

Analysis of the absorption spectra of epitaxial lead telluride and lead selenide layers

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Abstract. *By processing the optical transmission spectra, the index of refraction, and reflection and absorption coefficients of epitaxial PbTe and PbSe layers (grown on BaF₂ (111) and NaCl (100) substrates, respectively, by molecular epitaxy with a “hot wall”) were measured sequentially by the elaborated scheme. The concentration of current carriers varied in the range from 10¹⁸cm⁻³ to 10¹⁹cm⁻³. The analysis of the obtained absorption spectra was performed, and the squared absorption coefficients were straightened with the purpose to determine the forbidden gap width. Special emphasis is placed on the effect of high concentration of the current carriers on the type of dispersion of the index of refraction, the absorption by free carriers and the degeneration of the current carriers. An approach to correct determination of the forbidden gap width with the formal transition from the degenerated semiconductor to the no degenerate one by modification of the absorption coefficient was worked out. With limitation of the range of energy at which the absorption undergoes changes, additional absorption between the absorption edge and the absorption by free carriers increasing with the wavelength was revealed in the PbSe layers. For the PbTe layers with a lower concentration of current carriers, the additional absorption was found by the model analysis of transmission spectra at zero absorption.*

Keywords: concentration of current carriers, model analysis, straightening of squared coefficients, forbidden gap width, additional absorption.

I. INTRODUCTION

The investigation of the absorption spectra of narrow-band IV-VI semiconductors has assumed typically due to the formation of the dielectric state in them [1],[2]. When growing the IV-VI semiconductor layers doped with impurities such as Cr or In on dielectric substrates with larger lattice constants, the forbidden gap width increases, while the impurity levels shift into the forbidden gap. The more expands the forbidden gap, the deeper shifts the impurity level, and the compensation of electrically active defects and impurities becomes efficient. Hence a comprehensive study on the forbidden gap width of epitaxial IV-VI semiconductor layers grown on dielectric substrates is of considerable importance. There is actual experience in studying the absorption spectra of various compositions of these semiconductors [3]-[5]. In work [3], the forbidden gap width at direct and indirect transitions in PbS was established. In

work [4], PbSe polycrystals with grains of different sizes were investigated, and the corresponding forbidden gap width was determined. Similar investigation of solid PbSe_{1-x}Te_x solutions [5] showed that the forbidden gap width monotonously varied with the composition.

II. OBJECTS AND METHODS OF INVESTIGATION

We constructed the absorption spectra by the results of the analysis of experimentally obtained transmission spectra of the epitaxial layers of lead telluride and lead selenide grown by molecular epitaxy with a “hot wall”. The lead telluride layers were grown on BaF₂ (111) substrates, while the lead selenide layers – on NaCl(100) ones. Polycrystalline PbTe and PbSe were used as the sources of epitaxy. The temperature of the PbTe and PbSe sources was 530°C and 510°C, respectively, the temperature of the BaF₂ and NaCl substrates was 360°C and 240°C, respectively. The growth rate of the layers varied in the range from 1 to 2 μm/hour. We selected for investigation the following layers: the PbTe layer 2.5μm thick with the lattice constant 6.459Å, and the PbSe layer 1.4 μm thick with the lattice constant 6.122Å, which was somewhat lower than in the monocrystal (a=6.126Å), i.e. the PbSe layer was somewhat strained. The concentration of current carriers in the PbTe layer made up 1.2·10¹⁸cm⁻³, and PbSe-1.5·10¹⁹cm⁻³. The thickness and lattice constants of the layers were determined by recording the X-ray diffraction lines of reflection from crystallographic planes (200), (400) and (600). The optical transmission spectra were recorded with the help of double-beam prism-diffraction spectrophotometer SPECORD-751R over the wave number range 4000-400cm⁻¹(the corresponding wavelength region – 2.5-25μm). The calibration of zero and hundred percent transmission lines was performed using the masks and correcting diaphragms. For the PbTe layer of the given thickness, 6 interference maxima were recorded over the wavelength range from 4 to 11μm, while, for the PbSe layer of the given thickness, 5 interference maxima were recorded over the wavelength range from 3 to 9μm.

III. RESULTS AND ANALYSIS

The optical absorption spectra of lead telluride and lead selenide were constructed by processing the transmission

spectra and determining sequentially the index of refraction, and the absorption and reflection coefficients.

Table 1 . Data on transmission T , indices of refraction N and calculated values of coefficients r₁ and r₂ for the PbTe layer (d=2.5µm)

N ₀	hν, eV	N _{PbTe}	N _{BaF₂}	r ₁	r ₂	T _{exp.}	T ₀
1	0.123	2.020	1.399	0.338	0.181	0.500	0.970
2	0.164	3.030	1.427	0.504	0.354	0.505	0.970
3	0.202	3.681	1.443	0.573	0.437	0.490	0.970
4	0.242	4.103	1.451	0.608	0.477	0.450	0.970
5	0.278	4.464	1.452	0.634	0.509	0.425	0.970
6	0.312	4.771	1.455	0.653	0.533	0.390	0.970
7	0.335	4.977	1.461	0.665	0.546	0.145	0.301
8	0.360	5.201	1.465	0.677	0.560	0.095	0.192
9	0.384	5.425	1.470	0.689	0.572	0.080	0.188

The spectra were processed based on a model of the Fabry-Perot interferometer [8]: heterostructures PbTe/BaF₂

and PbSe/NaCl were considered as resonators. The absorption coefficient α was determined by the known values of transmission T and the calculated values of the reflection coefficient by the amplitude :

$$r_1 = (N_1 - N_{air}) / (N_1 + N_{air}) , r_2 = (N_1 - N_{sub.}) / (N_1 + N_{sub.})$$

by the relation

$$T = (1 - r_1^2) (1 - r_2^2) \exp(2\beta_1 d) / (1 - R)^2 + 4R \sin^2(\beta d) \quad (1)$$

where N₁ and N_{sub.} are the indices of refraction of the layers and substrates, R = r₁ r₂ exp(2β₁d) is the reflection coefficient by power, β = 2πN / λ being the real part of the complex constant of propagation, β₁ = -α/2. For determination of the absorption coefficient α we introduced notations

$$y = \exp(2\beta_1 d) = \exp(-\alpha d) . \text{ From } E_q \text{ (1) , we get:}$$

$$T(r_1 r_2)^2 y^2 - \{(1 - r_1^2) (1 - r_2^2) + T r_1 r_2 - 4 T r_1 r_2 \sin^2 2\pi N d / \lambda\} y + T = 0 \quad (2)$$

The transmission coefficients of the layers were determined by interference maxima by the relation 2Nd = m λ.

Table 2 . Calculated values of absorption coefficients α , differences α - α_{fr.car.} , their squared values (α - α_{fr.car.})² and [(α - α_{fr.car.})hv]² for the PbTe layer

N ₀	hν, eV	α, cm ⁻¹	α _{fr.car.} , cm ⁻¹	α* = α - α _{fr.car.} , cm ⁻¹	(α*) ² , cm ⁻²	(α*·hv) ² , cm ⁻² eV ²
1	0.123	2426	2426	0	6.62E+08	0
2	0.164	1950	910	1041	1.08E+06	2.90E+04
3	0.202	1801	491	1310	1.72E+06	7.01E+04
4	0.242	1896	308	1589	2.52E+06	1.48E+05
5	0.278	1930	214	1716	2.94E+06	2.27E+05
6	0.312	2054	159	1895	3.59E+06	3.49E+05
7	0.335	3593	132	3460	1.20E+07	1.34E+06
8	0.360	4449	110	4339	1.88E+07	2.43E+06
9	0.384	5125	92	5032	2.53E+07	3.74E+06

The transmission coefficients of the layers were determined by interference maxima by the relation 2Nd = mλ. The correctness of selection of the order of interference was checked by three conditions : transmission at zero absorption for each maximum,

fulfilment of the relation λ/2N < d (for nonextremal points) and consideration for the cone-shaped relationship of the indices of refraction .

The absorption spectra of epitaxial PbTe and PbSe layers are presented and analyzed below .

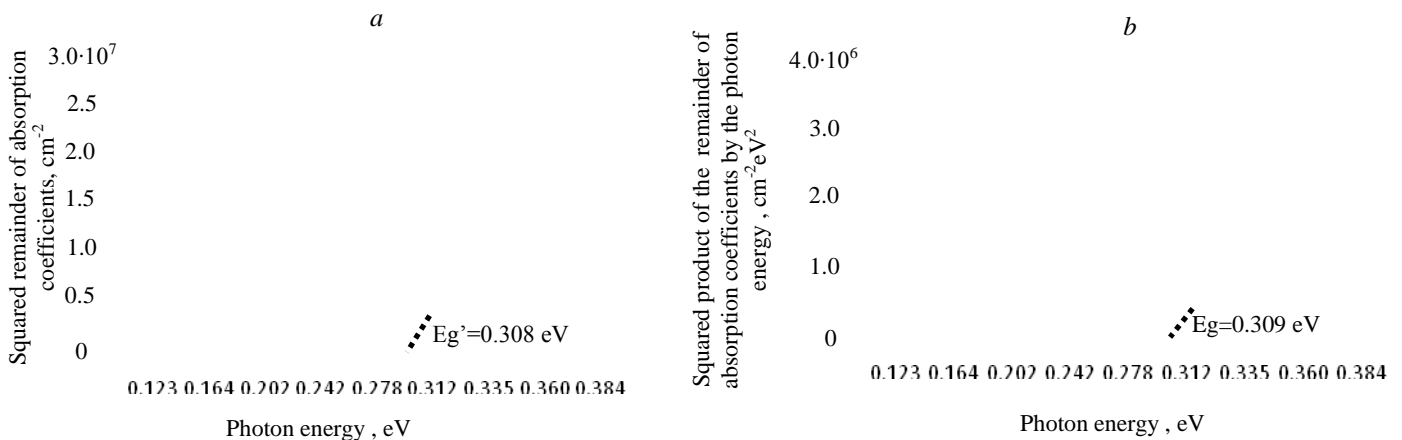


Fig 1a,b. Photon energy dependence of the squared remainder of absorption coefficients (a) and the squared product of the remainder of absorption coefficients by the photon energy (b) for the PbTe layer

The correctness of selection of the order of interference was checked by three conditions: transmission at zero absorption for each maximum, fulfillment of the relation λ/2N < d (for nonextremal points) and consideration for the

cone-shaped relationship of the indices of refraction. The absorption spectra of epitaxial PbTe and PbSe layers are presented and analyzed below. Table 1 gives the values of the

indices of refraction N_{PbTe} and N_{BaF_2} corresponding to the energy at interference maxima of the transmission spectrum, the calculated values of reflection coefficients r_1 and r_2 , transmission T_{exp} , and the calculated transmission at zero absorption T_0 . Three points are added in the short-wave interval. The values of the indices of refraction for them were studied by considering the dispersion relation $N^2=f(\lambda^2)$ for the PbTe layer at the concentration of current carriers $n=1.2 \cdot 10^{18} \text{cm}^{-3}$.

The values of the indices of refraction given in Table 1 satisfy all three conditions listed above. The order of interference was determined correctly and there are no "false" maxima, though the values of the determined indices of refraction are less than the ones given in work [9] and are characterized by more clearly defined dispersion. In that work it was also noted that such a tendency was manifested at the layer thickness less than $6 \mu\text{m}$.

Table 3 . Data on the real and correct values of transmission T under zero absorption α^* at $\lambda > 3.7 \mu\text{m}$ ($< 0.334 \text{eV}$) for the PbTe layer

N_0	$h\nu, \text{eV}$	α, cm^{-1}	$\alpha_{fr.car}, \text{cm}^{-1}$	$\alpha^* = \alpha - \alpha_{fr.car}, \text{cm}^{-1}$	$(\alpha^*)^2, \text{cm}^{-2}$	$(\alpha^* \cdot h\nu)^2, \text{cm}^{-2} \text{eV}^2$	T_{exp}	T_{cor}	$T_{cor} - T_{exp}$
1	0.123	2426	2426	0	0	0	0.500	0.500	0.000
2	0.164	910	910	0	0	0	0.505	0.707	0.202
3	0.202	491	491	0	0	0	0.490	0.794	0.304
4	0.242	308	308	0	0	0	0.450	0.845	0.395
5	0.278	214	214	0	2.18E-24	0	0.425	0.874	0.449
6	0.312	180	159	21	4.35E+02	4.23E+01	0.390	0.884	0.494
7	0.335	2850	132	2718	7.39E+06	8.28E+05	0.145	0.171	0.026
8	0.360	4000	110	3890	1.51E+06	1.96E+06	0.095	0.104	0.009
9	0.384	5125	92	5033	2.53E+07	3.74E+06	0.080	0.080	0.000

Table 2 gives the values of absorption coefficients α calculated by the spectrum according to E_q (2). As is obvious from the data given in Table 2, the absorption coefficient α goes through a minimum.

This means that, when the wavelength increases, the contribution of absorption by free carriers shows up, especially at the concentration of current carriers $\geq 10^{18} \text{cm}^{-3}$.

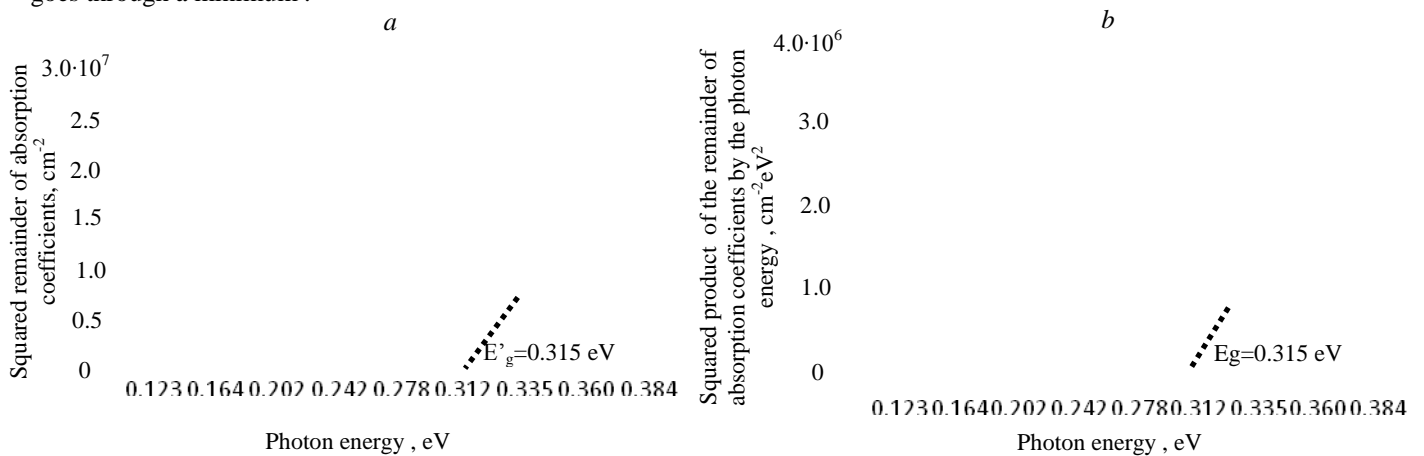


Fig 2a,b. Photon energy dependence of the squared remainder of absorption coefficients (a) and the squared product of the remainder of absorption coefficients by the photon energy (b) with reducing the absorption to zero at $\lambda > 3.7 \mu\text{m}$ for the PbTe layer

For such concentrations of current carriers, the following expression takes place:

$$\alpha_{fr.car.} = e^3 n \lambda^2 / \pi^2 \mu N m^2 \quad (3)$$

where n is the concentration of current carriers; m is the effective mass of electron; N is the index of refraction.

Equating the total absorption coefficient α to the coefficient of absorption by free carriers $\alpha_{fr.car.}$ at $\lambda \approx 10 \mu\text{m}$, we determined the factor to λ^2/N in (3) and then the value of $\alpha_{fr.car.}$ and the difference $\alpha - \alpha_{fr.car.}$ by the spectrum (Table 2).

Table 4 . Data on transmission T indices of refraction N and calculated values of coefficients r_1 and r_2 for the PbSe layer ($d=1.4 \mu\text{m}$)

N_0	$h\nu, \text{eV}$	N_{PbSe}	N_{NaCl}	r_1	r_2	T_{exp}	T_0	T_{cor}	$T_{cor} - T_{exp}$
1	0.167	2.646	1.505	0.451	0.275	0.285	0.959	0.285	0.000
2	0.222	3.990	1.515	0.599	0.450	0.318	0.958	0.318	0.000

3	0.279	4.762	1.521	0.653	0.516	0.250	0.957	0.250	0.000
4	0.334	5.311	1.522	0.683	0.554	0.185	0.957	0.190	0.005
5	0.360	5.542	1.523	0.694	0.569	0.102	0.180	0.089	-0.013
6	0.384	5.760	1.524	0.704	0.582	0.110	0.957	0.091	-0.019
7	0.417	5.846	1.525	0.708	0.586	0.060	0.164	0.056	-0.004

The forbidden gap width E_g of epitaxial PbTe layers was determined by two types of straightening $\alpha^2 = f(h\nu)$ and $(\alpha h\nu)^2 = f(h\nu)$. Fig. 1a,b shows the corresponding dependences, and the straightening of squared absorption coefficients gives the values of E_g 0.308 and 0.309eV, respectively. Such close values of E_g can be explained by a narrow energy range 0.01-0.02eV over which the straightening was performed.

The analysis of the data given in Table 2 and the obtained

values of E_g point to the additional absorption beyond the absorption edge. In an effort to find out whether the emergence of the additional absorption was associated with the necessity in correcting the transmission spectra and its possible effect on the determination of the forbidden gap width E_g , we performed gradual modeling of the transmission spectra with reducing the absorption to zero in the long -wave interval.

Table 5 . Calculated values of absorption coefficients α , differences $\alpha - \alpha_{fr.car.}$, their squared values $(\alpha - \alpha_{fr.car.})^2$ and the values of $1/\gamma$ and $[(\alpha - \alpha_{fr.car.})/1/\gamma]^2$ for the PbSe layer

hν, eV	α , cm ⁻¹	$\alpha_{fr.car.}$, cm ⁻¹	$\alpha^* = \alpha - \alpha_{fr.car.}$, cm ⁻¹	α^{*2} , cm ⁻²	1/γ	$\alpha^2 = (\alpha^* \cdot 1/\gamma)^2$, cm ⁻²	$(\alpha \cdot h\nu)^2$, cm ⁻² eV ²
0.167	7419	7419	0	7.9E-05	14.52	1.66E-02	4.65E-04
0.222	5342	2798	2544	6.5E+06	6.82	3.01E+08	1.48E+07
0.279	5976	1484	4493	2.0E+07	3.41	2.35E+08	1.83E+07
0.334	6955	931	6024	3.6E+07	2.04	1.51E+08	1.68E+07
0.360	8366	768	7598	5.7E+07	1.69	1.65E+08	2.14E+07
0.384	10648	646	10002	1.0E+08	1.47	2.17E+08	3.21E+07
0.417	11531	542	10989	1.2E+08	1.29	2.00E+08	3.48E+07

Table 3 gives the data with zero absorption beyond the absorption edge . As is obvious from Table 3, the reduction of the additional absorption to zero was achieved with considerable deformation of the real spectrum: the transmission should have increased by 0.4, while the possible inaccuracy of calibration of the transmission scale was equal to 0.05. In this case , the straightening of squared absorption coefficients yielded $E_g = 0.315$ eV (Fig.2a,b), which coincided with the previous results within the accuracy of 0.005-0.006eV. This means that the actual additional absorption hardly affects the determination of the forbidden gap width E_g , which could be inferred from our previous works as well [10],[11]. In a similar manner the PbSe layer was analyzed . Table 4 gives the values of the indices of refraction N_{PbSe} . Besides the data by the interference maxima, two points were added: a minimum between the fourth and fifth maxima ($h\nu=0.360$ eV) and one short-wave point ($h\nu=0.417$ eV). The existence of five maxima was determined in this case as well by checking the transmission without absorption and with consideration of the concentration dependence of the indices of refraction. In

Taking into account this coefficient, in the no degenerate case, we have $\alpha' = (\alpha - \alpha_{fr.car.})/\gamma$. It is evident that, at energy $h\nu - E_g = (1+m_c/m_v)E_F$ the value of $1/\gamma$ approximates 2 , and the coefficient α' in the nondegenerate case also increases approximately 2 times as compared to the band-band absorption $\alpha - \alpha_{fr.car.}$. With close values of the effective mass

Table 5 are also given the absorption coefficients α calculated by E_q (2) . It is evident that, despite the less thickness of the PbSe layer, the transmission was less than in the PbTe one and hence the absorption was more. The presence of the minimum in the absorption coefficient α in this case also points to the significant contribution of the absorption by free carriers. Table 5 gives the values of $\alpha_{fr.car.}$, the difference $\alpha - \alpha_{fr.car.}$ and their squared values $(\alpha - \alpha_{fr.car.})^2$ as a function of $h\nu$. The straightening of this function yields the values of the absorption edge at the concentration of current carriers $1.5 \cdot 10^{19} \text{cm}^{-3}$ and corresponds to the sum (Fig. 3):

$$E_g + 2E_F = 0.320 \text{eV} \tag{4}$$

In work [12] the coefficient γ considering for the degeneration was introduced:

$$\gamma = \{1 + \exp[(1+m_c/m_v)E_F - (h\nu - E_g)] / (1 + m_c/m_v)kT\}^{-1} \tag{5}$$

where m_c and m_v are the effective mass of electrons and holes , E_F is the Fermi level, E_g is the forbidden gap width .

m_c and m_v , the straightening of squared difference $\alpha - \alpha_{fr.car.}$ allows obtaining the values of $E_g + 2E_F$.

For reliable calculation of $1/\gamma$, we took the values $E_F = 0.02$ eV and $E_g = 0.286$ eV (which corresponds to the value of E_g in monocrystal PbSe at 300K). Fig.4 shows the

relationship $(\alpha' \cdot hv)^2 = f(hv)$. The straightening of function $[(\alpha - \alpha_{fr.car.})/1/\gamma]^2$ results in the value $E_g = 0.276\text{eV}$. The value of E_g less than in the monocrystal or the unstrained PbSe layer

($E_g = 0.286\text{eV}$) resulted from some compression of the layer on the NaCl substrate.

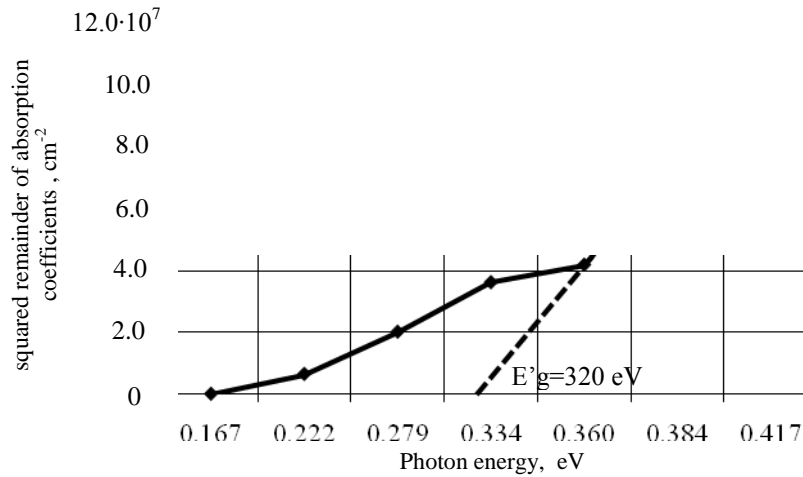


Fig 3. Photon energy dependence of the squared remainder of absorption coefficients for the PbSe layer

Using relationship (4), $E_F = 0.022\text{eV}$ was obtained. By analyzing the data on the dependence of the Fermi level in PbSe on the concentration of current carriers at 80K given in work [13] and determining the coefficients of conversion

of the Fermi level at 300K from work [14], at $E_g = 0.276\text{eV}$ and the concentration of current carriers $1.5 \cdot 10^{19}\text{cm}^{-3}$, we get $E_F = 0.023\text{eV}$, which is close to the value of E_F obtained by calculation.

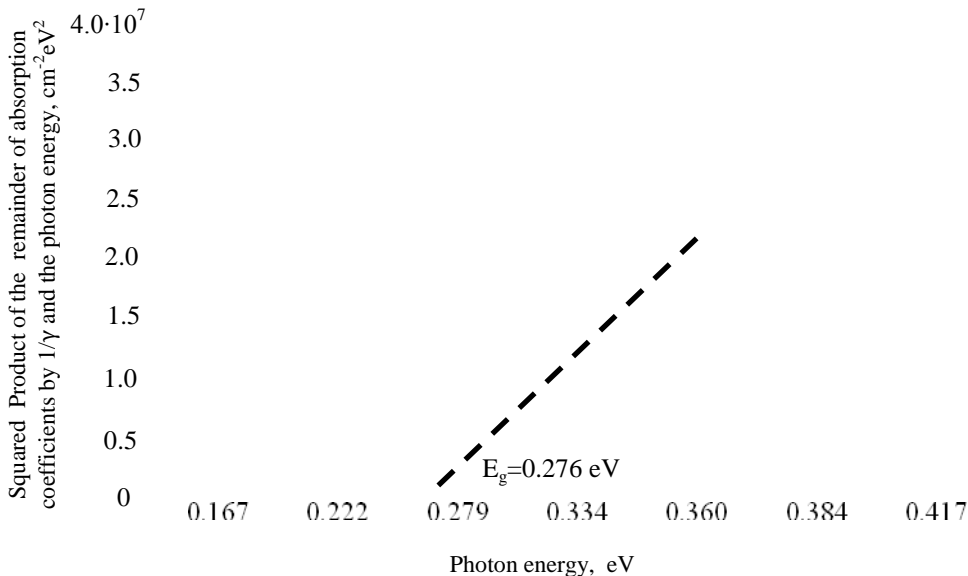


Fig 4. Photon energy dependence of the squared product of the remainder of absorption coefficients by $1/\gamma$ and the photon energy for the PbSe layer

Taking into account that the coefficient $1/\gamma$ can be considered at the energy above the bottom of the conduction band, the absorption spectrum beyond the absorption edge is determined by the difference $\alpha - \alpha_{fr.car.}$. Without multiplication by $1/\gamma$. Then, in the case of PbSe, the additional absorption higher than that in the PbTe layer is also observed beyond the absorption edge (Table 5). The energy threshold for the additional absorption between the absorption edge and the absorption by free carriers increasing with the wavelength for the PbTe layer approximates 0.164eV and for the PbSe it

approximates 0.222eV (Tables 2,5). In both cases these values were revealed at the given thickness of the layers and the absorption is likely to be connected with virtual transitions between different valleys of allowed bands. Therefore, for correct determination of the forbidden gap width in the IV-VI semiconductors, for straightening the squared absorption coefficients, we choose the regions in the absorption spectra where the forbidden gap width corresponds to deformation if it is revealed by the lattice constants. Besides, the two-fold increase in the absorption

coefficient takes place in the nondegenerate semiconductor at energy $2E_F$ (at the same time the Fermi level E_F at 300K is located in the allowed band). The results of this investigation will be useful for the creation of new type of infrared lasers, photo detectors and modulators on the base of IV-VI semiconductors.

IV. CONCLUSIONS

- Because the index of refraction in the IV-VI semiconductors depends heavily on the concentration of current carriers and so do the reflection and absorption coefficients, in the study on the optical transmission spectra with the use of the model of the Fabry-Perot interferometer, we determined these coefficients and the index of, refraction in the epitaxial PbTe and PbSe layers at different concentrations of current carriers $1.2 \cdot 10^{18} \text{ cm}^{-3} - 1.5 \cdot 10^{19} \text{ cm}^{-3}$. It was observed that, as the concentration of current carriers increased and the thickness of layers decreased, the dispersion of the indices of refraction became more pronounced, while the absorption by free carriers and their degeneration enhanced considerably.
- By straightening the squared absorption coefficients, the forbidden gap width of PbTe and PbSe was determined at

300K, which made up 0.309 and 0.276eV, respectively. At high concentration of current carriers, an approach to the choice of the interval for straightening with the purpose to determine correctly the forbidden gap width was justified. The analysis showed that, at the energy $2E_F$, formally the absorption coefficient in the nondegenerate semiconductor increases 2 times in comparison with the degenerated one.

- With model reduction of the absorption to zero in the transmission spectra of epitaxial PbTe layers and modeling of the absorption spectra with consideration for degeneration in the epitaxial PbSe layers, the additional absorption between the absorption edge and the absorption by free carriers increasing with the wavelength was found. It was established that the additional absorption depended on the concentration of current carriers and increased in the PbSe layers. The additional absorption is likely to be related to virtual transitions between different valleys of allowed bands. It is shown that the additional absorption hardly affects the determination of the forbidden gap width.

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