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Photoacoustic spectroscopy analysis of thin semiconductor samples

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ABSTRACT

This paper is an analysis of determination possibility of the optical absorption coefficient spectra of thin semiconductor layers from their normalized photoacoustic amplitude spectra. Influence of multiple reflections of light in thin layers on their photoacoustic and optical absorption coefficient spectra is presented and discussed in detail. Practical formulae for the optical absorption coefficient spectrum as a function of the normalized photoacoustic amplitude spectrum are derived and presented. Next, they were applied for computations of the optical absorption coefficient spectra of thin In_2S_3 thin layers deposited on a glass substrate. This method was experimentally verified with the optical transmission method.

1. Introduction

One of the first papers about the photoacoustic spectroscopy theory for multi-layered samples was published in 1981 [1]. Theoretical relations for the amplitude of a photoacoustic signal for thick semiconductor samples, as a function of the optical absorption coefficient including the influence of multiple reflections of light, are presented in Ref. [2]. The influence of multiple reflections of light on the normalized photoacoustic amplitude spectra, of thin semiconductor layers, was analyzed in Ref. [3]. Ref. [4] presents the dependence of the amplitude of a photoacoustic signal on the optical absorption coefficient of the sample, its optical reflection of light and its thickness. The influence of multiple reflections of light on the normalized photoacoustic amplitude spectra of thin silicon layers was also analyzed in Ref. [5]. The influence of multiple reflections of light on photoacoustic frequency characteristics of thin layers are presented in Refs. [6 and 7]. The model of temperature spatial distribution, in a two layer structure with multiple reflections of light, applied for determination of thermal parameters was presented in Ref. [8].

There are three main ways of analysis of experimental photoacoustic amplitude spectra met in the literature.

The first way is based on a qualitative analysis of experimental PA spectra.

The second one is based on a numerical computation of the optical absorption coefficient spectrum from the photoacoustic amplitude spectrum. In this approach dependence of the optical absorption coefficient on the normalized photoacoustic amplitude spectrum is not given in the analytical way. In a general case it is not possible to express the optical absorption coefficient by the normalized PA amplitude.

The third one is devoted to estimation of the value of the energy gap of the semiconductor sample from the Tauc's characteristics. It assumes the linear dependence of a photoacoustic signal amplitude on the optical absorption coefficient. Condition of this linear dependence is met for small values of the optical absorption coefficient.

In the present paper another way of computation of the optical absorption coefficient spectrum $\beta(\lambda)$ from the normalized photoacoustic amplitude spectrum $q(\lambda)$ is presented. It is based on the application of the analytical formula $\beta(q)$ derived in a thin layer approximation.

There are many papers in the literature presenting experimental photoacoustic amplitude spectra of thin semiconductor layers deposited on optically transparent substrates and their interpretations. They can be divided into three groups:

a) papers presenting the experimental photoacoustic normalized amplitude spectra of thin layers (Table 1)

b) papers about the optical absorption coefficient spectra of thin layers, calculated from the photoacoustic normalized amplitude spectra, using the numerical method (Table 2)

c) papers assuming the optical absorption coefficient spectrum proportionality and the normalized PA amplitude spectrum (Table 3).

In references above experimental PA spectra of thin semiconductor layers were interpreted qualitatively.

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Table 1
Experimental PA amplitude spectra of thin layers presented in the literature.

Semiconductor	Reference
$CuIn_{0.75}Ga_{0.25}Se_2$	[9,10,11]
$CuInSe_2$	[12]
$Cu(In,Ga)Se_2$	[13]
CdTe	[14]
$Hg_{1-x}Cd_xTe$	[15]
In_2S_3	[16,17]
Er_2O_3	[18]
TiO_2	[19,20]
ZnO	[21,22,23]
Fe_2O_3:Cr	[24]
4H-SiC	[25]
Mg_xZn_{1-x}	[26]
$Cd_{1-x}Zn_xS$	[27]
CdS	[28]
Cd_2SnO_4	[29]

Table 2
Optical absorption coefficient spectra computed with the numerical method.

Semiconductor	Reference
$CuIn_{0.75}Ga_{0.25}Se_2$	[30,31]
$CuInSe_2$	[32]
$Cu(In, Ga)Se_2$	[33]

Table 3
Energy gap of semiconductors computed with the Tauc's method.

Semiconductor	Reference
$CuInSe_2$	[34]
µc-Si	[35]
$GaInN_xAs_{1-x}$	[36,37,38]
$CdO + CdTiO_3$	[39]
TiO_2 with CdS QD	[40]
$ZnO + Zn_2TiO_4$	[41]
TiO_2+Ag	[42]
CdS:Al	[43]
In_2S_3	[44]

In references above the numerical method of computations of the optical absorption coefficient spectra from the PA spectra was used. The dependance complex form, in a general case, does not allow for a computation of the optical absorption coefficient spectrum from the PA spectrum in the analytical way.

Also, in references above, a linear dependence of the photoacoustic signal amplitude on the optical absorption coefficient value was assumed. With this assumption it is possible to determine the semiconductor energy gap value by the Tauc's characteristic method.

2. Theoretical model

As a result of performed investigations, practical formulae for computations of the optical absorption coefficient spectra from the normalized photoacoustic spectra are presented. They were applied for computations of the optical absorption coefficient spectra of In_2S_3 thin layers.

The paper presents derivations of the formulae for different cases:

a) the optical absorption coefficient of a thin sample as a function of R_{opt1}, R_{opt2}, q,d, with multiple reflections of light $\beta = f\left(R_{opt1}, R_{opt2}, q, d\right)$

$$\beta = f\left(R_{opt1}, R_{opt2}, q, d\right),$$

where: R_{opt} is the optical reflection coefficient of light between the sample and the medium, e.g., gas or a transparent substrate material, q is the normalized amplitude of the photoacoustic signal, d is the thickness of a sample;

b) the optical absorption coefficient of a thin sample as a function of R_0, q, d with multiple reflections of light when R_0=R_{opt1}=R_{opt2}

$$\beta = f\left(R_0, q, d\right);$$

c) the optical absorption coefficient of a thin sample as a function of q, d when the effect of multiple reflections of light in the sample can be neglected

$$\beta = f\left(q, d\right).$$

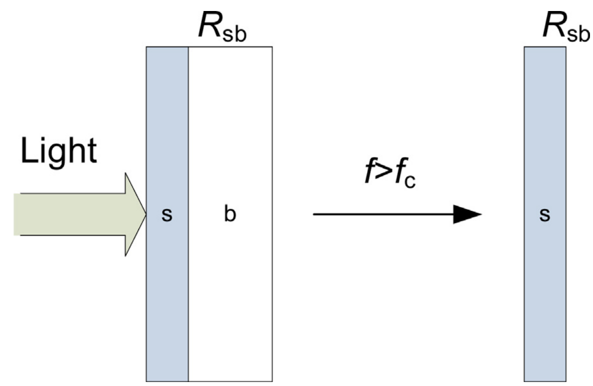

Fig. 1. Two layer model considered as one layer model, s- is the sample, b- is the glass backing, R_{sb} - is the thermal reflection coefficient between the sample and the substrate.

When an optically transparent and thermally thick glass substrate is considered, a one layer model with a proper thermal wave reflection coefficient can be used for the analysis of a photoacoustic signal. The theoretical situation is presented in Fig.1.

The substrate can be considered as thermally thick for frequencies higher than its characteristic frequency f_c,

$$f_c = \frac{D_{Tb}}{\pi d_b^2}, \tag{1}$$

where D_{Tb} is the thermal diffusivity and d_b is the thickness of the glass substrate.

Temperature of the front (illuminated) surface of the sample T, when the multiple reflections of light in the thin layer are taken into account, is given by Eq. (2) derived in Ref. [4]

$$T = \frac{\left(1 - R_{opt1}\right)\left(T_F + R_{opt2}\exp\left(-\beta d\right)T_R\right)}{\left(1 - R_{opt1}R_{opt2}\exp\left(-2\beta d\right)\right)}, \tag{2}$$

where: T_F and T_R are given by Eqs. (3) and (4)

$$T_F = \frac{\beta}{\lambda\sigma\left(1 - R_{sb}\exp\left(-2\sigma d\right)\right)}\left(\frac{1 - \exp\left(-\left(\beta + \sigma\right)d\right)}{\beta + \sigma} + \frac{R_{sb}\exp\left(-2\sigma d\right)\left(1 - \exp\left(-\left(\beta - \sigma\right)d\right)\right)}{\beta - \sigma}\right) \tag{3}$$

$$T_R = \frac{\beta\exp\left(-\sigma d\right)}{\lambda\sigma\left(1 - R_{sb}\exp\left(-2\sigma d\right)\right)}\left(\frac{R_{sb}\left(1 - \exp\left(-\left(\beta + \sigma\right)d\right)\right)}{\beta + \sigma} + \frac{1 - \exp\left(-\left(\beta - \sigma\right)d\right)}{\beta - \sigma}\right), \tag{4}$$

where: d – thickness of a thin layer, σ =(1+i)/μ, μ - thermal diffusion length, λ thermal conductivity, R_{sb} – thermal reflection coefficient defined as R_{sb}=(e_s-e_b)/(e_s+e_b), e_s e_b are effusivities of the thin layer and the glass substrate, respectively, β - optical absorption coefficient, R_{opt1}, R_{opt2}- optical reflection coefficients of air-thin layer and a thin layer-glass substrate, respectively.

Case a)

When light penetration depth in the sample is smaller than its thermal diffusion length, i.e.,

β^{-1}«μ, and the thermal diffusion length is bigger than the thickness of the sample, i.e., μ»d then Eqs. (2)–(4) can be simplified.

$$T_F = \frac{\left(1-\exp\left(-\beta d\right)\right)(1+R_{sb}\exp(-2\sigma d))}{\lambda\sigma(1-R_{sb}\exp(-2\sigma d))} = \frac{\left(1-\exp\left(-\beta d\right)\right)(1+R_{sb})}{\lambda\sigma(1-R_{sb})} \tag{5}$$

$$T_R = \frac{\left(1-\exp\left(-\beta d\right)\right)(1+R_{sb})\exp(-2\sigma d)}{\lambda\sigma(1-R_{sb}\exp(-2\sigma d))} = \frac{\left(1-\exp\left(-\beta d\right)\right)(1+R_{sb})}{\lambda\sigma(1-R_{sb})} \tag{6}$$

For this case T_F=T_R=T_0 and temperature of the sample illuminated side T takes the form of Eq. (7).

$$T = \frac{\left(1-R_{opt1}\right)\left(1+R_{opt2}\exp\left(-\beta d\right)\right)}{\left(1-R_{opt1}R_{opt2}\exp\left(-2\beta d\right)\right)}T_0 = \frac{\left(1-R_{opt1}\right)\left(1+R_{opt2}\exp\left(-\beta d\right)\right)\left(1-\exp\left(-\beta d\right)\right)(1+R_{sb})}{\left(1-R_{opt1}R_{opt2}\exp\left(-2\beta d\right)\right)\lambda\sigma(1-R_{sb})}. \tag{7}$$

When the optical absorption coefficient β goes to infinity, the PA signal saturates, and Eq. (7) simplifies to Eq. (8).

$$T_{sat} = \frac{\left(1-R_{opt1}\right)(1+R_{sb})}{\lambda\sigma(1-R_{sb})}. \tag{8}$$

The normalized amplitude of the PA signal is defined by formula (9)

$$q = \left|\frac{P}{P_{sat}}\right| = \left|\frac{T}{T_{sat}}\right|, \tag{9}$$

where: P is the amplitude of periodical component of the gas pressure in PA chamber, P_{sat} is the periodical component of the gas pressure amplitude in PA chamber for a big value of the optical absorption coefficient when PA signal reaches saturation.

The normalized amplitude of the PA signal can be expressed by Eq. (10).

$$q = \frac{\left(1+R_{opt2}\exp\left(-\beta d\right)\right)\left(1-\exp\left(-\beta d\right)\right)}{\left(1-R_{opt1}R_{opt2}\exp\left(-2\beta d\right)\right)}. \tag{10}$$

The optical absorption coefficient β can be computed from the normalized PA amplitude q by the general Eq. (11).

$$\beta = -\frac{1}{d}\ln\left(\frac{\sqrt{(1-R_{opt2})^2+4R_{opt2}(1-R_{opt1}q)(1-q)}-(1-R_{opt2})}{2R_{opt2}(1-R_{opt1}q)}\right). \tag{11}$$

Case b)

When the optical reflection coefficients R_{opt1}=R_{opt2}=R_o are equal then Refs. [7, 8 and 10] can be written as Refs. [12–14], respectively.

$$T = \frac{(1-R_o)\left(1+R_o\exp\left(-\beta d\right)\right)T_0}{\left(1-R_o^2\exp\left(-2\beta d\right)\right)} = \frac{(1-R_o)\left(1+R_o\exp\left(-\beta d\right)\right)}{\left(1-R_o^2\exp\left(-2\beta d\right)\right)}\frac{\left(1-\exp\left(-\beta d\right)\right)(1+R_{sb})}{\lambda\sigma(1-R_{sb})} \tag{12}$$

$$T_{sat} = \frac{(1-R_o)(1+R_{sb})}{\lambda\sigma(1-R_{sb})} \tag{13}$$

$$q = \frac{\left(1+R_0\exp\left(-\beta d\right)\right)\left(1-\exp\left(-\beta d\right)\right)}{1-R_0^2\exp\left(-2\beta d\right)} = \frac{1-\exp\left(-\beta d\right)}{1-R_0\exp\left(-\beta d\right)}. \tag{14}$$

In this case the optical absorption coefficient β can be computed from the normalized PA amplitude q by Eq. (15).

$$\beta = -\frac{1}{d}\ln\left(\frac{1-q}{1-R_o q}\right). \tag{15}$$

Case c)

When multiple reflections of light in the sample can be neglected, and there are not any optical absorption of light in the substrate the following relations take place

$$T = \frac{\left(1-R_{opt1}\right)\left(1-\exp\left(-\beta d\right)\right)}{\lambda\sigma(1-R_{sb})}(1+R_{sb}) \tag{16}$$

$$T_{sat} = \frac{\left(1-R_{opt1}\right)(1+R_{sb})}{\lambda\sigma(1-R_{sb})} \tag{17}$$

$$q = \left|\frac{T}{T_{sat}}\right| = 1-\exp(-\beta d). \tag{18}$$

The value of the optical absorption coefficient β can be determined from the value of the normalized PA amplitude q by Eq. (19).

$$\beta = -\frac{\ln(1-q)}{d}. \tag{19}$$

2.1. Numerical verification of the theoretical model

Let's assume the optical absorption coefficient spectrum in the form presented by Eq. (20). It is spectrum for direct electron type transitions and Urbach absorption tail for energy of photons $h\nu$ smaller than the energy gap E_g = 2.07 eV. This optical absorption coefficient spectrum is presented in Fig.2a). The normalized PA amplitude spectrum computed for this spectrum with Eq. (18) and the thickness of the sample d = 179 nm is presented in Fig.2b).

$$\beta = \begin{cases} \frac{6.5\times10^5}{h\nu}(h\nu-2.07)^{0.5}+2\times10^4 \text{ for} h\nu > 2.07 \\ 6\times10^4\left(\frac{h\nu-2.07}{0.1}\right) \text{ for} h\nu \leq 2.07 \end{cases} \tag{20}$$

For both optical absorption coefficient spectrum and normalized PA amplitude spectrum the Tauc's characteristics were next computed. They are presented in Fig.3.

From the optical absorption coefficient spectrum the energy gap Eg = 2.05 eV was found.

The fitting results: $\beta h\nu$=$A(h\nu$-$Eg)^{0.5}$ where: A = 6.9 × 10^5 $eV^{0.5}$/cm, Eg = 2.05 eV.

From the normalized PA spectrum the energy gap Eg = 1.97 eV was estimated. It is much smaller than the assumed value Eg = 2.07 eV in Eq. (20).

The fitting results:$(PAS\ h\nu)^2$ = 18($h\nu$ -Eg) where Eg = 1.97 eV.

One can see from Fig.3. that the value of the energy gap obtained from the PAS spectrum is smaller than the value assumed in equation for optical absorption coefficient spectrum.

2.2. Interpretation of In_2S_3 experimental PA amplitude spectra

The above Eqs. (11) and (19) were next applied for computations of optical absorption coefficient spectra of In_2S_3 thin layer deposited on the thermally thick and optically transparent glass

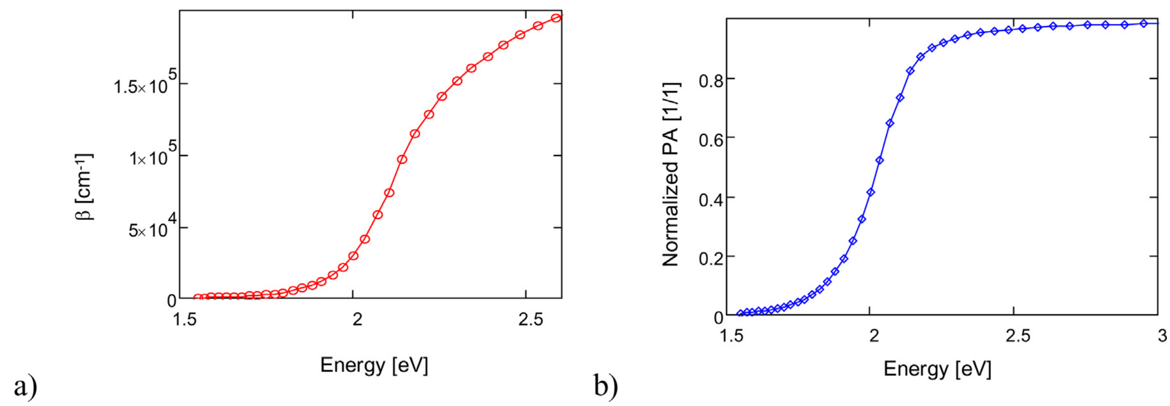

Fig. 2. a) Optical absorption coefficient spectrum taken for computations; b) Normalized PA spectrum computed for the optical absorption coefficient spectrum.

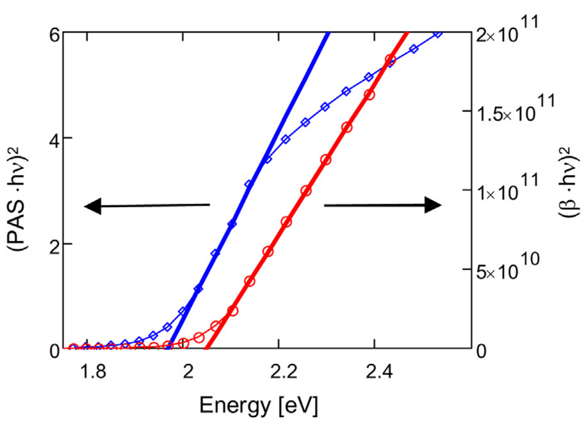

Fig. 3. The comparison of Tauc's characteristics obtained from normalized PA amplitude spectrum q and optical absorption coefficient spectrum β.

substrate. Thickness of the layer was of $d = 0.179\,\mu$m, thickness of the substrate was of 1.2 mm, refractive index of the substrate was of 1.5, refractive index of the sample $n = 2.7$ and $R_{opt1} = 0.21$, $R_{opt2} = 0.08$. Fig. 4a) presents a normalized photoacoustic amplitude spectrum of this layer measured at the frequency $f = 36$ Hz. Fig. 4b) presents an optical absorption coefficient spectra of the In_2S_3 sample computed by Eqs. (11) and (19).

Comparison of the optical absorption coefficient spectra computed by different Eqs. (11) and (11) presented in Fig.4b) shows that the spectra are similar. There is not any big influence of a reflection coefficient on the obtained optical absorption spectra in the observed range of R_{opt1}=(0,0.2). Of course, a bigger R_{opt1} will give a bigger difference between a full model [Eq. (11)] and a simplified model [Eq. (19)].

2.3. Experimental verification of the proposed method

The experimental verification of the proposed method of computation was performed on the In_2S_3 sample of thickness of $d = 0.57$ u m. The experimental normalized PA spectrum of the sample at $f = 36$ Hz is presented in Fig. 5a). The optical transmission spectrum of the sample is presented in Fig.5b). The optical absorption coefficient spectra obtained from these experimental spectra are presented in Fig.5c).

Comparison of these experimental optical absorption coefficient spectra obtained by different methods shows that they are similar. Differences in the obtained spectra are the result of a different influence of interference effects on the transmission and PA experimental spectra.

3. Conclusions

The paper presents several equations for computations of the optical absorption coefficient spectra, from the normalized photoacoustic amplitude spectra, of thin semiconductor samples deposited on a thermally thick and optically transparent substrate.

It also analyses differences between two approaches used for computations of the energy gap values of semiconductors from the Tauc's characteristics. The first one is based on the photoacoustic amplitude spectrum and the second one is based on the optical absorption coefficient spectrum computed from the photoacoustic amplitude spectrum. It was proved that different values

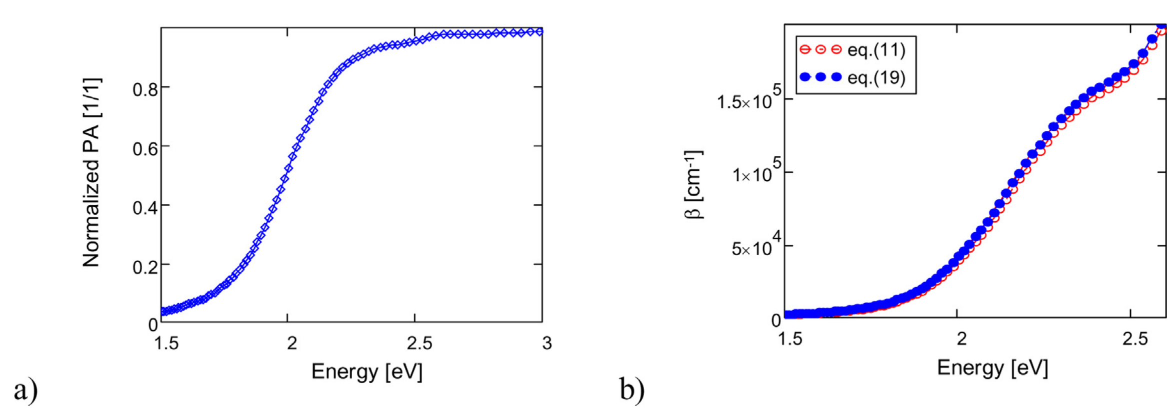

Fig. 4. a) Normalized photoacoustic spectrum of In_2S_3 at $f = 36$ Hz; b) Comparison of optical absorption coefficient spectra computed with Eqs. (11) and (19). Thickness of the sample of $d = 0.179\,\mu$m.

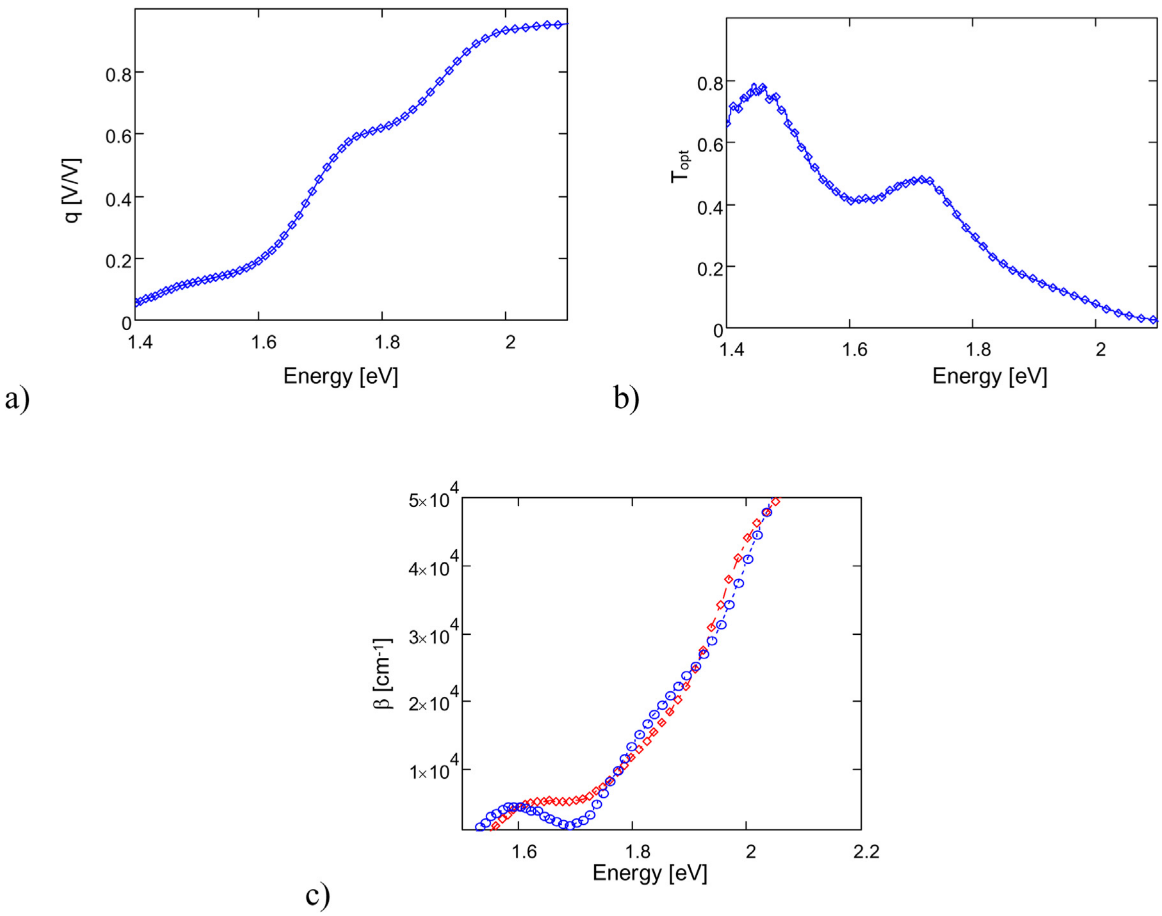

Fig. 5. a) Normalized, experimental photoacoustic amplitude spectrum q of the In_2S_3 sample; b) Experimental, optical transmission spectrum of the In_2S_3 sample; c) Optical absorption coefficient spectra computed from the experimental optical transmission spectrum (circles), computed from experimental normalized PA amplitude spectrum (diamonds). Thickness of the sample $d = 0.57$ µm.

of the energy gap are obtained depending on the approach. Only the values of the energy gaps computed from the optical absorption spectrum are correct.

In this paper the influence of the optical reflection coefficients on the optical absorption coefficient spectra computed from the PAS spectra is shown and discussed, too.

Comparison of the optical absorption coefficient spectra obtained from PA spectra and the spectra obtained from the optical transmission spectra showed that the spectra are similar. It is the experimental verification of the proposed method.

This paper shows advantages of application of the presented equations for numerical interpretations of the experimental photoacoustic amplitude spectra of thin semiconductor layers as they allow to get the optical absorption coefficient spectra of thin semiconductor layers in the analytical way and allow for a calculation of the correct values of their energy gaps.

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