

The Absorption Properties of p-Type $TlIn_xGa_{(1-x)}Se_2$ and $TlGaSe_2$

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Abstract

$p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ single crystals were grown by the modified Bridgman-Stockbarger method in our crystal growth laboratory. The absorption measurements were carried out on $p-TlIn_xGa_{(1-x)}Se_2$ samples in temperature range 10-300 K in steps of 10 K. The binding energies of $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ were obtained as 35.0 meV, 16.5 meV and 14.5 meV, respectively. The direct band gaps were calculated as 2.244 eV, 2.195 eV, 2.164 eV in $p-TlGaSe_2$, 2.158 eV, 2.131 eV, 2.098 eV in $p-TlIn_{0.3}Ga_{0.7}Se_2$, and 2.107 eV, 2.075 eV, 2.019 eV in $p-TlIn_{0.5}Ga_{0.5}Se_2$ respectively, at sample temperatures of 10 K, 140 K and 300 K. The indirect band gaps were calculated as 2.196 eV, 2.127 eV, 2.073 eV in $p-TlGaSe_2$ 2.130 eV, 2.101 eV, 2.064 eV in $p-TlIn_{0.3}Ga_{0.7}Se_2$ and 2.090 eV, 2.054 eV, 2.004 eV in $p-TlIn_{0.5}Ga_{0.5}Se_2$ respectively, at 10 K, 140 K and 300 K. There is an abrupt change in the energy peak for $p-TlGaSe_2$ in the temperature range 135-150 K. The values that we obtained from the energy peak change may be at a phase transition temperature.

Key Words: $TlGaSe_2$, $TlIn_{0.3}Ga_{0.7}Se_2$, $TlIn_{0.5}Ga_{0.5}Se_2$ single crystals, absorption, binding energy, phase transition.

Introduction

In recent years growing interest has been paid to the physics of one-dimensional and two-dimensional systems. The latter are represented vividly by $A^{III}B^{III}C_2^{IV}$ crystals, such as $TlGaSe_2$, $TlInSe_2$ and $TlInS_2$. These crystals are characterized by a clearly defined anisotropy of mechanical properties. The compound $TlGaSe_2$ falls under the category of new incomplete-valance $A^{III}B^{III}C_2^{IV}$ -type semiconductors. Some of their electrical, photoelectrical and optical properties are dealt with in a number of publications^[1-9].

The crystals of the system $TlGa_xLn_{1-x}Se_{2x}S_{(1-x)}$ solid solutions ($x=0; 0.1, 0.2, 0.3; 0.6; 0.7; 0.8; 0.9$) were grown by the Bridgman-Stockbarger method in evacuated quartz ampoules^[10]. Measurements of the absorption spectra of $TlGaSe_{2x}S_{(1-x)}$ solid solutions have been obtained at temperatures in the range from 110 to 300 K, with the long-wave region of the edge absorption corresponding to indirect optical transitions^[11] p-type $TlGaSe_2$ crystals were prepared by the Bridgman-Stockbarger method^[11-14]. Large crystals of $TlGaSe_2$ were grown by using the Bridgman method^[15]. The compound crystallises in a monoclinic lattice with the parameters $a=b=7.771 \text{ \AA}$, $c=15.636 \text{ \AA}$, $\beta=100.06$, the densities $\rho = 6.42g/cm^3$, $z=16$ space group $C2/c$ ^[11] and tetragonal lattice with the parameters; $a=b=7.644 \text{ \AA}$, $c=30.832 \text{ \AA}$, $\beta=100.07$ and $z=16$ ^[16]. The monoclinic structure of $TlGaSe_2$ is considered as a deformed tetragonal one. The band structure of the metastable tetragonal modification of $TlGaSe_2$ is calculated by the pseudo potential method^[17]. The energy gap structure and the lattice dynamics of ternary layer $A^{III}B^{III}C_2^{III}$ semiconductor, particularly that of $TlGaSe_2$, which is known to exhibit a strong anisotropy of the electronic and vibrational spectra, has been attracting considerable attention^[12]. The authors^[18] assumed that there exist two different valance bands in $TlGaSe_2$ and the one near the band edge is split. With this fact in mind one could estimate the energy gap value in $TlGaSe_2$.

The long-wavelength tail of the optical absorption in $TlGaSe_2$ at $\alpha = 30 - 150cm^{-1}$ is shown to obey the Urbach rule in the temperature range 4.2 K to 293 K^[19]. The anomalous behavior of the parameters of this rule suggests the presence of two phase transitions in $TlGaSe_2$ at 246 K and 101 K beside the known phase transitions at 120 K and 107 K^[19]. The presence of phase transitions in $TlGaSe_2$ at 246 K and 101 K is also comformed by means of the heat capacity measurements^[19,20]. It has been shown from transition measurements that $TlInS_2$ has an incommensurate phase at 220 K. It has been suggested that $TlGaSe_2$ transforms to a commensurate phase at 120 K. The behavior of an additional fourth material such as Indium (In) in $TlGaSe_2$ and other III-III-VI compounds is interesting. This paper presents new data on the edge absorption in $TlIn_xGa_{(1-x)}Se_2$ mixed crystals inthe 10 to 300 K temperature range. The samples were grown by the modified Bridgman-Stockbarger method.

Basic Equations

The absorption coefficients were obtained from transmission data using the^[21].

$$T = (1 - R)^2 \exp(-A) = (1 - R)^2 \exp(-\alpha d), \quad (1)$$

where R is the reflectivity, (while R is 0.2605^[22]), A is he absorbance, α is the optical absorption coefficient (cm^{-1}) and d is the sample thickness. The optical absorption coefficients determined for all temperatures using the values of R at room temperature by assuming that the temperature change from 10 to 320 K produce a small change in R ^[21]. The multiple reflection and interference fringes were generated by placing the samples with a small angle with respect to the incident beam.

The fundamental absorption edge in most semiconductors follows the exponential low.

Above the exponential tail, the absorption coefficient of semiconductor has been observed to obey the equation

$$\alpha\hbar\omega = B(\hbar\omega - E_g)^n, \quad (2)$$

where $\alpha\hbar\omega$ is the absorption coefficient at angular frequency of $\omega = 2\pi\nu$, ν is a constant and n is an index which can be assumed to have values of 1/2, 3/2, 2 and 3, depending on the nature of electronic transition responsible for the absorption. $n=1/2$ for direct allowed transitions in the high energy part of the spectra, $n=3/2$ for forbidden direct transition; $n=2$ for the indirect allowed transitions in the low energy part of the spectra and $n=3$ for forbidden indirect transitions^[24,25]

The thermal variation of the band gap is given as follows:

$$\frac{dE_g}{dT} = \left(\frac{dE_g}{dT}\right)_{1-ex} + \left(\frac{dE_g}{dT}\right)_{e-p}, \quad (3)$$

where the first term depends on lattice expansion and the second term depends on electron-phonon interaction.

The following empirical expression is often used to describe the temperature dependence of the energy gap^[26]:

$$\begin{aligned} E_{gd}(T) &= E_{gd}(0) - \delta \frac{T^2}{T + \beta} \\ E_{gi}(T) &= E_{gi}(0) - \delta \frac{T^2}{T + \beta} \end{aligned} \quad (4)$$

where $E_g(T)$ is the energy gap, $E_g(0)$ is the energy gap at 0 and δ and β are constants depending on the material. The constant β is approximately equal to the Debye temperature θ_D .

Experimental Procedures

$p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ single crystals were grown by the modified Bridgman-Stockbarger method in our crystal growth laboratory. The melting point of $p-TlGaSe_2$ compound was determined from the phase diagram to be $820 \pm 5^\circ\text{C}$ ^[27]. A sealed quartz ampule was annealed at 1000°C for 10 hours in an outgassing furnace designed in our laboratories. The temperature of quartz ampule was decreased to room temperature over 7 hours. The mix $Tl - In_x - Ga_{(1-x)} - Se_2$ was put into the quartz ampule and the quartz ampule was sealed under a vacuum of 10^{-6} mbar. In order to grow $p-TlGaSe_2$ polycrystal with Bridgman-Stockbarger method, a quartz carbon coated crucible (13 mm in diameter and about 250 mm in length) was used. The crucible was suspended in the middle of a two zone vertical furnace designed in our laboratories. The heated the sample at furnace 205°C for 5 hours and then heated at 310°C for 5 hours. The temperature of the furnace was increased to 920°C for

33 hours. The temperature of the low zone of the furnace was lowered to 550°C at a rate of 6°C/h. Both of the furnace zones cooled to 320°C in 78 hours. The solidified ingot was cooled to room temperature in 52 hours. The prepared $p-TlGaSe_2$ and $TlIn_xGa_{(1-x)}Se_2$ ingot single crystals were 12 mm in diameter and about 70 mm in length. The ingots neither had cracks nor voids on the surface. The freshly cleaved crystals had a mirror-like surface and there was no need for mechanical or chemical polishing treatments. Samples for experiments were cleaved along cleavage planes (001). The X-ray Laue back reflection method was used to test the crystallinity of prepared samples. $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ samples were cleaved from the ingots with a razor blade and cut into 250 μm , 210 μm to 270 μm in thickness and about $1.4 \times 1.4 \text{ mm}^2$, $1.3 \times 1.5 \text{ mm}^2$ and $1.5 \times 1.5 \text{ mm}^2$ in sizes, respectively. The absorption measurements were carried out on $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ samples over the temperature range 10-300 K in 10 K increments.

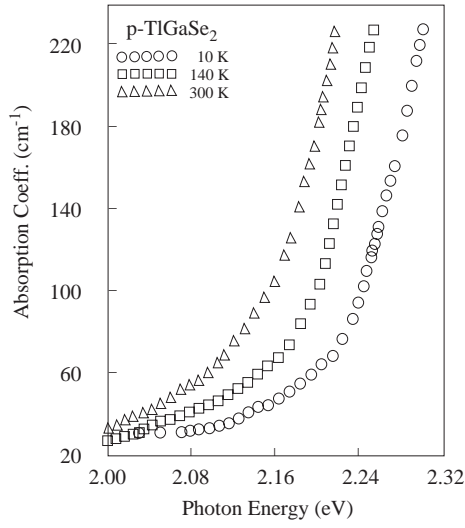
The optical measurements as a function of temperature were made in a closed-cycle He Cryostat. For optical measurements a Perkin Elmer UV/VS Lambda 2S spectrometer was used which works in the range 190-1100 nm. Wavelength accuracy of the spectrometer is better than ± 0.3 nm. Therefore E_g can be calculated with an accuracy better than of about ± 0.0004 eV, considering the wavelength accuracy of our spectrometer.

Results and Discussion

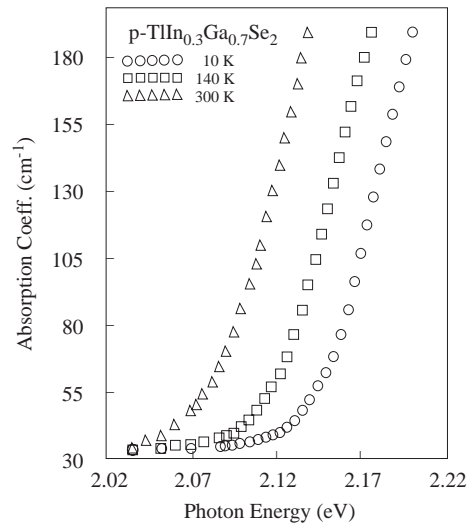
$p-TlGaSe_2$ and $TlIn_xGa_{(1-x)}Se_2$ samples were found to be p-type by hot probe techniques. The absorption coefficient spectra have been obtained from the experimental absorbance values at different sample temperatures using Eq. 1. In Fig.1(a), (b) and (c), we show some typical absorption spectra obtained at sample temperatures of 10 K, 140 K and 300 K. Analyzing the experimental data for $p-TlGaSe_2$, and $TlIn_xGa_{(1-x)}Se_2$ samples, we found that absorption coefficients of $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ were between 25 cm^{-1} and 230 cm^{-1} , 29 cm^{-1} and 191 cm^{-1} and 29 cm^{-1} and 154 cm^{-1} , respectively (Fig.1) in the temperature range of 10-300 K. Note from Fig. 1, that the absorption coefficient of $p-TlGaSe_2$ is smaller than those for $TlIn_xGa_{(1-x)}Se_2$.

The energy gaps of the indirect and direct optical absorptions for $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ are obtained from the upper and lower parts of Figs. 1, using the dependencies $(\alpha^{1/2} - \hbar w)$ and $(\alpha^2 - \hbar w)$ by extrapolation of the dashed straight lines down to $\alpha^2 = 0$ and $\alpha^{1/2} = 0$, respectively (Fig. 2 and 3). The direct absorption spectra of $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ samples versus photon energy at 10 K, 140 K and 300 K are showed in Fig. 4. The direct adsorption edges of investigated compounds shifted considerably when the temperature was changed from 10 to 300 K. As seen Fig. 4, the absorption coefficients $p-TlGaSe_2$ are grater than those of $TlIn_xGa_{(1-x)}Se_2$. The binding energies calculated for $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ are 35.0 meV, 16.5 meV and 14.5 meV, respectively.

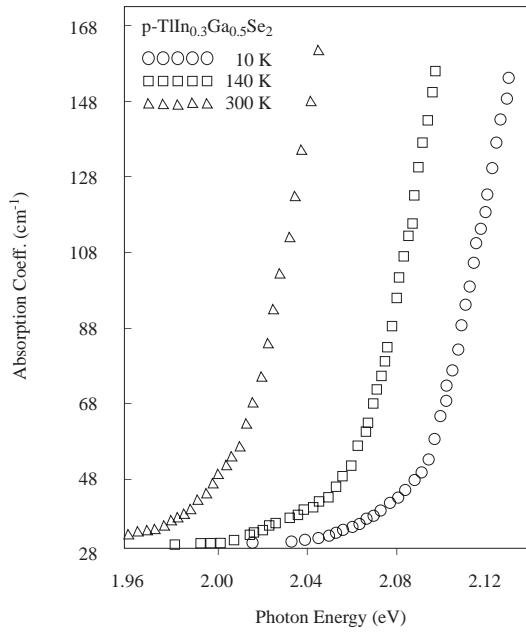
The temperature dependence of the direct and indirect band gap of these samples in



(a). $p - TlGaSe_2$

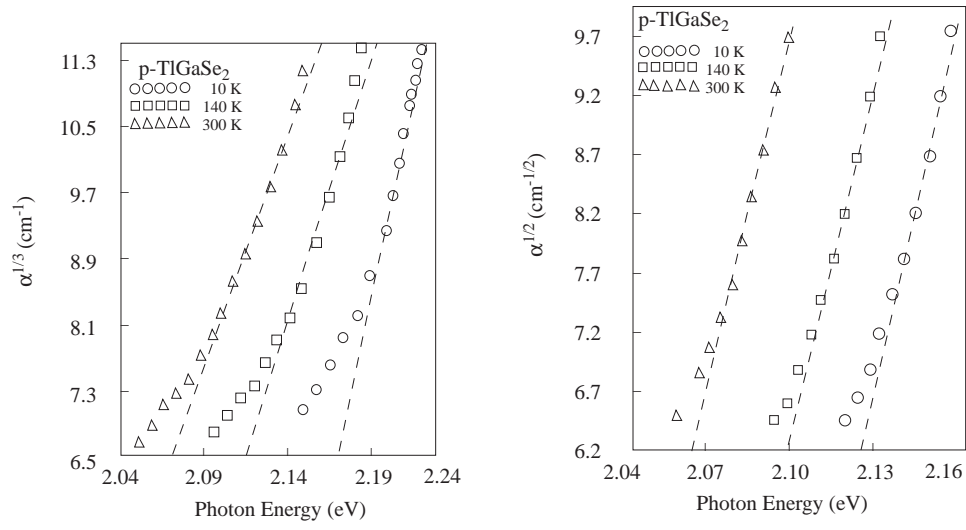


(b). $p - TlIn_{0.3}Ga_{0.7}Se_2$



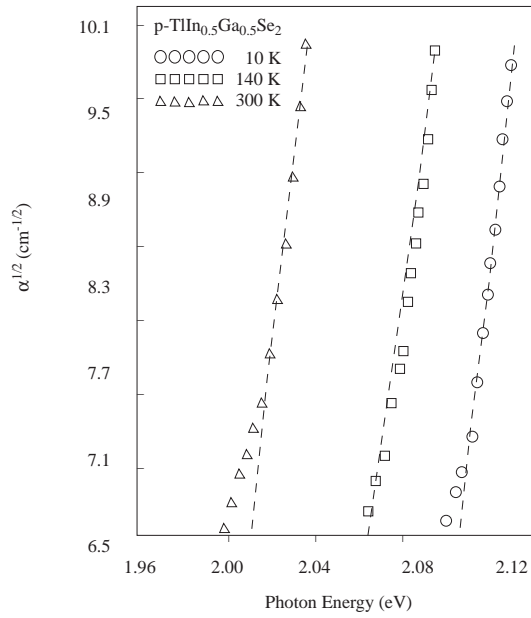
(c). $p - TlIn_{0.5}Ga_{0.5}Se_2$ samples.

Figure 1. Typical absorption spectra obtained at 10 K, 140 K, and 300 K for



(a). $p\text{-TlGaSe}_2$

(b). $p\text{-TlIn}_{0.3}\text{Ga}_{0.7}\text{Se}_2$



(c). $p\text{-TlIn}_{0.5}\text{Ga}_{0.5}\text{Se}_2$ samples.

Figure 2. The indirect absorption spectra versus photon energy for

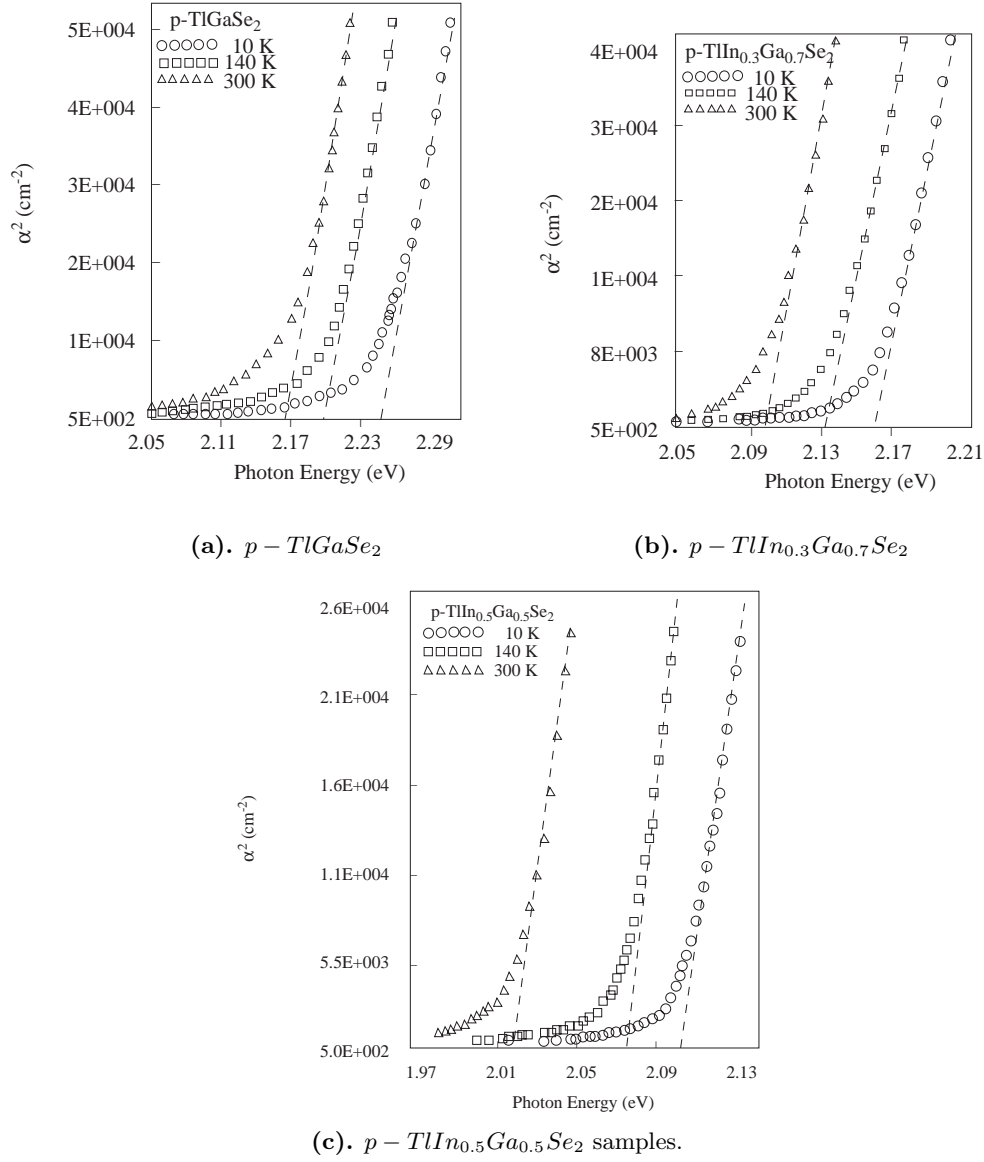


Figure 3. The direct absorption spectra versus photon energy for

the range 10-300 K are shown in

Fig. 5(a), (b) and (c). The absorption spectra for $p - TlGaSe_2$, $p - TlIn_{0.3}Ga_{0.7}Se_2$ and $p - TlIn_{0.5}Ga_{0.5}Se_2$ samples were shown in Fig. 2 and 3 plotted as α^2 versus $\hbar\omega$ gave a dashed straight line indicating the values of direct and indirect band gaps. The direct and indirect band gaps for $p - TlGaSe_2$, $p - TlIn_{0.3}Ga_{0.7}Se_2$ and

$p - TlIn_{0.5}Ga_{0.5}Se_2$ samples are given in Table 1. These results are in agreement with literature^[4,10-12,16]. The temperature dependence of the experimental and theoretical (obtained from Eq. (4)) band gaps is shown in Fig. 5. This curve represents the values of $E_g(T)$ found by Eq. (4). In addition, the temperature dependencies of the band gap for $p - TlGaSe_2$, $p - TlIn_{0.3}Ga_{0.7}Se_2$ and $p - TlIn_{0.5}Ga_{0.5}Se_2$ are presented in Fig. 6. The effect of In on direct and indirect energy gap of $p - TlGaSe_2$ is great (see Fig. 6). It is evident that the direct and indirect energy gap decreases with increasing temperature.

The temperature coefficients were calculated for direct band gap as 3.906×10^{-4} eV/K for $p - TlGaSe_2$, 3.25×10^{-4} eV/K for $p - TlIn_{0.3}Ga_{0.7}Se_2$ and 4.725×10^{-4} eV/K for $p - TlIn_{0.5}Ga_{0.5}Se_2$ from a satisfactory fitting of the experimental curve using the Eq. (4) above and these results agree with results of Guseinov et al.^[29] However, for $p - TlGaSe_2$, $p - TlIn_{0.3}Ga_{0.7}Se_2$ and $p - TlIn_{0.5}Ga_{0.5}Se_2$ samples, β were calculated as 145 K, 165 K and 159 K, respectively. The temperature coefficients were calculated the same way for indirect band gap as 6.245×10^{-4} eV/K for $p - TlGaSe_2$, 3.385×10^{-4} eV/K for $p - TlIn_{0.3}Ga_{0.7}Se_2$ and 4.325×10^{-4} eV/K for $p - TlIn_{0.5}Ga_{0.5}Se_2$ and for $p - TlGaSe_2$, $p - TlIn_{0.3}Ga_{0.7}Se_2$ and $p - TlIn_{0.5}Ga_{0.5}Se_2$ samples, β were calculated as 160 K, 162 K and 157 K, respectively.

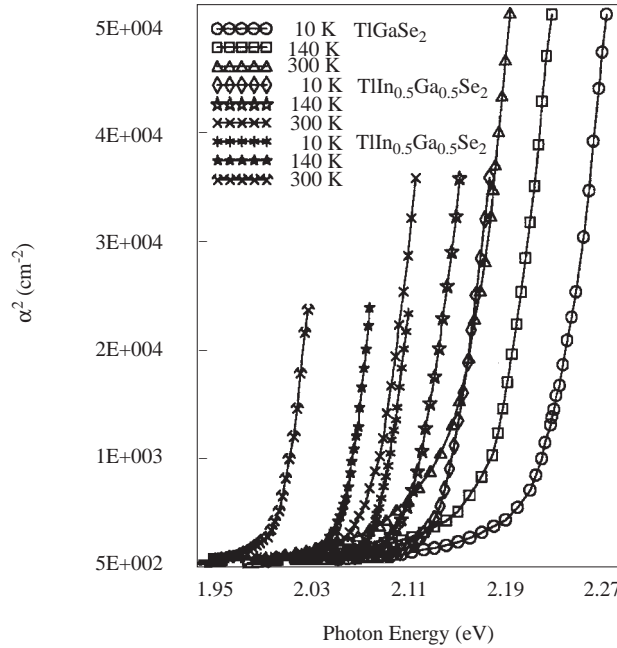


Figure 4. The absorption spectra versus photon energy for $p - TlGaSe_2$, $p - TlIn_{0.3}Ga_{0.7}Se_2$, $p - TlIn_{0.5}Ga_{0.5}Se_2$ samples.

Table 1. Direct and indirect band gap values of $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$ samples versus sample temperatures.

Samples	Sample temperatures	E_{gd} (eV)	E_{gi} (eV)
$p-TlGaSe_2$	10 K	2.244	2.196
	140 K	2.195	2.127
	300 K	2.164	2.073
$p-TlIn_{0.3}Ga_{0.7}Se_2$	10 K	2.158	2.130
	140 K	2.131	2.101
	300 K	2.098	2.064
$p-TlIn_{0.5}Ga_{0.5}Se_2$	10 K	2.107	2.090
	140 K	2.075	2.054
	300 K	2.019	2.004

The dE_g/dT sensitively depends upon the contributing terms positively and negatively. It depends both on lattice expansion and electron-phonon interaction. The sign of the temperature coefficient given in Eq. 3 neither depend solely on the sign of first term nor that of the second term. The total temperature coefficient depends upon the nature of the system and especially on the electronic wave functions. The positive temperature variation is not surprising. There are similar examples in the IV-VI compounds and this variation depends on the specific properties of the electronic states of the band extremely and the ordering of the levels relatively^[29]. The temperature coefficients of indirect and direct band gaps of $p-TlGaSe_2$ were found to have negative signs which suggest that the electron-phonon interaction term is larger than the lattice expansion contribution. Phase transitions in $p-TlGaSe_2$ single crystals have been investigated by analyzing the optical energy gap^[30].

For $p-TlGaSe_2$, $p-TlIn_{0.3}Ga_{0.7}Se_2$ and $p-TlIn_{0.5}Ga_{0.5}Se_2$, the direct and indirect energy gaps decrease towards lower values as the temperature increases and the temperature coefficient has a negative sign (Fig. 6). Such a shift is generally a typical property for crystals having layered structures^[23]. There is an abrupt change in the energy peak for $p-TlGaSe_2$ in the temperature ranges 135-150 K. Our values obtained from the energy peak change may be a phase transition temperature. The maximum values of the phase transition temperature obtained from heat-capacity measurements are 340 K, 253 K, 117.2 K, 108.9 K, 106 K and 101 K^[30]. It should be noted that the phase transition found in at 250 K^[19] was blurred, whereas in this study, a sharp heat-capacity peak has been obtained for a $p-TlGaSe_2$ single crystal. The values that we obtained from the energy peak change may be phase transition temperature with respect to above authors.

In conclusion, these results are in good agreement with literature^[4,13,17,19,23,28-31]. It is seen from the experimental absorption results that the band gap of $p-TlGaSe_2$ is greater than those of $TlIn_xGa_{(1-x)}Se_2$. Addition of indium causes the appearance of a new band gap energy and reduces the intensity of absorption peak in $p-TlGaSe_2$. As seen Figure 4, the slopes of the absorption peaks in $p-TlIn_xGa_{(1-x)}Se_2$ are smaller

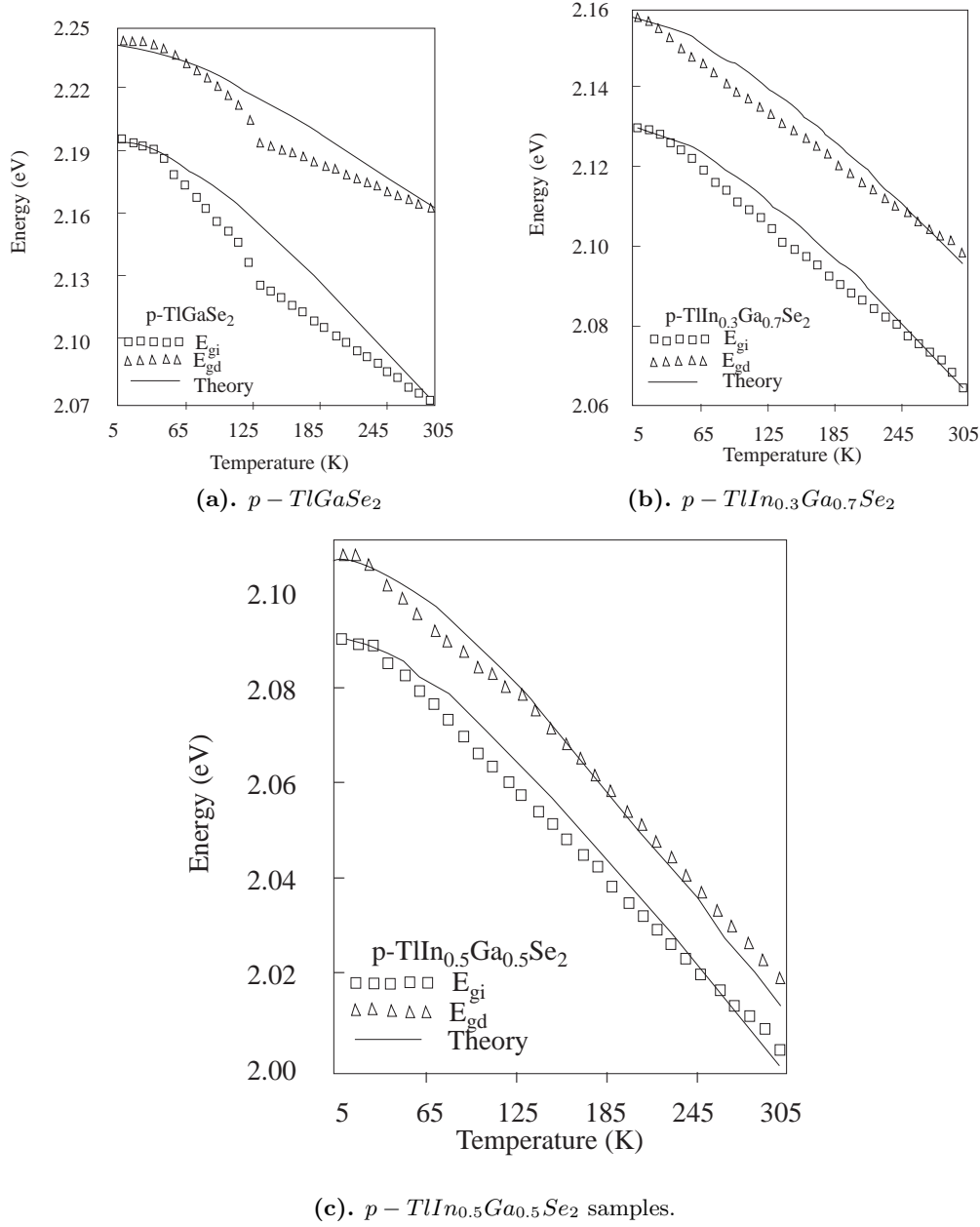


Figure 5. The temperature dependence of the direct and indirect band gap for than that of $p-TlGaSe_2$. The reason of these decreases in the slops is due to disappearing defects existing in the original material by adding indium (Figure 4). Indium atoms in

p - $TlGaSe_2$ have reduced the amount of defects. These defects are optically active in band gap of p - $TlGaSe_2$. The peaks which give the absorption coefficient in $TlIn_xGa_{(1-x)}Se_2$ are smaller than that of p - $TlGaSe_2$ (Figure 4). This result shows that there are defects in p - $TlGaSe_2$ single crystal and these defects in p - $TlGaSe_2$ are cleaned by adding indium. These are expected results, due to the behavior of the indium atoms in the p - $TlGaSe_2$. That is to say, Indium atoms substitute Gallium atoms.

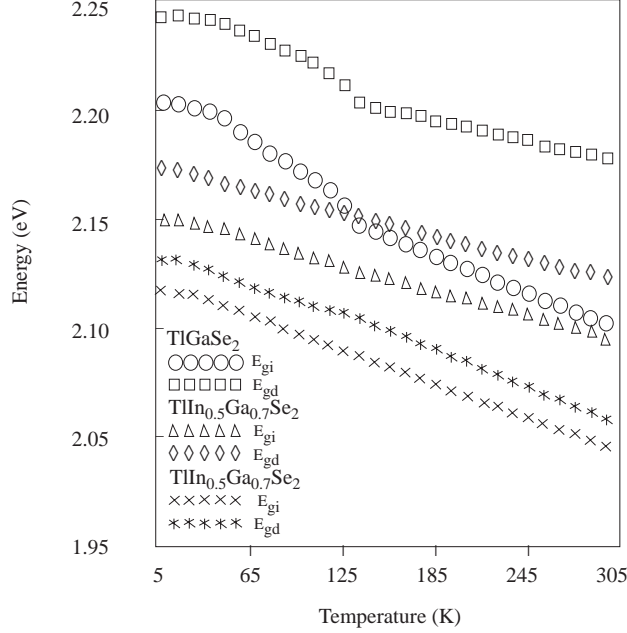


Figure 6. The temperature dependence of the direct and indirect band gap for p - $TlGaSe_2$, p - $TlIn_{0.3}Ga_{0.7}Se_2$, p - $TlIn_{0.5}Ga_{0.5}Se_2$ samples.

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