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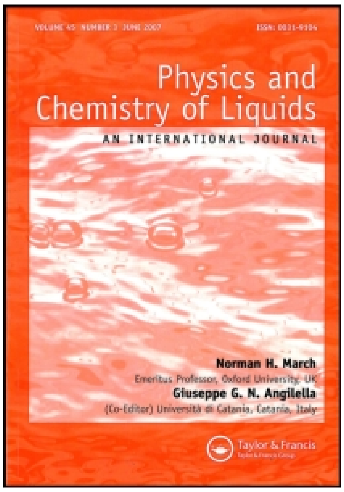

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N. Abdelmohsen[a]; A. Abdelghany[a]; K. Sharaf[a]
[a] Physics Department, Faculty of Science, Al Azhar University, Nasr City, Cairo, Egypt

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THERMAL CONDUCTIVITY OF $CuSbSe_2$ AND $CuBiSe_2$ IN THE SOLID AND LIQUID STATE

N. ABDELMOHSEN, A. ABDELGHANY and K. SHARAF

Physics Department, Faculty of Science,
Al Azhar University, Nasr City, Cairo, Egypt.

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Thermal Conductivity of $CuSbSe_2$ and $CuBiSe_2$ have been studied in the solid and liquid state in a wide range of temperatures. Measurements of thermal conductivity were carried out using the concentric cylinder method which is based on the flow of heat through a cylindrical wall.

KEY WORDS: Rotational molten, heat of melting

INTRODUCTION

Ternary chalcopyrite semiconductors have attracted recently a great deal of attention because of their possible applications in electro-optical devices[1] and infrared generation and detection. They can be regarded as valence analogs of the III-V and II-VI semiconductors respectively[2] and they crystallize in the chalcopyrite structure[3,4] which is closely related to zincblende. Very little work has been reported on the thermal properties of these compounds. Only measurements of the thermal conductivity at room temperature of some II-IV-V_2 compounds have been reported[5]. Oshcherin[6] has shown that the melting entropies of chalcopyrites are like their binary alloys.

The aim of the present contribution is to study the thermal conductivity of $CuSbSe_2$ and $CuBiSe_2$ in a wide range of temperatures in the solid and liquid state.

EXPERIMENTAL DETAILS

The samples were prepared by melting the proper amounts of highly pure component elements (99.999 %). The material was sealed in evacuated quartz tubes at 10^{-3} pa and heated at 1200°C for 12 h with frequent rocking to ensure homogenization of the melt. Then the tubes were quenched in ice to obtain the samples in the amorphous state. The solid material is then heated in inert atmosphere until it melts and then transferred to the measuring cell.

Measurements of thermal conductivity were carried out using the concentric cylinder method where the material in the liquid state was poured into the cylindrical gap between two concentric graphite cylinders kept in nitrogen atmosphere[7]. The system was fitted with a heater and sensitive thermocouples for accurate measurements of temperature and the thermal conductivity was calculated using the formula

$$k = \frac{Q \ln(d_2/d_1)}{2\pi L(t_1 - t_2)}$$

where d_1 and d_2 are the diameters of the inner and outer cylinders, t_1 and t_2 are the temperatures on both sides of the sample, L is the length of the cylinders and Q is the quantity of heat generated. The quantity of heat flowing per second through the sample is given by

$$Q = m\left(\frac{\Delta T}{\Delta t}\right)S$$

where m is the mass of external cylinder, $(\Delta T/\Delta t)$ is the temperature gradient and S is the specific heat of graphite. Measurements were carried out in a wide range of temperatures below and above the melting point.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the thermal conductivity of $CuBiSe_2$. In the solid state the increase of thermal conductivity with temperature can be explained with the relation[8]

$$K_{\text{lattice}} = \tfrac{1}{3}CVL$$

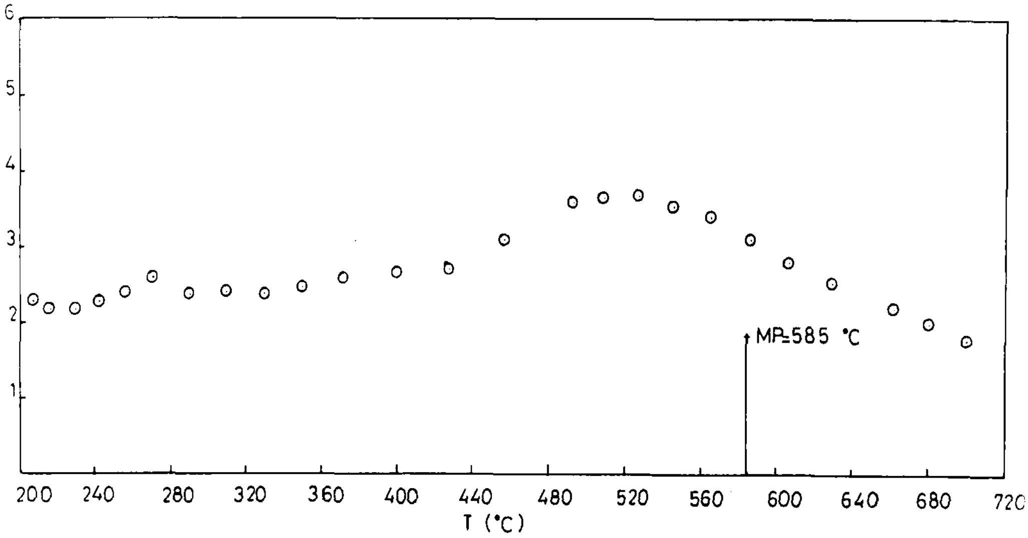

Figure 1 Temperature dependence of the thermal conductivity of $CuBiSe_2$ in the solid and liquid state.

where C is the heat capacity per unit volume, V is the phonon velocity, and L is the phonon mean free path. In the amorphous solids the wavelength of phonons carrying thermal energy is equal to the average distance between the molecules[9] and is nearly temperature independent and the linear temperature dependence of thermal conductivity in the solid state is due to the linear increase of the heat capacity and phonon mean free path.

Moreover the additional increase of thermal conductivity before melting may be attributed to a transition from the amorphous to a highly elastic state characterized by increase of the mobility of the molecules and the distance between them which is followed by the stimulation of rotational oscillations of molecules.

These rotational oscillations will spread along and between the molecules, transferring thermal energy and so the value of k will increase. The decrease of the thermal conductivity on melting can be attributed to the decrease of density on melting and the subsequent increase of the distance between the molecules which leads to a weakening of the bonds between the molecules and an increase in the disordered arrangement. As a result, the heat capacity and the mean free path of the phonons decrease and the thermal conductivity decreases correspondingly.

Powell's[10] theoretical calculations show that the ratio between the thermal conductivity of material at melting point in the crystalline k_s and liquid state K_1 is given by

$$\frac{K_s}{KL} = e\frac{2}{3}\frac{r}{KT_{\text{melt}}}$$

where T_{melt} is the melting point and r is the atomic heat of melting of the material. According to Eq. (1) the thermal conductivity should decrease on melting which explains the drop of thermal conductivity at the melting point.

Figure 2 shows the temperature dependence of the thermal conductivity of $CuSbSe_2$ in the solid and liquid states.

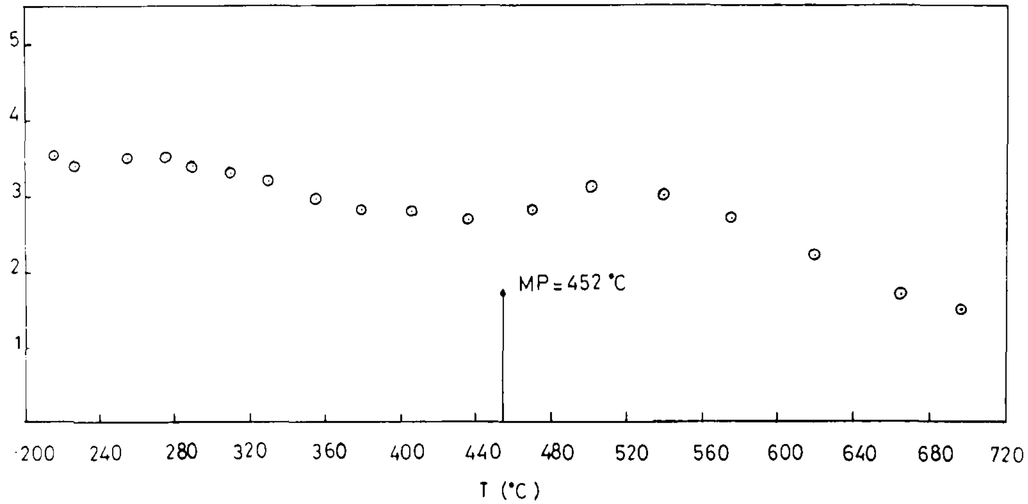

Figure 2 Temperature dependence of the thermal conductivity of CuSbSe2 in the solid and liquid state.

The thermal conductivity of $CuSbSe_2$ varies with temperature as T^{-1} up to melting point indicating that the low-temperature conductivity is of the phonon type. On further heating beyond melting point the thermal conductivity of $CuSbSe_2$ and $CuBiSe_2$ decreases with increasing temperature, indicating that there is no photon heat transfer and K is attributed mainly to moleculer component.

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