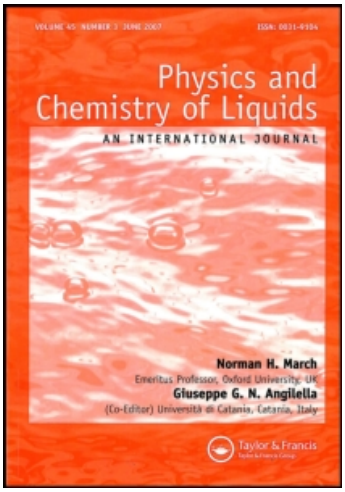

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Electrical Properties of $AgSbTe_2$ in the Solid and Liquid States

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ELECTRICAL PROPERTIES OF $AgSbTe_2$ IN THE SOLID AND LIQUID STATES

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The electrical conductivity and thermoelectric power of $AgSbTe_2$ have been investigated as a function of temperature from 240°C to 850°C. Experimental data are analyzed in terms of a model developed from the density of states and electrical transport in solid amorphous semiconductors[1]. The activation energy calculated from electrical conductivity data is found to be 0.218 eV for the solid and 0.43 eV for the liquid. Moreover, the coefficient of the linear decrease of energy gap with temperature was found to be 3.367×10^{-4} eV/°K.

KEY WORDS: Conductory, thermopower, activation energy

1 INTRODUCTION

Ternary chalcopyrite semiconductors have recently attracted considerable interest because of their potential applications in optoelectronic devices[2,3] and nonlinear optics.

The electronic transport properties of some liquid chalcopyrite semiconductors is of considerable interest. There are serious reasons for believing that the electronic states at the band edges are perturbed on melting[4,5]. These states which are 'tailed' in the forbidden gap become localized.

The transport properties of $CuSbSe_2$, $CuBiSe_2$ and $AgTlTe_2$ were studied and published [6–8] where recent models for amorphous semiconductors are successfully applied. The aim of the present contribution is to investigate the electrical behaviour of $AgSbTe_2$ in the solid and liquid phases.

The melting point of $AgSbTe_2$ is about 556°C.

2 EXPERIMENTAL

The sample was prepared by melting the proper amounts of high pure components elements taken in their stoichiometric ratio. The material was sealed in a silica tube at 10^{-3} Pa, and heated at 1100°C for 12h with frequent rocking using an oscillating oven to ensure homogeneity of the melt. Then the tube was quenched in air to obtain the sample in the amorphous state. The solid material is then transferred to the measuring

cell. The measuring cell was fitted with graphite electrodes, heaters and thermocouples for accurate measurements of temperatures[9].

Measurements of the electrical conductivity were carried out in inert atmosphere using a highly stabilized power supply, a sensitive voltmeter and a sensitive digital electronic multimeter (type Systron Donner model 7003) capable of measuring currents as low as 10^{-6} A. The sample was investigated up to 240°C above its melting point.

3 RESULTS AND DISCUSSION

Figures (1) and (2) show the electrical conductivity and thermoelectric power data in the solid and liquid states. In the solid state the electrical conductivity increases with temperature, then decreases near melting point. In the liquid state the electrical conductivity increases exponentially with temperature, the activation energy in the solid $E_{a\,\text{solid}} = 0.218$ eV and in the liquid state $E_{a\,\text{liquid}} = 0.43$ eV. The thermoelectric power is positive in the solid state which indicates that $AgSbTe_2$ is a *p*-type semiconductor. After melting the thermoelectric power decreases with temperature and near 838°C it changes sign and becomes negative. At the melting point a sudden decrease in the electrical conductivity and thermoelectric power is observed, which may be attributed to changes in the short range order on melting, and the subsequent decrease in carrier mobility. Moreover, the electrical conductivity increases with temperature.

The negative behaviour of the thermoelectric power in the liquid state at high temperature can be understood in terms of a transition between transport in two different bands. The region between the peaks—in magnitude—of the thermoelectric power is explained by ambipolar transport[10], which suggests that there is cancellation between the positive and negative contributions to the thermoelectric power due to hole and electron transport[10].

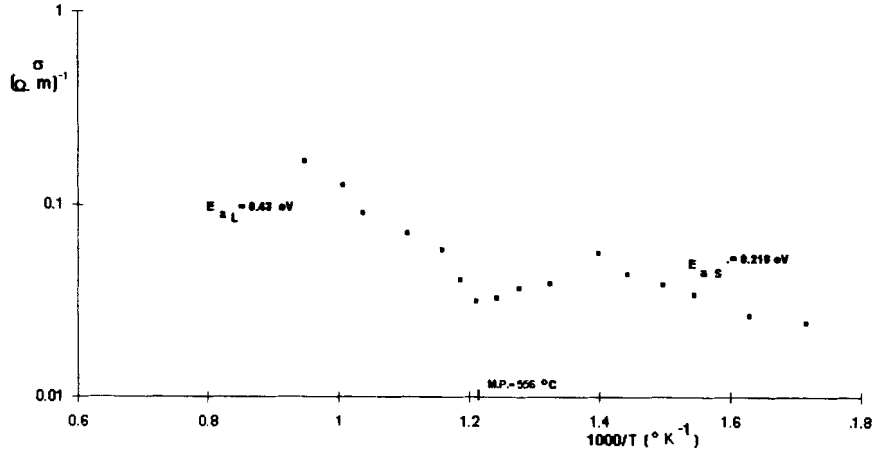

Figure 1 Temperature dependence of the electrical conductivity of $AgSbTe_2$ in solid and liquid states.

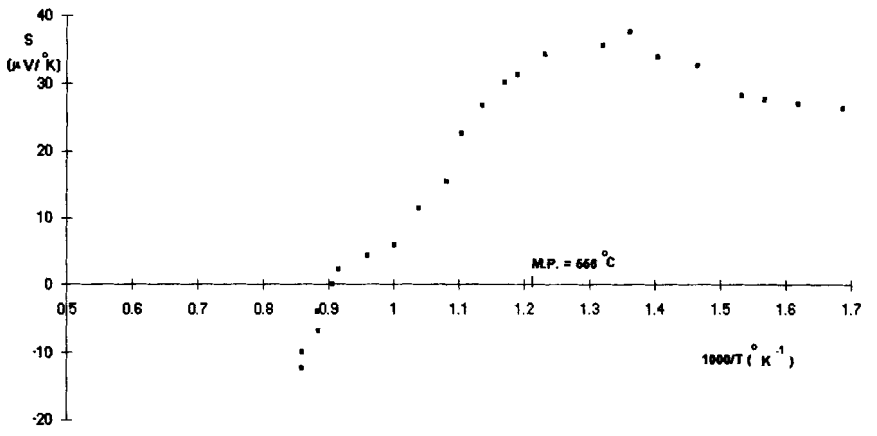

Figure 2 Temperature dependence of the thermoelectric power of $AgSbTe_2$ in solid and liquid states.

Various workers[11,12] have attributed the negative values of the thermoelectric power to a large predominance of electrons in electrical transport.

The present data can be adequately interpreted according to the model developed by Mott[1] for amorphous semiconductors. This model is derived from that proposed by Cohen *et al.*[4] and by Davis and Mott[13] for a solid amorphous semiconductor. According to this model, the electronic structure of energy bands is not significantly different from that in the corresponding crystal. The main difference is that the electronic states at the band edges are 'tailed' into the forbidden gap and become localized. Therefore, the conduction mechanism changes radically from crystalline to amorphous structure. To explain the positive sign of the thermoelectric power, it is supposed that the range of localized states in the conduction band is wider than in the valence band.

Two conduction processes may occur:

(a) Conduction due to holes excited into extended states at E_v.
(b) Conduction due to holes excited in localized states near the band edge with an activated mobility.

If it is assumed that the range of localized states and the activation energy for hole mobility is small compared with the energy gap, then the electrical conductivity in both cases may be expressed as

$$\sigma = \sigma_0 \exp\left[-\frac{E_f - E_v}{kT}\right] \tag{1}$$

The value of the constant σ_0 varies strongly with the conduction process $(E_f - E_v)$ depends on temperature and is given by:

$$E_f - E_v = E - \gamma T \tag{2}$$

Thus:

$$\sigma = \sigma_0 \exp(\gamma/k) \exp(-E/kT) \tag{3}$$

The temperature coefficient γ may be calculated directly from the thermoelectric power which is expressed as:

$$S = k/e(E/kT - \gamma/k + A) \tag{4}$$

where the constant A is related to scattering mechanisms. If A is known, γ can be determined directly from the intercept on the $1/T = 0$ axis of a plot of S versus $1/T$. By combining expressions (1) and (4), we obtain the relation between σ and S:

$$\sigma = \sigma_0 \exp(-eS/k + A) \tag{5}$$

from which the constant σ_0 can be calculated.

In $AgSbTe_2$ the straight line between $\ln\sigma$ and S (Figure 3) extrapolated to $1/T = 0$ yields $\gamma/k = (S_0/86) + A$. Thus for $A \geqslant 0$, is $\gamma/k \geqslant S_0/86$. According to Cutler and Mott[14], the constant A is of order unity for disordered structures. However, recent investigation of the thermoelectric power of amorphous chalcogenides[15] suggests that A may be larger than unity for highly disordered materials. In our discussion we assume $A = 1$. Then the coefficient γ is found to be 3.367×10^{-4} eV/°K. These high values of γ which give the temperature dependence of the gap for liquid $AgSbTe_2$ is of the same order as that found for liquid Te–Se alloys[16]. It seams likely that the large linear decrease of the gap with increasing temperature is related to the fact that the difference between the distances from one atom to nearest and next-nearest neighbours decrease, Mott[1].

Figure 3 shows the dependence of $\ln\sigma$ as a function of S. The variations of $\ln\sigma$ are linear with S according to relation (5) in the temperature range where the expressions

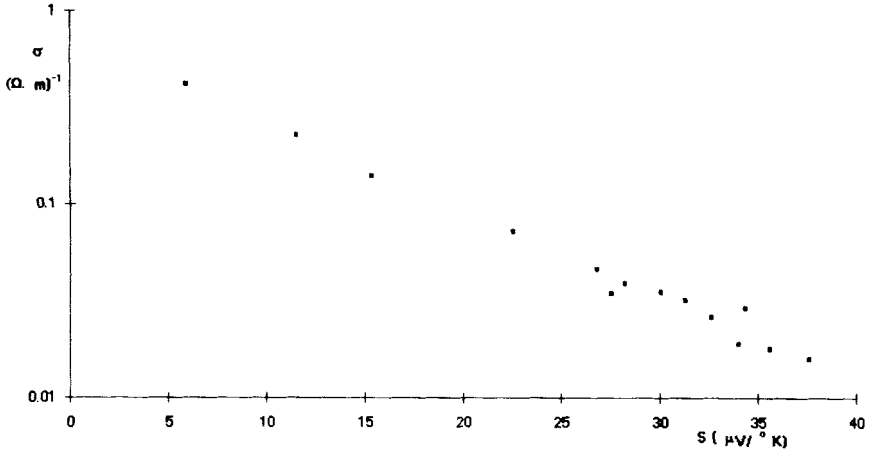

Figure 3 Electrical conductivity versus thermoelectric power for $AgSbTe_2$ in liquid state.

(1) and (4) can be satisfied. With the assumption $A = 1$, the value of the intercept on the $S = 0$ axis yields $\sigma_0 = 0.78\,\Omega^{-1}\,m^{-1}$. For $AgSbTe_2$ with high value of σ_0, the conduction is probably due to holes excited in extended states.

The large linear decrease of the gap may give rise to the transition from semiconducting to metallic behaviour expected at high temperature. As the gap contracts with increasing temperature, the tails of conduction and valence bands become more pronounced. At sufficiently high temperature these tails overlap, leading to a filling in of the gap and the disappearance of localization. Consequently, the electrical conductivity shows a weak temperature dependence, and the thermoelectric power should approach zero slowly.

4 CONCLUSION

The electrical conduction in liquid $AgSbTe_2$ could be analyzed by the model developed by Mott. The electrical conduction mechanism is due to holes excited into extended states near the band edge and the energy gap is 0.86 eV. The factor which gives the temperature dependence of the energy gap is found to be 3.367×10^{-4} eV/°K, which shows a large decrease of the gap with temperature.

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