33 nm from the distribution function data of 80 at.% Na alloy and these alloys are considered as a ternary mixture composed of the atomic association of Na_4Pb and its dissociated atoms of Na and Pb. In this model, the chemical interaction between unlike atom pairs is somewhat squeezed in the compound formation of " Na_4Pb molecule" and the measured structure factors can be estimated using the model structure factors of ternary hard spheres proposed by Hoshino [30]. Figure 10 shows the comparison between calculation and experiment in the structure factor of liquid $\text{Na}_{80}\text{Pb}_{20}$ [8]. The overall agreement between these two structure factors can be recognized. The oscillating profiles in the large Q region beyond 60 nm^{-1} are also quite in phase with measured structure factor. These facts suggest that the simple hard sphere mixture model basically work well by reproducing the characteristic structural features of compound forming melts such as liquid Na-Pb alloys.

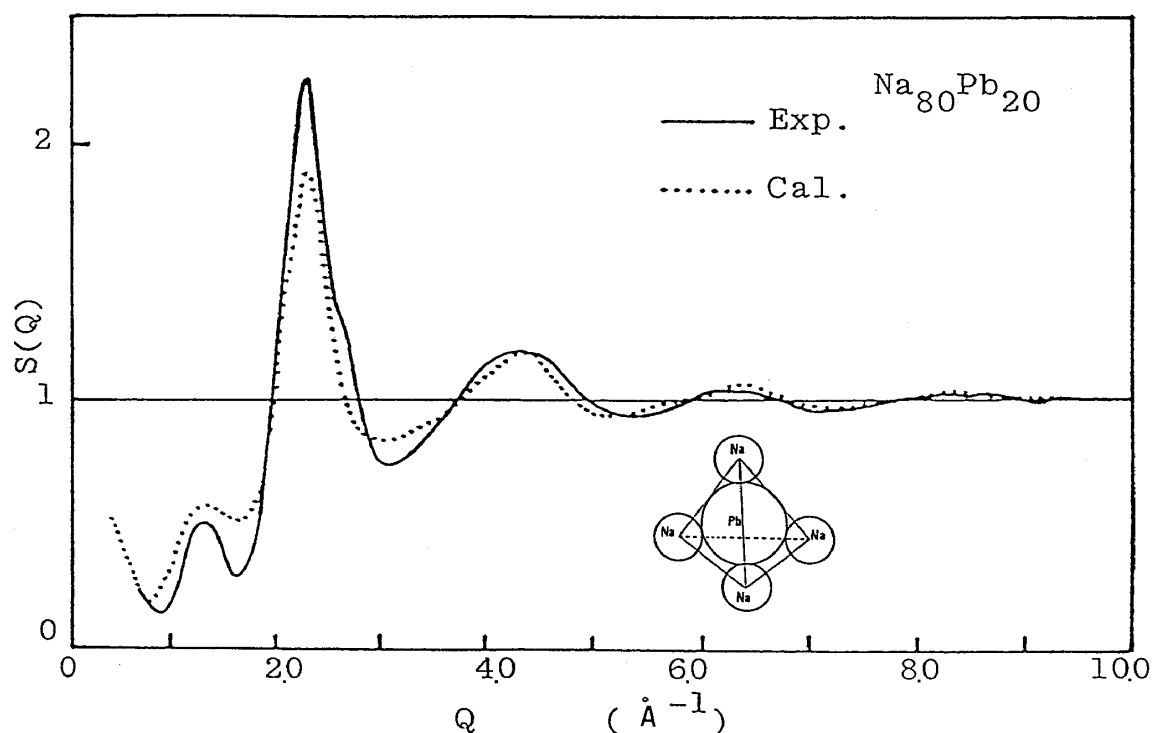


Fig. 10 Comparison of the calculated structure factor of liquid Na_4Pb with the experimental data of liquid $\text{Na}_{80}\text{Pb}_{20}$ [8].

The compound forming character has also been suggested in various liquid metal-chalcogen alloys such as Te-based and Se-based alloys. Figure 11 show the total structure factors and radial distribution functions of liquid Ga-Te alloys as a function of Te concentration determined by X-ray diffraction [31]. The basic profiles of these structural functions are rather close to the profiles found in the case of pure Te. From these figures the following facts could be recognized:

- 1) The total structure factors of all alloys show the characteristic second peak at about $Q=33 \text{ nm}^{-1}$ which is detected in pure Te.
- 2) The measured RDFs of alloys show the split first peak and its splitting feature become intense with an increase of Te concentration.
- 3) The particular peak at a distance of about 0.43 nm corresponding to the third peak in pure Te appears in alloys.

The compound forming character of liquid Ga-Te alloys is known to be distinct in the close vicinity of Ga_2Te_3 . In order to obtain insight into the compound forming features in this melts, the partial

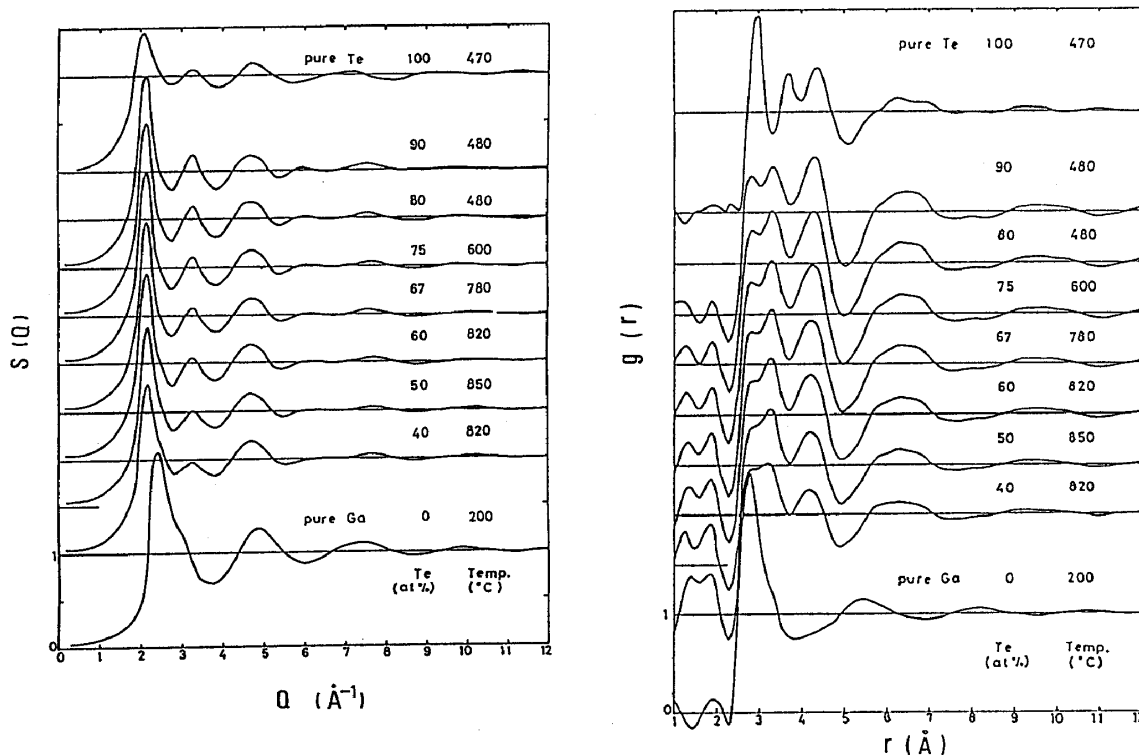


Fig. 11 Total structure factors and RDFs of liquid Ga-Te alloys measured by X-ray diffraction [31].

structures of the Ga_2Te_3 alloy were estimated and they are given in Fig.12 [31]. The basic profiles of the partial functions of like atom pairs, Ga-Ga and Te-Te, are very similar to those of pure Ga and Te. On the other hand the profile of the partial RDF of Ga-Te pairs does not lie in the simple average values of two like atom pairs. These observation can not certainly be explained by the random mixing of Ga and Te and these particular structural features should be attributed by the crystal-like chemical ordering formed by the constituents, Ga_2Te_3 .

The spherical approximation used in the case of Na_4Pb is not allowed in the present Ga_2Te_3 compound. A stereoscopic model is rather convenient for this purpose using the structural parameters such as the interatomic distance and its coordination number in near neighbor region experimentally determined from the structure data.

The partial coordination numbers, Z_{ij} , of Ga_2Te_3 melt are summarized in Table 1 together with the values of crystalline Ga_2Te_3 . A local ordering picture of Ga_2Te_3 melt can be constructed using the value of the coordination number. The following inconsistency is pointed out from the results of Table 1. If the local ordering units

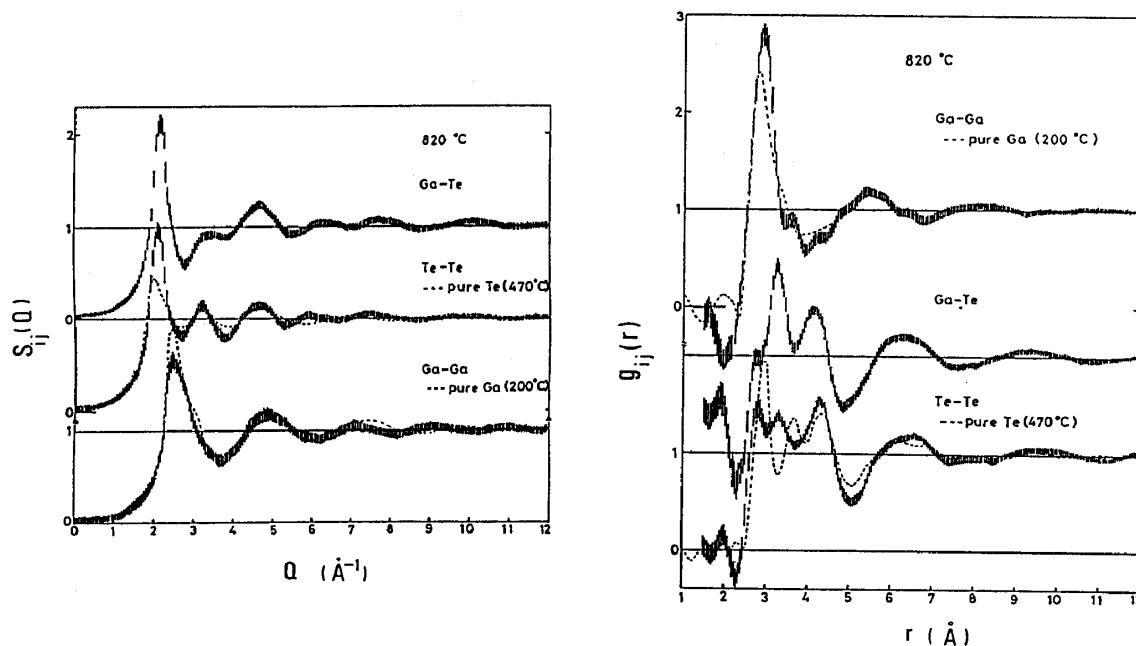


Fig. 12 Partial structural functions of liquid Ga_2Te_3 [31].
The vertical lines indicates the experimental uncertainties .

Table 1 Comparison of the experimental partial coordination numbers of liquid Ga_2Te_3 with those of a defected zinc blend type local ordering crystal structure suggested by Wyckoff [31].
(r:A)

Ga-Ga				Ga-Te					Te-Te			
Observed in the liquid state		Estimated from crystal structure		Observed in the liquid state		Estimated form crystal structure			Observed in the liquid state		Estimated form crystal structure	
\bar{r} [Å]	$Z_{\text{Ga-Ga}}$	\bar{r} [Å]	Ga-Ga	\bar{r} [Å]	$Z_{\text{Ga-Te}}$	\bar{r} [Å]	Ga-Te	Te-Ga	\bar{r} [Å]	$Z_{\text{Te-Te}}$	\bar{r} [Å]	Te-Te
2.9	1.9	2.8	1	2.8	1.6	2.81	3	2 or 3	2.8	1.4	2.81	2
3.6	0.7			3.3	3.2	3.44	1	1 or 2	3.4	1.7		
						3.97	2	2 or 1				
4.3	1.1	4.44	2	4.1	8.1	4.44	2	2 or 1	4.3	4.8	4.44	4

Table 2 Comparison of the experimental partial coordination numbers of liquid Ga_2Te_3 with those of a model structure employed in this work. When considering the partial dissociation, the first coordination numbers are given in the bottom line in this table [31].
(r:A)

Ga-Ga				Ga-Te					Te-Te			
Observed		Estimated		Observed		Estimated			Observed		Estimated	
\bar{r} [Å]	$Z_{\text{Ga-Ga}}$	\bar{r} [Å]	Ga-Ga	\bar{r} [Å]	$Z_{\text{Ga-Te}}$	\bar{r} [Å]	Ga-Te	Te-Ga	\bar{r} [Å]	$Z_{\text{Te-Te}}$	\bar{r} [Å]	Te-Te
2.9	1.9	2.81	2	2.8	1.6	2.81	2 or 3	2 or 1	2.8	1.4	2.81	2
				3.3	3.2	3.44	1 or 0	1 or 0	3.4	1.7	3.44	1 or 0
						3.97	2 or 1	1 or 2			3.97	1 or 0
						4.44	5 or 4	4 or 3			4.44	3 or 2
4.3	1.1	4.44	1	4.1	7.8				4.3	4.8		
2.9	1.9	2.81	2.0	2.8	1.6	2.81	1.72		2.8	1.4	2.81	1.55

be concluded that a feasible origin for the structural inhomogeneity for explaining the particular features of Ga_2Te_3 melt is the existence of the defected-zinc-blend type local ordering units found in crystalline Ga_2Te_3 with some modification and such local ordering units are gradually dissociated to form simpler mixture as the temperature increase. It may be stressed that similar interpretation has been well approved in other semiconducting melts such as In-Te, Sn-Te [32] and Pb-Te [33].

6. Concluding remarks

In this paper, we describe an outline of the structural inhomogeneity in semiconducting melts with special reference to the fundamentals of the crystal growth process. Although the information is still limited at the present time, the structural inhomogeneity in melts attribute to a certain chemical short range ordering (cluster or macro-molecule) which has been well recognized in several melts. The possible chemical short range ordering producing the structural inhomogeneity in melts near the melting point well correlates to the structural units found in a certain crystalline compound. Further quantitative studies in connection with structural inhomogeneity in melts are strongly required for fuller understanding the mechanism of the crystal growth process.

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