JOURNAL OF NANO- AND ELECTRONIC PHYSICS
Vol. **5** No 3, 03038(3pp) (2013)

ЖУРНАЛ НАНО- ТА ЕЛЕКТРОННОЇ ФІЗИКИ
Том **5** № 3, 03038(3cc) (2013)

Periodicity of the Distribution of Intrinsic Defects in Epitaxial PbTe Films

Ya.P. Saliy, D.M. Freik, I.K. Yurchyshyn, I.M. Freik

Vasyl Stefanyk Precarpathian National University, 57, Shevchenka Str., 76000 Ivano-Frankivsk, Ukraine

(Received 14 February 2013; published online 17 October 2013)

The *p*-type PbTe films have been grown on the mica substrates by the method of open evaporation in vacuum. It was established that dimensional effects in the films are connected with periodic distribution of donors. Approximation of the experimental effective dependences of the conductivity σ_d and the product of the Hall coefficient and square of conductivity $R_d\sigma_d^2$ on the thickness by the theoretical dependences was executed. These dependences are integrals of local concentration $n(x)$ represented by the sinusoid which is determined by the distribution of donors in depth, and mobility μ, which are independent on coordinate. Spatial parameters of the distributions of defects were obtained. Based on the layered inhomogeneity of semiconductor PbTe films, we have suggested the hypothesis of diffusion instability of point defects initiated by the substrate-film interface.

Keywords: Thin films, Dimensional effects, Lead chalcogenides, Periodicity, Damped deflection of characteristics, Methodological averaging, Diffuse instability.

PACS numbers: 75.50.Lw, 73.63.Hs, 73.61.Ey

1. INTRODUCTION

Film structures based on lead chalcogenides are studied for the purpose of their application in infrared technology, optoelectronics, and thermoelectricity [1-3]. Thickness dependence of the characteristics of the lead chalcogenide thin films grown by epitaxial methods was discussed repeatedly. Thus, it is established in the works [4-6] that stratified inhomogeneity of the distribution of defects in PbTe films is connected with substrate-film interface. Obviously, the thinner film is, the more significant influence of the substrate on the concentration and mobility of charge carriers in the film is.

It was natural to perform the analysis of just those integral electrophysical values and their combinations, which in the measurements, for example, of the conduction and the Hall constant, are really the layer-by-layer integrals of the corresponding local characteristics or their combinations. In the work [4] the main attention was devoted to the processes of free carrier scattering, however, it was not noted that carrier concentration in the films of different thickness differed by an order of magnitude. Analysis of the dimensional effects in semiconductor films in [6] already took into account distributions of the donor and acceptor centers and scattering centers. However, identity or differences of the scattering centers and centers which determine the free carrier concentration in lead chalcogenides at high temperatures with impurity conduction is not still established.

Concentration of structural defects, conditioned by the inconsistency of crystal structure of the condensate and substrate, can considerably influence the concentration of point defects removed far from the substrate because of their diffusion instability [7]. Free surface of the film can be also the source of defects, however, one can expect appreciable influence at room temperature of defects conditioned by evaporation and segregation of the condensate components, adsorption and diffusion of atmospheric oxygen etc. for a long time after growth.

2. ADDITIVE ELECTROPHYSICAL CHARACTERISTICS

If current density is changed with the coordinate *x* along the axis perpendicular to the film surface as in the case of stratified-non-uniform sample, then certain effective coefficients and their combinations can be expressed through the averaged in thickness local analogs as follows [5]:

$$\sigma_d = d^{-1}\,{}_0\!\int^d \sigma(x)dx, \quad (1)$$

$$R_d\ \sigma_d^2 = d^{-1}\,{}_0\!\int^d R(x)\sigma^2(x)dx, \quad (2)$$

where σ_d is the effective conduction; R_d is the effective Hall coefficient. Expressing local conduction $\sigma(x)$ and Hall coefficient $R(x)$ through the concentration of free charge carriers $p(x)$ connected with the concentration of acceptors and donors and mobility $\mu(x)$ connected with the distribution of scattering centers, we obtain from (1) and (2) [6]

$$\sigma_d = d^{-1}\,{}_0\!\int^d ep(x)\mu(x)dx, \quad (3)$$

$$R_d\ \sigma_d^2 = d^{-1}\,{}_0\!\int^d ep(x)\mu^2(x)dx. \quad (4)$$

We note that average value of the constant is equal to this magnitude, and, for example, of the oscillating one – damps to zero. At averaging, contribution of the oscillating component of the characteristic of the layer, whose thickness is multiple to the oscillation period, is equal to zero.

3. MEASUREMENTS AND EXPERIMENT REPRESENTATION

Films of the stoichiometric composition were grown in a vacuum chamber by the open evaporation method on the mica substrates. Thickness of the *p*-type PbTe films was varied in the range of 0.16-0.8 μm and measured by using microinterferometer MII-4 with the accuracy of 0.02 μm. Measurements for one sample were carried out in air at room temperature for three time intervals after growth 10^3, 10^4 and 10^5 s in the magnetic field of 2 T.

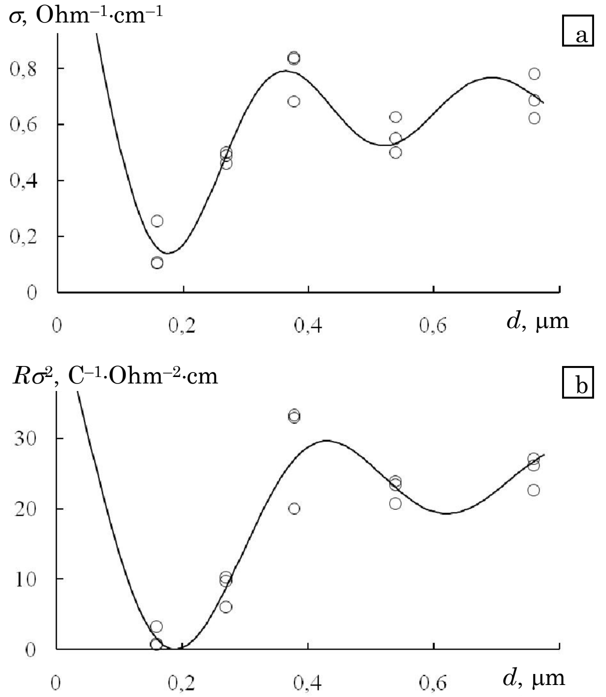

Fig. 1 – Dependences of the conductivity σ (a) and product of the Hall constant and square of conductivity $R\sigma^2$ (b) for the *p*-type PbTe films on the thickness: dots denote the experiment, lines are the approximation

Conductivity and product of the Hall constant and square of conductivity for the films of different thickness and three time intervals are represented in Fig. 1. Effect of the moment of measurement on the experimental values is not revealed. Therefore, in Fig. 1 dots which correspond to different measurement times are given by the same symbols and their spread for one thickness is considered to be an experimental error.

We observe that both sets of the experimental data behave as damped vibrations with saturation for large thicknesses. One can reveal 1,5 vibrations.

4. THEORETICAL MODEL AND APPROXIMATION FUNCTION

At high temperatures concentration of the electrically active defects determines concentration of free charge carriers. For the analysis of the experimental data we have used the function which is the solution of diffusion equations for intrinsic defects with taking into acount their thermal generation and recombination [7]. Therefore, local concentrations of the scattering centers which define carrier mobility and acceptor defects are considered to be coordinate-independent – a trivial solution, and local concentrations of donors are described by the sinusoid – non-trivial solution:

$$ep(x)\mu = e\mu\,(N_a - N_d \sin(2\pi x / \lambda - \varphi)), \qquad (5)$$

$$ep(x)\mu^2 = e\mu^2\,(N_a - N_d \sin(2\pi x / \lambda - \varphi)), \qquad (6)$$

where λ is the wavelength of the distribution of defects; φ is the initial phase.

Model validity of the periodic distribution of point defects by thickness will be confirmed by further results.

Defect states are completely ionized at the specified measurement temperatures, therefore, concentration of free charge carriers is equal to the difference of their concentrations $p(x) = N_a(x) - N_d(x)$. Mobility of free charge carriers is inversely proportional to the concentration of scattering centers, however, for high temperatures it is determined by the scattering by phonons, whose distribution does not depend on the coordinate. Using formulas (3) and (4) for integral dependences and (5) and (6) for functions, we obtain expressions which will be used in approximation of the experimental data

$$\sigma_d = e\mu N_a\,(1 - N_d / N_a\,\lambda / 2\pi d\,\sin(\pi d / \lambda - \varphi)\sin(\pi d / \lambda)), \qquad (7)$$

$$R_d\,\sigma_d^2 = e\mu^2 N_a\,(1 - N_d / N_a\,\lambda / 2\pi d\,\sin(\pi d / \lambda - \varphi)\sin(\pi d / \lambda)). \qquad (8)$$

5. DISCUSSION OF THE APPROXIMATION RESULTS

Four parameters of approximation functions (7) and (8) obtained by the least square method for two sets of the dependences in 15 dots are represented in Table 1 in the first and second rows. It is seen from Table 1 that spatial characteristics of the distributions of λ and φ in the range of accuracy weakly depend on the set. Ratio N_d / N_a of the amplitude value of donor concentration to the acceptor concentration does not depend on the data set. Wavelength of donor distribution λ exceed by an order of magnitude the free path of charge carriers $l = 0.04$ μm and Debye screening length $L_D = 0.025$ μm [3], therefore, it cannot be defined by them. We assume that films are thick, because their thickness is larger than the free path of charge carriers and Debye screening length.

Table 1 – Parameters of the dependences which approximate the experimental data

	$e\mu N_a$, Ohm^{-1}cm^{-1}	$e\mu^2 N_a$, C^{-1}Ohm^{-2}cm	N_d / N_a	λ, μm	φ, rad
σ_d – (7)	0.74	–	1.71	0.33	0.75
$R_d\,\sigma_d^2$ – (8)	–	29	1.70	0.40	0.42
$1 - N_d / N_a$	–	–	1.70	0.37	0.56

We will represent the thickness dependences for the compensation factor $(N_a - N_d) / N_a$, whose values one can obtain dividing the experimental values given in Fig. 1 by the corresponding dimensional coefficients taken from the second and third columns of Table 1. The dependence common for two sets is illustrated in Fig. 2. Parameter $N_d / N_a = 1.7$ was fixed, and two other parameters were obtained from the approximation for 30 dots and represented in the fourth row of Table 1. It is seen that spatial characteristics of this distribution $\lambda = 0.37 \pm 0.03$ μm and $\varphi = 0.56 \pm 0.15$ with the stated accuracy lie in the range of the values determined separately for two sets.

In Fig. 2, besides the integral distribution of the compensation factor on the film thickness *d*, we present the local one versus the coordinate *x* along the axis perpendicular to the film surface. As seen from the local distribution, minima of sinusoids are shifted to lesser spatial values in comparison with minima of damped oscillations of the integral distribution. Minima correspond to the regions of donor predominance, and maxima – acceptor.

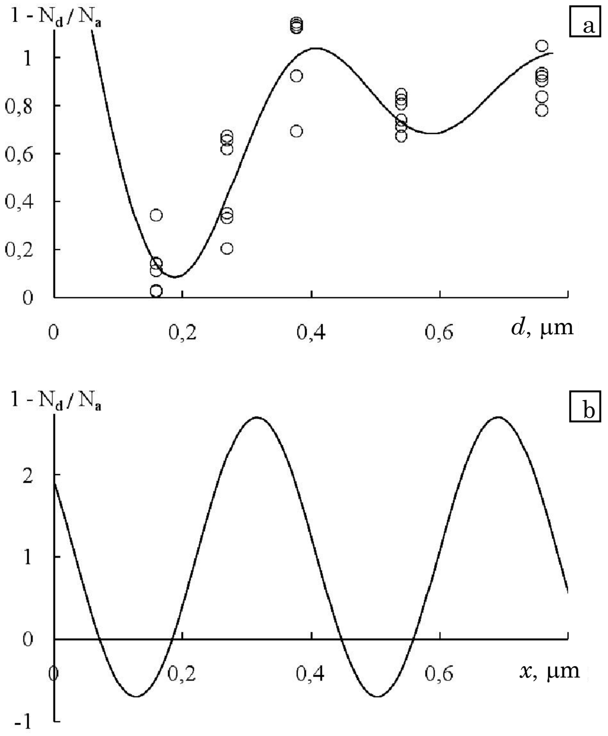

Fig. 2 – Distributions of the compensation factor $(N_a – N_d) / N_a$: integral – on the film thickness d and local – on the coordinate x along the axis perpendicular to the p – PbTe film surface ($x = 0$ corresponds to the substrate/film interface)

It is known that IV-VI compounds are the promising materials among effective thermoelectric ones of medium temperatures; and increase in the thermoelectric Q-factor is extraordinarily important question. The authors of a number of publications have noted repeatedly the growth of Q-factor in artificially formed PbTe / $Pb_{1-x}Eu_xTe$ [8], PbTe / PbSe [9] superlattices in comparison with uniform materials. However, it was revealed that periodic structures are also formed during the thin film growth by thermal deposition of PbS, PbSe and PbTe [10-12]. In this case, the Seebeck coefficient and electrical conduction differ in minima and maxima of the thickness dependences by 2 times. Such periodicity with the period of $\lambda \approx 0{,}03$ µm is connected with the dimensional quantization, though this explanation of periodicity $\lambda \approx 0{,}2$ µm, as it is shown in [13], gives rise to doubts.

6. CONCLUSIONS

Damped thickness oscillations of electrophysical film characteristics are connected with periodic distribution of defects.

Concentration of donor defects in lead telluride films is distributed periodically along the direction perpendicular to their surface. Spatial parameters and amplitude values of the periodic distribution of defect concentrations in the film are obtained.

Ionized scattering centers are distributed almost uniformly over the sample thickness; a certain deviation is observed near the substrate, where mobility of charge carriers slightly decays.

Generation of the periodic distribution of defects of different types is one of the ways to increase the thermoelectric Q-factor of the materials and can promote the formation of superlattices, whose properties are used for the design of new functional elements of semiconductor devices of micro-, opto-, and thermoelectricity.

REFERENCES

1. A.V. Dmitriev, I.P. Zvyagin, *Phys.-Usp.* **53**, 789 (2010).
2. B.A. Volkov, L.I. Ryabova, D.R. Khokhlov, et al., *Phys.-Usp.* **45**, 819 (2002).
3. D.M. Zayachuk, V.A. Shenderovskiy, *UFZh* **36** No11, 1692 (1991).
4. O.A. Aleksandrova, R.Ts. Bondokov, I.V. Saunin, Yu.M. Tairov, *Semiconductors* **32**, 953 (1998).
5. R.L. Petritz, *Phys. Rev.* **110** №6, 1254 (1958).
6. Ya.P. Saliy, N.Ya. Stefaniv, *FKhTT* **13** No1, 69 (2012).
7. A.S. Vasin, M.I. Vasilevsky, *Phys. Solid State* **48**, 37 (2006).
8. L.D. Hicks, T.S. Harman, X. Sun, M.S. Dresselhaus, *Phys. Rev. B* **53**, R10493 (1996).
9. G. Springholz, M. Pinczolits, P. Mayer, V. Holy, G. Bauer, H. Kang, L. Salamanca-Riba, *Phys. Rev. Lett.* **84**, 4669 (2000).
10. E.I. Rogacheva, O.N. Nashchekina, Y.O. Vekhov, M.S. Dresselhaus, S.B. Cronin, *Thin Solid Films* **423**, 115 (2003).
11. E.I. Rogacheva, T.V. Tavrtna, O.N. Nashchekina, S.N. Grigorov, K.A. Nasedkin, *Appl. Phys. Lett.* **80**, 2690 (2002).
12. E.I. Rogacheva, O.N. Nashchekina, S.N. Grigorov, M.S. Dresselhaus, S.B. Cronin, *Nanotechnology* **14**, 53 (2003).
13. D.M. Freik, I.K. Yurchyshyn, V.M. Chobanyuk, *Sensor Electron. Microsyst. Technol.* **2** No8, 41 (2011).