Influence of Sb substitution on thermal and electrical characteristics of Ge-Sn-Se chalcogenide glass system

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Abstract. Present research work reports the systematic investigation of thermal and electrical characterizations of Ge₂₀Sn₁₀Se₇₀₋ₓSbx (x = 0, 3, 6, 9, 12, 15) glass system to probe the structural modifications. Bulk samples are prepared using melt quenching technique. Differential scanning calorimetric technique (DSC) is used at the constant heating rate of 10°C under nonisothermal conditions. Different kinetic parameters viz. glass transition temperature Tg, Hruby parameter Hr, crystallization temperature Tc, etc., have been calculated. Glasses under study shows good thermal stability, hence, can find practical applications especially in optical fiber technology. Further, in the current study, an attempt has been made to observe the variation in conductivity with respect to the increase in temperature for amorphous Ge₂₀Sn₁₀Se₇₀₋ₓSbx (x = 0, 3, 6, 9, 12, 15) using two probe method and hence the band structure and corresponding conduction mechanism is studied.

1. Introduction
Chalcogenide based thin films have attracted great attention of scientific community owing to their unique features like interesting electron transport mechanism, good thermal, mechanical and chemical properties [1-2]. Amorphous nano-scale structures have various advantages over crystalline nano-structures. It is well known that synthesis of crystalline nano-structures is controlled by deposition of atoms one by one. On the other hand in amorphous chalcogenide thin films atomic level handling is meaningless owing to its inherent property of disorderness. Thus chalcogenide films exhibit various varieties of structures due to lack of bonding constraints those arises in crystalline unit cells [3].

In Ge-Se system, Ge atoms acts as bond modifiers, which increase average bond strength by crosslinking with Se. This make the system more rigid, thus, increasing the glass transition temperature and resistivity [4]. The addition of third impurity introduces modifications into the local atomic structure, which further influence physical properties of the system. Various studies reveal that addition of Sn to Ge-Se matrix weakens the system and makes it at higher wavelength [5].
Much attention has been focused on quaternary chalcogenide glass (C.G) systems. Thus, study [6] revealed that the addition of Sb to Ge-Sn-Se matrix enhanced the glass-forming ability and increased the glass transition temperature \( T_g \).

Thermal characterization of glasses involves the study of glass transition temperature, heat capacity and crystallization kinetics of chalcogenides, which, in turn, helps one to determine the thermal stability, effective working limits, and their practical applications.

In the present work, we have attempted to study the effect of Sb concentration on the thermal properties Ge-Sn-Se chalcogenide glass system. Differential Scanning Calorimetry (DSC) is very useful technique for the study of crystallization in glass system. This technique is particularly important due to the fact that (i) it is easy to carry out (ii) it is quite sensitive (iii) it requires little apriori sample preparation, etc. [7]. In the present study, non-isothermal method for calorimetric measurements is used.

Electrical properties of chalcogenide glasses C.Gs are important to understand the transport mechanism and hence the phenomenon of conductivity in glasses. Electrical study offer vital information regarding electronic band structure, conduction mechanism in chalcogenide materials. Study of electrical properties of C.Gs is still under extensive study. In the current study, an attempt has been made to observe the variation in conductivity with respect to the increase in temperature for amorphous \( \text{Ge}_{20}\text{Sn}_{10}\text{Se}_{70-x}\text{Sb}_x \) (\( x = 0, 3, 6, 9, 12, 15 \)) using two probe method and hence to predict the band structure and corresponding conduction mechanism.

2. Experimental techniques

Bulk samples of chalcogenide glass system \( \text{Ge}_{20}\text{Sn}_{10}\text{Se}_{70-x}\text{Sb}_x \) (\( x = 0, 3, 6, 9, 12, 15 \)) were prepared using conventional melt quenching technique. The constituent elements of 5N purity were weighed according to their atomic percentages and were sealed in evacuated quartz ampoules of 8 mm inner diameter. The ampoules were then heated to 1000 \( ^\circ \text{C} \) at slow rate of 2-3 \( ^\circ \text{C} \) per min. and the peak temperature was maintained for 48 hours. During heating, ampoules were shaken regularly to ensure the homogeneous mixing of constituent elements. The samples were quenched in ice cold water. The samples were recovered from the ampoules by dissolving the ampoules in solution of HF+H\(_2\)O\(_2\).

Thin films of \( \text{Ge}_{20}\text{Sn}_{10}\text{Se}_{70-x}\text{Sb}_x \) (\( x = 0, 3, 6, 9, 12, 15 \)) were then produced using thermal evaporation process (Hind High Vacuum Coating Unit (model no. 12A4D)). Thin films were deposited onto a pre-cleaned glass substrate at the rate of 5-8 nm/sec, in a vacuum better than 10\(^{-5}\) torr. For cleaning of substrate three steps were used (i) washing with soap solution (ii) cleaning with acetone (vapor cleaning) (iii) cleaning with methanol. Washing with soap solution involved scrubbing substrates with soap and rinsing with distilled water. This removes dirt, oil and grease from the substrate. Cleaning with acetone removes organic impurities whereas methanol removes inorganic impurities from it. After cleaning, substrate was dried by placing in oven at temperature \( \sim 110^\circ \text{C} \). Then substrates were placed in deposition chamber.

In order to confirm the amorphous nature of specimen so produced, the X-Ray diffraction technique was used. Absence of prominent peaks confirms the amorphous nature of specimens. Figure 1 shows the XRD diffractograms of \( \text{Ge}_{20}\text{Sn}_{10}\text{Se}_{70-x}\text{Sb}_x \) (\( x = 0, 3, 6, 9, 12, 15 \)).
In the present study, FESEM-EDAX was carried out to verify the compositions of as prepared thin films. FESEM was conducted using an Hitachi SU 8010 model with field emission tungsten-based gun. The EDAX analysis of the samples was performed by using Bruker EDX flash detector model 6130. For FESEM platinum coating of the specimen was done using sputtering.

![XRD diffractograms of Ge\textsubscript{20}Sn\textsubscript{10}Se\textsubscript{70-x}Sbx (x = 0, 3, 6, 9, 12, 15)](image)

**Figure 1.** XRD diffractograms of Ge\textsubscript{20}Sn\textsubscript{10}Se\textsubscript{70-x}Sbx (x = 0, 3, 6, 9, 12, 15)

![EDAX results for Ge\textsubscript{20}Sn\textsubscript{10}Se\textsubscript{70}](image)

**Figure 2.** EDAX results for Ge\textsubscript{20}Sn\textsubscript{10}Se\textsubscript{70}
Figure 3. EDAX results for Ge$_{20}$Sn$_{10}$Se$_{68}$Sb$_{6}$

Figure 4. EDAX results for Ge$_{20}$Sn$_{10}$Se$_{58}$Sb$_{12}$
Figure 5. FESEM images of Ge$_{20}$Sn$_{10}$Se$_{70}$

Figure 6. FESEM images of Ge$_{20}$Sn$_{10}$Se$_{64}$Sb$_{6}$

Figure 7. FESEM images of Ge$_{20}$Sn$_{10}$Se$_{58}$Sb$_{12}$
In the present study, the differential scanning calorimetric DSC technique was used under nonisothermal conditions for thermal characterization. DSC instrument was first calibrated with alumina powder as standard material before recording thermal history of the sample. DSC scan was produced under the constant flow of N2 (200 ml/min) to avoid oxidization of the sample, which could lead to some unwanted features in the scan. About 10mg of the sample of Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_{x}$ (x = 0, 3, 6, 9, 12, 15) was taken in alumina pan with 10mg of alumina powder as the reference material. Scan is produced in the temperature range of 70°C to 500°C with the constant heating rate of 10°C/min.

For electrical characterization of thin films two probe methods was used by authors. Measurements were taken in the temperature range of 289-333K. Electrodes were designed using silver paste with ~99.99% purity. For I-V characteristics, D.C voltage ranging 0-500V was applied across the ohmic contacts, and the corresponding current was noted through picometer, whereas Keithley (model 6487) was used by the authors for electrical characterization.

3. Results and discussion

3.1 Thermal characterization of Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_{x}$ (x = 0, 3, 6, 9, 12, 15) chalcogenide glasses

DSC thermograms of all the samples in the temperature range 70 °C − 500 °C are shown in figure 8. Single glass transition temperature observed in thermograms for all the samples indicates that the samples were compositionally homogenous. From DSC curves, glass transition temperature has been calculated from the intersection of tangents from base line and slope of the step. The values of glass transition temperature (T$_{g}$), onset crystallization temperature (T$_{c}$) and peak crystallization temperature (T$_{p}$) calculated from thermograms are summarized in table 1.

![Figure 8. DSC thermograms of Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_{x}$ (x = 0, 3, 6, 9, 12, 15) chalcogenide glasses.](image)

The values of glass transition temperature T$_{g}$, crystallization temperature T$_{c}$, peak temperature T$_{p}$, melting temperature T$_{m}$, Thermal stability temperature T$_{c}$-T$_{g}$ and Hruby parameter H, has been calculated for Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_{x}$ (x = 0, 3, 6, 9, 12, 15) chalcogenide glasses are given in table 1.
Table 1. Glass transition temperature ($T_g$), onset crystallization temperature ($T_c$) and peak crystallisation temperature ($T_p$), Hruby parameter ($H_r$), reduced glass transition temperature ($T_{rg}$) for Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_{x}$ ($x = 0, 3, 6, 9, 12, 15$) chalcogenide glasses.

| Composition       | $T_g$ ($^\circ$C) | $T_c$ ($^\circ$C) | $T_p$ ($^\circ$C) | $T_m$ ($^\circ$C) | $T_c-T_g$ ($^\circ$C) | $H_r$ | $T_{rg}$ |
|-------------------|-------------------|-------------------|-------------------|-------------------|----------------------|-------|----------|
| Ge$_{20}$Sn$_{10}$Se$_{70}$ | 209.63            | 369.91            | 432.10            | -                 | 160.28               | -     | -        |
| Ge$_{20}$Sn$_{10}$Se$_{67}$Sb$_{3}$ | 216.41            | 316.31            | 332.56            | 394.42            | 99.90                | 1.28  | 0.55     |
| Ge$_{20}$Sn$_{10}$Se$_{64}$Sb$_{6}$ | 219.72            | 298.13            | 319.11            | 461.92            | 78.41                | 0.48  | 0.48     |
| Ge$_{20}$Sn$_{10}$Se$_{61}$Sb$_{9}$ | 214.33            | 295.92            | 314.92            | 457.76            | 81.59                | 0.50  | 0.47     |
| Ge$_{20}$Sn$_{10}$Se$_{58}$Sb$_{12}$ | 210.92            | 297.41            | 311.23            | 454.15            | 86.49                | 0.55  | 0.46     |
| Ge$_{20}$Sn$_{10}$Se$_{55}$Sb$_{15}$ | 206.47            | 283.62            | 305.56            | 451.12            | 77.15                | 0.46  | 0.46     |

It can be observed from table 1 that glass transition temperature $T_g$ increases up to 6 at.% of Sb, after which a decrease in $T_g$ is observed. This trend can be explained on the basis of CON (chemically ordered network) model. Calculating the hetropolar bond energies using the Pauling relation [8], we get

$$E(A - B) = \left[ E(A - A) \times E(B - B) \right]^{\frac{1}{2}} + 30(\chi_A - \chi_B)^2$$

(1)

where E(A-A) and E(B-B) are energies of homonuclear bonds and $\chi_A$ and $\chi_B$ are electronegativity values for atoms involved, the values of bond energies are given in table 3. According to CON model bonds are formed in the sequence of decreasing energy. Thus Ge-Se stronger bonds will form first followed by Sn-Se, Se-Sb and so on. It is evident from table 2 that upto 6 at.% of Sb stronger bonds viz. Ge-Se, Se-Sn, Se-Se dominates over weaker homopolar Se-Se, Sb-Sb bonds leading to the increase in mean bond energy and hence glass transition temperature. But with further increase in Sb atoms, concentration of weaker Sb-Sb, Sn-Sb, Sn-Sn bonds increases resulting into decrease in glass transition temperature $T_g$.

Table 2. Values of distribution of bonds for Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_{x}$ ($x = 0, 3, 6, 9, 12, 15$).

| Distribution of bonds |
|-----------------------|
| Ge-Se | Se-Sn | Se-Sb | Se-Se | Sn-Sn | Sn-Sb | Sb-Sb |

| Ge$_{20}$Sn$_{10}$Se$_{70}$ | 0.57 | 0.28 | -     | 0.14 | -     | -     |
| Ge$_{20}$Sn$_{10}$Se$_{67}$Sb$_{3}$ | 0.60 | 0.30 | 0.06  | 0.04 | -     | -     |
| Ge$_{20}$Sn$_{10}$Se$_{64}$Sb$_{6}$ | 0.60 | 0.31 | 0.05  | -    | -     | 0.03  |
| Ge$_{20}$Sn$_{10}$Se$_{61}$Sb$_{9}$ | 0.59 | 0.30 | -     | 0.01 | -     | 0.10  |
| Ge$_{20}$Sn$_{10}$Se$_{58}$Sb$_{12}$ | 0.59 | 0.26 | -     | -    | 0.01  | 0.13  |
| Ge$_{20}$Sn$_{10}$Se$_{55}$Sb$_{15}$ | 0.58 | 0.22 | -     | -    | 0.02  | 0.02  | 0.15  |
Table 3. Values of bond energy for Ge-Sn-Se-Sb System [9]

| Bonds   | Bond Energy (Kcal/mol) |
|---------|------------------------|
| Ge-Se   | 49.42                  |
| Sn-Se   | 49.23                  |
| Se-Sb   | 43.96                  |
| Se-Se   | 44.00                  |
| Sn-Sn   | 34.20                  |
| Sn-Sb   | 32.38                  |
| Sb-Sb   | 30.20                  |

Thermal stability of glasses is very important from their practical application point of view, which reflects the resistance to devitrification after the glass formation. Glass forming ability GFA is the ease with which the melt can be cooled to form glass. Nascimento et al [10] suggested that slower is the rate of crystallization higher will be the glass formation ability [10]. Although thermal stability and glass forming ability GFA are two important and independent characteristics of glasses but number of studies are made to relate these two parameters [11-15]. Glass transition temperature $T_g$ represents the rigidity of glassy structure. Thus, $T_g$ can be associated with thermal stability of glass but GFA cannot be determined with $T_g$ alone. Sakka and Meckenzie [16] used the parameter of reduced glass transition temperature $T_{rg}=T_g/T_m$ as glass stability parameter. For glass formation, $T_{rg}$ has range i.e. $1/2 \leq T_{rg} \leq 2/3$. In the present work, $T_{rg}$ is approximately 0.5 i.e. lies in the lower accepted range. Turnbull [17] proposed that for a given composition glass forming ability decreases with the decrease in $T_{rg}$. So, there is a need to search for another parameter that can relate both GFA and $T_g$.

Dietzel [18] suggested parameter $\Delta T=T_c-T_g$ which is the strong indicator for both thermal stability and GFA of glasses, as calculated in table 1. Unstable glasses generally have small difference between crystallization temperature and glass transition temperature and soon these glasses tend to crystallize. Higher difference between $T_c$ and $T_g$ retards the nucleation process, thus GFA increases. [19]. Hruby [20] introduced a new parameter, which combines both growth and nucleation aspects of phase transformation:

$$H_r = \left(\frac{T_c-T_g}{T_m-T_c}\right)$$  

(2)

Glass formation will be difficult for $H_r \leq 0.1$ and easy for $H_r \geq 0.4$ [20]. In this paper, glasses under study have good glass-forming ability as indicated in table 1. Hence as prepared glasses can find practical applications viz for optical fibres, phase-change memory, etc.

3.2 Improvement of electrical properties of ternary Ge-Sn-Se chalcogenide thin films by the Sb additions

In the present study of amorphous Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sbx ($x = 0, 3, 6, 9, 12, 15$), the whole mechanism is divided into two parts (i) deviation from ohmic to non-ohmic behavior of I-V curve at higher voltage (ii) dependence of conductivity on composition.

Efforts have been made to evaluate DC conductivity at room temperature via relation

$$\sigma_{RT} = \frac{L}{RA}$$  

(3)

where $L$ is sample thickness, $A$ is cross-sectional area, and $R$ is sample resistance. The values of $R$ for all samples at room temperature are derived from the ohmic (linear) part of I-V curve.
Figure 9. Current (A) vs. voltage (V) for Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_x$ (x = 0, 3, 6, 9, 12, 15).

Figure 9 shows the variation of current with respect to voltage. The non-ohmic nature of samples is shown in figure 3 for a specific range of voltages, where the ohmic part for 0-5V is observed, after which the graph deviates from its linear path at high voltage. Linear portion of the graph reveals that the charge carriers are generated due to thermal excitations. However, the deviation from the linear path reflects the non-ohmic aspect of films. Space charge limited conduction (SCLC) model implies that higher voltages applied to the sample causes electrodes to inject the non-equilibrium density of charge carriers. The latter populate above the Fermi level. The larger population of these charges, as compared to thermally generated charge carriers, causes a deviation from the linearity [21].

Table 4. Values of activation energy $E_a$, optical band gap $E_g$, conductivity at room temperature $\sigma_{RT}$, resistivity $\rho_{RT}$, pre-exponential function $\sigma_0$, cohesive energy C.E for Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sb$_x$ (x = 0,3, 6, 9, 12, 15).

| S. No. | Composition | Conductivity $\sigma_{RT}$ (mho m$^{-1}$) | Resistivity $\rho_{RT}$ (ohm m) | Activation energy $E_a$ (eV) | $E_g$ (eV) | Optical band gap (Tauc) | $\ln\sigma_0$ | Pre-exponential function $\sigma_0$ (mho m$^{-1}$) | C.E° (Kcal/mol) |
|--------|-------------|----------------------------------------|-------------------------------|----------------------------|---------|----------------------|----------------|--------------------------------|-----------------|
| 1      | Ge$_{20}$Sn$_{10}$Se$_{70}$ | 1.88 x 10$^{-4}$ | 5308.54 | 0.75 | 1.66 | -1.57 | 2.08 x 10$^{-1}$ | 48.59 |
| 2      | Ge$_{20}$Sn$_{10}$Se$_{70}$Sb$_3$ | 2.06 x 10$^{-2}$ | 48.49 | 0.44 | 1.54 | -9.74 | 5.88 x 10$^{-4}$ | 48.79 |
| 3      | Ge$_{20}$Sn$_{10}$Se$_{67}$Sb$_6$ | 1.12 x 10$^{-2}$ | 89.31 | 0.37 | 1.49 | -13.05 | 2.16 x 10$^{-6}$ | 47.54 |
| 4      | Ge$_{20}$Sn$_{10}$Se$_{64}$Sb$_9$ | 9.00 x 10$^{-3}$ | 111.11 | 0.33 | 1.45 | -13.13 | 1.98 x 10$^{-6}$ | 47.39 |
| 5      | Ge$_{20}$Sn$_{10}$Se$_{61}$Sb$_{12}$ | 8.22 x 10$^{-3}$ | 121.68 | 0.29 | 1.42 | -13.15 | 1.94 x 10$^{-6}$ | 46.60 |
| 6      | Ge$_{20}$Sn$_{10}$Se$_{58}$Sb$_{15}$ | 7.97 x 10$^{-3}$ | 125.46 | 0.28 | 1.38 | -14.81 | 3.71 x 10$^{-7}$ | 45.64 |
From table 4, it is observed that there is an overall decrease in conductivity at room temperature \( \sigma_{RT} \) with the increase in concentration of Sb in Ge-Sn-Se thin films. Conductivity at room temperature \( \sigma_{RT} \) shows an abrupt increase for 3 at% of Sb. This observation is in agreement with cohesive energy as shown in the table 4. Similar trend is observed for glass transition temperature \( T_g \) as studied in previous section of our work which shows the system is rigid for this composition and with the further increase in Sb concentration the system loses its rigidity. Overall decrease in \( \sigma_{RT} \) can be explained by the fact that Se has tendency to form polymerized network. Scottmiller et al. [2] reported that, in glassy state, 60% of Se atoms are bonded as polymeric chains, while rest of atoms are bonded in the ring-like structure [22]. The addition of Sb to the ternary Ge-Sn-Se glass system causes the formation of stronger Se-Sb (43.96 Kcal/mol) bonds leading to an increase in C.E. However, further increase in Sb concentration leads to the formation of weaker Sn-Sb (32.38 Kcal/mol) and Sb-Sb (30.20 Kcal/mol) bonds as the cost of stronger bonds, which reduces the cohesive energy and, thus, conductivity. In other words, conductivity at room temperature decreases with Sb concentration due to the increase in concentration of defect states associated with impurity atoms [23]. Observations are in agreement with the results of Jain and Saxena [24]. The decrease in \( \sigma_{RT} \) can be correlated with the decrease in mean free path owing to the short range order of glasses. The temperature dependence of DC conductivity has been determined for the temperature range of (289-333K) for as prepared thin films of amorphous Ge\(_{20}\)Sn\(_{10}\)Se\(_{70-x}\)Sbx (x = 0, 3, 6, 9, 12, 15). At higher temperature, dependence of DC conductivity with temperature is given by the Arrhenius formula

\[
\sigma(T) = \sigma_0 \exp\left(\frac{-E_a}{k_B T}\right)
\]

where \( \sigma_0 \) is the pre-exponential factor which includes charge carrier mobility and density of states. Pre-exponential factor \( \sigma_0 \) indicates whether the conduction mechanism takes place in extended states above mobility edge or by hopping in localized states. \( E_a \) represents the activation energy. According to Mott and Davis (1979) for conduction in localized states, \( \sigma_0 \) is in the range of \( 10^3 \) to \( 10^5 \) \( \Omega^{-1} \text{m}^{-1} \) [25]. Value of \( E_a \) has been determined by fitting a slope for the plot between \( \ln \sigma \) and \( 1000/T \), as shown in figure 4. Activation energy \( E_a \) is the smallest energy required by electrons to overcome the potential barriers, thus influencing the structural relaxation. Thus, \( E_a \) implies the energy differences either (\( E_C - E_F \)) or (\( E_F - E_V \)) for conduction in localized states.

![Figure 10. \( \ln \sigma \) versus 1000/T (K\(^{-1}\)) for various compositions of Sb in Ge\(_{20}\)Sn\(_{10}\)Se\(_{70-x}\)Sbx (x = 0, 3, 6, 9, 12, 15) chalcogenide thin films.](image)
$E_a$ is generally found to be half of the optical band gap $E_g$ ($E_g \sim 2E_a$) [26] implying that the Fermi level lies near the mid gap. In the present study, it is found that values of $E_a$ is less than the half of $E_g$ thus Fermi level in the present study is not pinned to the middle of band gap.

It is observed that $E_a$ and $\sigma_0$ decrease with the concentration of Sb. It is known that Sb forms ionic bonds and, thus, makes the system ionic in nature. This weakens the network and deviates the structure towards fragility causing the decrease in C.E and conductivity [27].

Also decrease in activation energy can be attributed to the formation of weaker homopolar bonds, which, in turn, reduce the average single bond energy [28]. Such small values of $E_a$ and pre-exponential function $\sigma_0$ imply that the conductivity is due to thermally generated charge carrier motion in the band tails of localized states, which finding is in accordance with the results of earlier studies [29-30].

4. Conclusions
This study confirmed that glass transition temperature $T_g$ increases with Sb concentration up to 6 at.% Sb, after which a decrease in $T_g$ is observed in Ge$_{20}$Sn$_{10}$Se$_{70-x}$Sbx ($x = 0, 3, 6, 9, 12, 15$) chalcogenide glass system. DSC thermographs reveal a higher difference between glass transition temperature $T_g$ and crystallization temperature $T_c$, which implies that the glassy system is quite stable and shows resistance to nucleation process. Further glass forming ability and stability is verified using Hruby parameter and reduced glass transition temperature. Glasses under study show good thermal stability and, hence, can find practical applications, especially, in optical fiber technology field.

An overall conductivity deterioration was observed at room temperature $\sigma_{RT}$ with the concentration of Sb in Ge-Sn-Se thin films. Conductivity at room temperature $\sigma_{RT}$ shows an abrupt increase for 3 at% of Sb. Although an overall decrease in $\sigma_{RT}$ has been observed, it can be correlated with the mean free path decrease due to the short range order of glasses.

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