

INVESTIGATION OF THE OPTICAL ABSORPTION
SPECTRA OF THIN EPITAXIAL LEAD SELENIDE
LAYERS APPROACHING THE NANOSCALE THICKNESS

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Abstract

By sophisticated treatment of the optical absorption spectra of thin epitaxial layers of lead selenide of different thickness approaching the nanorange, the presence of similar additional absorption for all layers was revealed. Such absorption was detected between the absorption edge and the absorption on free current carriers increasing with the wavelength. When constructing the absorption spectra by the experimentally investigated transmission spectra, the criteria of their treatment were strengthened by considerations for the absorption relation between the layers with low and high concentrations of current carriers, and from which level of absorption the transitions related to the absorption edge were to be considered. This resulted in the fact that, upon straightening the squared absorption coefficients, the obtained values of the forbidden gap width were in good correlation with the results obtained with the corresponding deformation of thin layers, and the contribution of the additional absorption to their determination was negligible.

1. Introduction

To suggest the use of strained epitaxial layers of IV–VI semiconductors for designing of high-sensitive and high-temperature IR photodetectors [1], it is essential to determine precisely the forbidden gap width of these layers. The additional absorption was detected in thin layers, especially in the ones $\geq 1 \mu\text{m}$ in thickness, in previous works [2, 3]. In recent works [4, 5], it was shown that the additional absorption between the absorption edge and the absorption on free carriers increasing with the wavelength has no effect on the determination of the forbidden gap width [6]. It is of interest to study such additional absorption in strained layers $< 200 \text{ nm}$ in thickness, which are promising for application. Besides, it is desirable to supplement the criteria of treatment of the optical absorption spectra for correct determination of the forbidden gap width of thin layers by addition of new verified considerations.

2. Objects and methods of investigation

The epitaxial PbSe layers were grown on KCl, NaCl and BaF₂ substrates by molecular epitaxy with a “hot wall”. The temperature of the source of epitaxy representing polycrystalline PbSe was over the range of 450 – 510 °C, while the temperature of the substrate was over the range of 240 – 320 °C. At the distance between the open tip of a quartz ampoule with the source of epitaxy and the substrate equal to 10 – 12 mm, the layers ~ 1 μm thick grew for 1 – 2 h, and the ones < 200 nm thick – for tens seconds [7].

The deformation of epitaxial layers $\varepsilon = (a_1 - a_s)/a_s$ was determined by the results of X-ray measurements of lattice constants by a symmetric scheme. Radiation of CoK α ($\lambda = 1.7889 \text{ \AA}$) was used.

The thickness of layers was determined by comparing the intensity of reflection from the layer-coated substrate and that of the uncoated one. The structure of the layers was also studied by the X-ray method.

The layers thicker than 20 nm grew uniform and monocrystalline. The optical transmission spectra were detected at $T = 300 \text{ K}$ by using the prism-diffraction spectrophotometer SPECORD-75IR. Precision measurements of the transmission of layers were carried out by using the masks in measurement and reference modes.

3. Results and discussion

The PbSe layers grown on the NaCl and KCl substrates oriented in the (100) direction were chosen for the investigation. The layers differed in thickness by about an order of magnitude, 1.4 μm and 180 nm for layers SL-592 and SL-578, respectively. **Table 1** gives the data on absorption coefficients α and $\alpha_{\text{fr.car.}}$, their squared remainder and their squared product by coefficient $1/\gamma$ considering for the degeneracy the concentration of current carriers in the layers of about 10^{19} cm^{-3} . The lattice constants of layers SL-592 and SL-578 made up $a = 6.122$ and 6.149 \AA , respectively. This means that layer SL-592 is somewhat contracted (lattice constants: $a_{\text{PbSe}} = 6.126 \text{ \AA}$ and $a_{\text{NaCl}} = 5.640 \text{ \AA}$), while layer SL-578 is stretched ($a_{\text{KCl}} = 6.290 \text{ \AA}$) with “negative” deformation $\varepsilon = 0.0037$.

Table 1. Data on absorption coefficients, their squared values and squared products by $1/\gamma$ for layer SL-592.

$h\nu$, eV	α , cm^{-1}	$\alpha_{\text{fr.car.}}$, cm^{-1}	$\alpha_{\text{fr.car.}}^2$, cm^{-2}	$\alpha^* = \alpha - \alpha_{\text{fr.car.}}$, cm^{-1}	$\alpha^{*2} = (\alpha - \alpha_{\text{fr.car.}})^2$, cm^{-2}	$1/\gamma$	$\alpha'^2 = (\alpha^*/\gamma)^2$, cm^{-2}
0.167	7419	7419	5.50E+07	0	5.5E+07	20.88	3.44E-02
0.222	5342	2798	7.83E+06	2544	6.5E+06	9.56	5.92E+08
0.279	5976	1484	2.20E+06	4493	2.0E+07	4.55	4.17E+08
0.334	6955	931	8.66E+05	6024	4.6E+07	2.53	2.32E+08
0.360	7669	768	5.89E+05	6901	4.8E+07	2.02	1.92E+08
0.384	10137	646	4.18E+05	9491	9.0E+07	1.70	2.61E+08
0.417	11531	542	2.94E+05	10989	1.2E+08	1.42	2.42E+08

The data on the absorption coefficients were obtained by solving the equation linking transmission, reflection coefficients and propagation constants with the absorption coefficient according to the model of the Fabry–Perot interferometer [8]. The corresponding initial data were taken from work [3] where the analysis had been performed by interference maxima in the transmission spectrum.

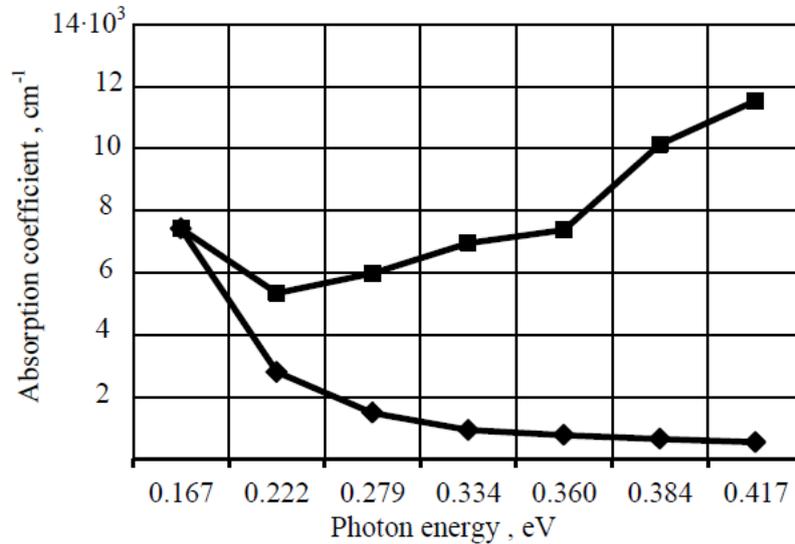


Figure 1. Spectra dependence of the total absorption coefficient α (■) and the absorption coefficient on free carriers $\alpha_{fr.car.}$ (◆) for layer SL-592.

Figure 1 shows the spectral dependences of the total absorption coefficient α obtained by the equation and the absorption coefficient on free carriers $\alpha_{fr.car.}$. The latter was obtained from the condition of equality of absorption α and absorption $\alpha_{fr.car.}$ ($\alpha_{fr.car.} \sim \lambda^2$) at the wavelength corresponding to the long-wavelength maximum (five maxima were considered).

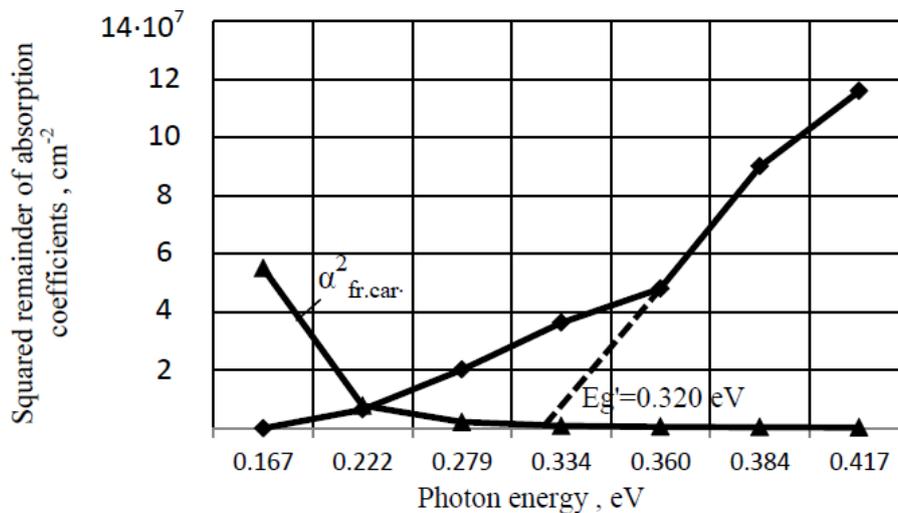


Figure 2. Dependence of the squared remainder of absorption coefficients $\alpha^* = \alpha - \alpha_{fr.car.}$ on the photon energy $h\nu$ for layer SL-592.

In **Figure 2**, the relations $(\alpha - \alpha_{\text{fr.car.}})^2 = f(h\nu)$ and $\alpha_{\text{fr.car.}}^2 = f(h\nu)$ are shown. By extrapolating the $(\alpha - \alpha_{\text{fr.car.}})^2 = f(h\nu)$ to zero energy of the photon beginning from the squared absorption coefficients $\geq 5 \cdot 10^7 \text{ cm}^{-2}$, the absorption edge making up 0.320 eV was determined. At close values of the effective mass of electrons and holes, the above mentioned value of the absorption edge corresponds to the sum $2E_{\text{F}} + E_{\text{g}}$ (where E_{F} is the Fermi level at the concentration of current carriers $1 \cdot 10^{19} \text{ cm}^{-3}$ at $T = 300 \text{ K}$).

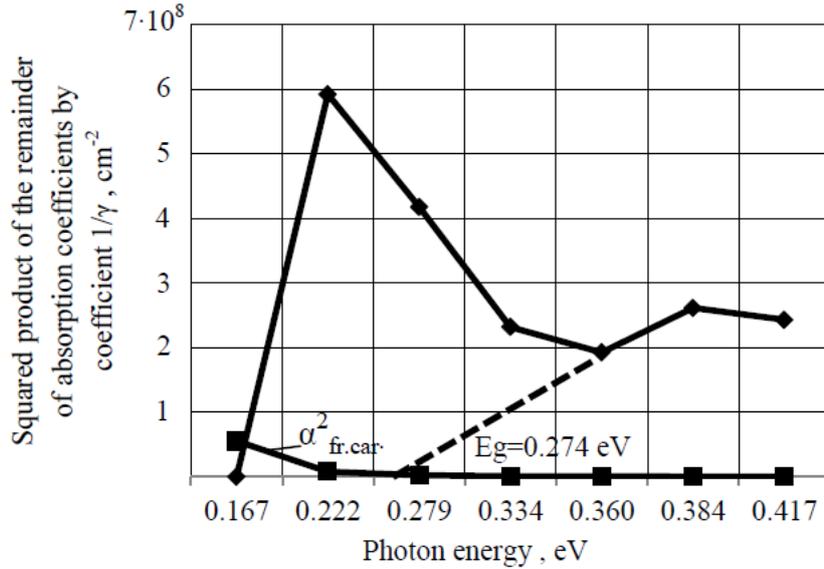


Figure 3. Dependence of the squared product of the remainder of absorption coefficients $\alpha^* = \alpha - \alpha_{\text{fr.car.}}$ by coefficient $1/\gamma$ (♦) on the photon energy $h\nu$ for layer SL-592.

The forbidden gap width E_{g} was determined by extrapolating the relation $((\alpha - \alpha_{\text{fr.car.}})/\gamma)^2 = f(h\nu)$ to the zero energy of the photon. The coefficient γ considering for degeneration was calculated at the Fermi level $E_{\text{F}} = 0.03 \text{ eV}$. **Figure 3** shows the extrapolation of this function, and the E_{g} value makes up 0.274 eV.

Unlike work [3], where, upon growing of the PbSe layers on the KCl substrate, the forbidden gap width made up 0.286 eV in the layers $\sim 1 \mu\text{m}$ thick. In our case its smaller value is related to somewhat contraction of the layer.

From **Figure 3** it is also obvious that multiplication by the coefficient $1/\gamma$ at the energy less than E_{g} yields an improbable result – transitions of current carriers to the energy levels with a maximum at $\sim 0.22 \text{ eV}$. Hence, the squared remainder $\alpha^{*2} = (\alpha - \alpha_{\text{fr.car.}})^2$ at the energy lower than E_{g} shows the presence of additional absorption. From **Table 1** and **Figure 1**, it is seen that the additional absorption above the absorption by free carriers exceeds the latter by $(2.5 - 3.0) \cdot 10^3 \text{ cm}^{-1}$.

Like **Table 1**, **Table 2** gives the data on the total absorption coefficient and their remainder $\alpha^* = \alpha - \alpha_{fr.car.}$, and also the corresponding squared values the spectrum under study and their multiplication by the coefficient $1/\gamma$ for layer SL-578. There is as well given the transmission which was used in the solution of the equation by the model of the Fabry-Perot interferometer for determining the coefficient α .

Table 2. Data on absorption coefficients, their squared values and square products by $1/\gamma$ for layer SL-578.

$h\nu$, eV	α , cm^{-1}	$\alpha_{fr.car.}$, cm^{-1}	$\alpha_{fr.car.}^2$, cm^{-2}	$\alpha^* = \alpha - \alpha_{fr.car.}$, cm^{-1}	$\alpha^{*2} = (\alpha - \alpha_{fr.car.})^2$, cm^{-2}	$1/\gamma$	$\alpha'^2 = (\alpha^*/\gamma)^2$, cm^{-2}	T_{max}
0.248	42322	42322	1.80E+09	0	1.01E-01	6.727	4.55E+00	0.205
0.261	39104	36756	1.35E+09	2347	5.51E+06	5.681	1.78E+08	0.197
0.276	33304	31791	1.01E+09	1513	2.29E+06	4.742	5.15E+07	0.195
0.292	28571	27363	7.49E+08	1208	1.46E+06	3.913	2.23E+07	0.189
0.310	25469	23418	5.48E+08	2051	4.21E+06	3.198	4.30E+07	0.179
0.335	22426	19430	3.78E+08	2996	8.98E+06	2.491	5.57E+07	0.172
0.347	21828	17846	3.18E+08	3982	1.59E+07	2.237	7.93E+07	0.169
0.353	22472	17096	2.92E+08	5376	2.89E+07	2.125	1.30E+08	0.166
0.360	23357	16377	2.68E+08	6979	4.87E+07	2.021	1.99E+08	0.163
0.366	23585	15738	2.48E+08	7847	6.16E+07	1.929	2.29E+08	0.162
0.378	24148	14568	2.12E+08	9579	9.18E+07	1.767	2.87E+08	0.161
0.381	24830	14294	2.04E+08	10536	1.11E+08	1.731	3.33E+08	0.160
0.397	27359	13068	1.71E+08	14291	2.04E+08	1.575	5.07E+08	0.158

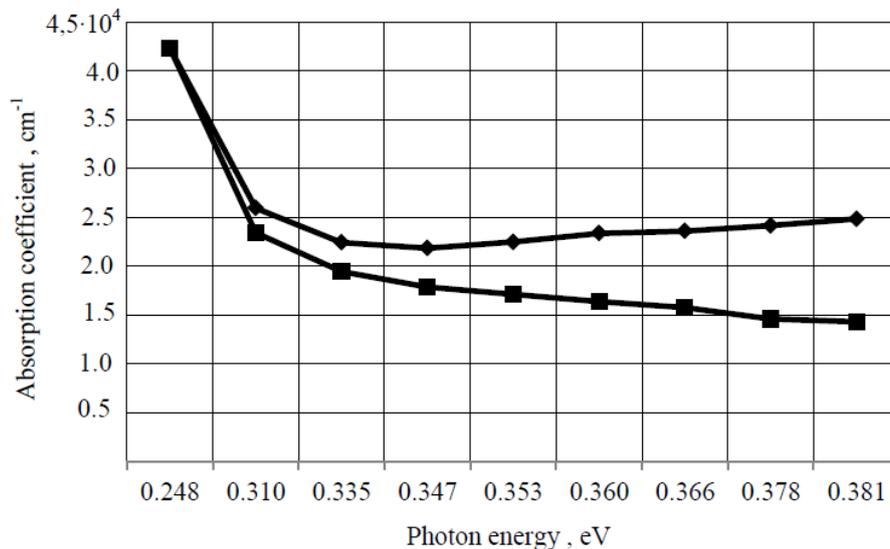


Figure 4. Spectra dependence of the total absorption coefficient α (\blacklozenge) and the absorption coefficient on free carriers $\alpha_{fr.car.}$ (\blacksquare) for layer SL-578.

In **Figure 4**, there are shown the spectral dependences of the total absorption coefficient α and coefficient of the absorption on free carriers $\alpha_{fr.car.}$. As in **Figure 1**, here is observed the

minimum α , which is caused by an increase in the band absorption with increasing energy and the reverse course of absorption on free carriers.

Figure 5 shows the relation $(\alpha - \alpha_{fr.car.})^2 = f(h\nu)$ for layer SL-578. The extrapolation to the zero energy of the photon results in the value 0.356 eV. In this case, in the straightening, the points with the values of squared remainders of absorption coefficients are considered for layer SL-578.

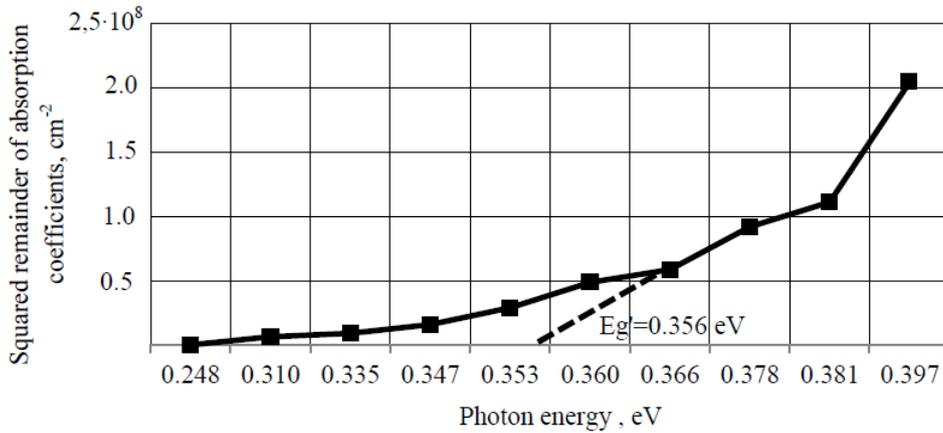


Figure 5. Dependence of the squared remainder of absorption coefficients $\alpha^* = \alpha - \alpha_{fr.car.}$ on the photon energy $h\nu$ for layer SL-578.

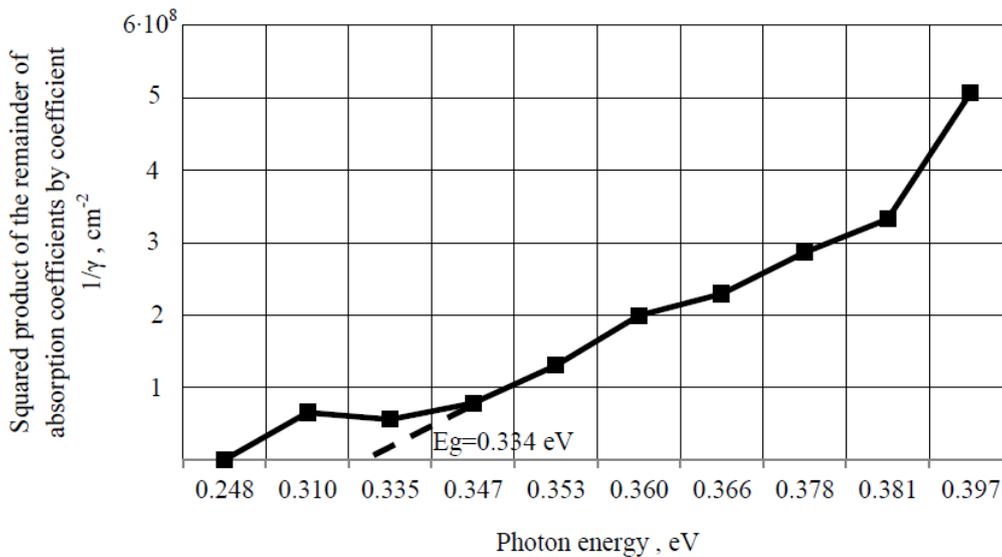


Figure 6. Dependence of the squared product of the remainder of absorption coefficients $\alpha^* = \alpha - \alpha_{fr.car.}$ by coefficient $1/\gamma$ on the photon energy $h\nu$ for layer SL-578.

Figure 6 shows the relation $((\alpha - \alpha_{fr.car.})/\gamma)^2 = f(h\nu)$. In the calculation of the coefficient γ [2], the Fermi level was equal to $E_F = 0.03$ eV, identical to that for layer SL-592.

The extrapolation of this relation to the zero energy of the photon results in the value $E_g = 0.334$ eV. From these extrapolations, it can be inferred that the value of $2E_F$ makes up 0.022 eV, hence $E_F = 0.011$ eV. Therefore, hereinafter there is good reason to take a smaller

value of E_F for calculation of the coefficient γ for such width of the forbidden gap and deformation $\varepsilon = 0.0037$. From **Table 2** and **Figure 4**, it is evident that the additional absorption over the spectral region from 0.310 to 0.334 eV also exceeds the absorption on free carriers by $(2.5 - 3.0) \cdot 10^3 \text{ cm}^{-1}$. Hence, the additional absorption in the epitaxial layers of different thickness has close values and is likely to be associated with virtual transitions between different branches of allowed bands. This fact received mention in work [9] as well.

It should be noted that, according to work [10], with the energy exceeding the forbidden gap width by $2.1E_F$, the absorption coefficient in the pure IV–VI semiconductor with a low concentration of current carriers increases almost twice as much as compared to the degenerate semiconductor. From **Tables 1** and **2**, it is seen that, at $1/\gamma \approx 2.1$, the energy with which this condition is fulfilled for layers SL–592 and SL–578 makes up 0.320 and 0.356 eV, respectively. This means that, with the energy exceeding E_g by $2E_F$, not only the absorption edge can be determined, but also the character of its growth in the pure semiconductor can be assessed.

4. Conclusion

By using the data of our previous works and by the optical transmission spectra by the model of the Fabry–Perot interferometer, we determined the total absorption coefficient α over the spectral region of 3 – 5 μm for the epitaxial PbSe layers grown on the NaCl and KCl substrates, the thickness of layers making up 1.4 μm and 180 nm, respectively. We sequentially determined the absorption coefficients on free carriers $\alpha_{\text{fr.car.}}$, their remainder $\alpha^* = \alpha - \alpha_{\text{fr.car.}}$ and also the value of $\alpha' = \alpha^*/\gamma$ for considering of the degeneration in the layers. By extrapolating the relation $((\alpha - \alpha_{\text{fr.car.}})/\gamma)^2 = f(h\nu)$ to the zero energy of the photon, the forbidden gap width was determined. It made up 0.274 and 0.334 eV, respectively, which was in good correlation with the deformation in the layers. The additional absorption was revealed between the absorption edge and the absorption on free carriers increasing with the wavelength. This absorption had equal values for the layers of different thickness.

It was found that additional absorption has little or no effect on the determination of the value of the of forbidden gap width. The 2-fold increase relation of the absorption with the energy twice as high as the Fermi level E_F in lead selenide could be an appropriate checking factor of correct determination of the forbidden gap width in the degenerate layer [11, 12].

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