

## OPTICAL PARAMETER SPECTRA OF Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) SINGLE CRYSTALS

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$R(E)$  reflection spectra of Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) single crystals in beam energy interval 1 ÷ 6 eV are investigated. For the case of Bi<sub>2</sub>Te<sub>3</sub> single crystals the measurements are carried out parallel and perpendicular to optical  $C$  axis. It is shown that the energy values of some optical transitions from crystalline to non-crystalline state in Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) are constant.

**Keywords:** optical spectra, reflection coefficient, optical conduction.

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### 1. INTRODUCTION

$R(E)$  reflection coefficients of some materials (amorphous and single-crystalline) Se, Se-S, InSnTe<sub>2</sub>, TlIn<sub>0.9</sub>Ce<sub>0.1</sub>Se<sub>2</sub>, TlInSe<sub>2</sub>Ce<sub>0.04</sub>, TlInSe<sub>2</sub>, Cu<sub>3</sub>GdTe<sub>3</sub>, Cu<sub>5</sub>GdTe<sub>4</sub>, CuGdTe<sub>2</sub>, Se<sub>95</sub>As<sub>5</sub> (with impurity Sm), Bi<sub>2</sub>Te<sub>3</sub>(Ni, Cu, Zn), Bi<sub>2</sub>Te<sub>3</sub>, Bi<sub>2</sub>Se<sub>3</sub> are measured and their optical parameters such as  $\theta$  reflected light phase, indexes of  $\kappa$  absorption and  $n$  reflection, real  $\epsilon_1$  and imaginary  $\epsilon_2$  parts of dielectric constant,  $\alpha$  absorption coefficient, function of characteristic volume  $-Img \epsilon^{-1}$  and surface  $-Img(\epsilon+1)^{-1}$  electron losses, electrooptical differential functions ( $\alpha$ ,  $\beta$ ), optical conduction  $\epsilon_2 E$ , integral function of bound state density  $\epsilon_0(E)$ , effective number of valence electrons  $n_{ef}(E)$  [1], taking part in transitions up to the given energy  $E$  are calculated by us.

The bismuth telluride is known as effective material for thermoelectric transformers. This material is easier to prepare in the form of enough perfect single crystals and obtain both  $n$ - and  $p$ -type by the way of doping [2,6].

Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) crystals has packet structure and the bound between neighbor packets has the mixed Van-der-Waals-covalent character [7]. The additional bond between packets is carried out because of transition of one  $p$ -electron on  $d$ -levels and overlapping of some  $d$ -levels with valence band. All this causes the significant metallic properties and comparably small energy values of forbidden bands in interval 0,15 ÷ 0,35 eV.

Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) and its analogues are the uniaxial crystals in optical relation. The dielectric constant in them is the tensor of second order and depends on direction of incident wave with respect to optical  $C$  axis. The bismuth telluride optical properties are investigated in region of higher frequencies in work [5].

The band structure of Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) crystal is theoretically calculated in work [5]. The absence of data on ( $\Delta$ ) spin-orbital interaction value and complexity of chemical bond character between Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) atoms make the essential difficulty.

The bismuth telluride and solid solutions on its base are used at preparation of different energy

transformers [7]. The single-crystalline or polycrystalline Bi<sub>2</sub>Te<sub>3</sub> and its solid solutions with Bi<sub>2</sub>Se<sub>3</sub> are mainly used. The single-crystalline samples Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) are easily sheared by cleavage plane [0001] forming the mirror surface which stable to the oxidation that is very important for carrying out of optical measurements and doesn't require the special thermal treatment.

The study of band structure Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) hasn't achieved the level study of germanium and  $A^{III}B^V$  compound that is connected with complexity of its crystalline and band structures [3]. This makes new investigations very important.

The measurement of crystal reflection coefficient of Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) of  $n$ - and  $p$ - types and also its polycrystalline film samples, parallel and perpendicular to  $C$  axis and the definition of spectra of their optical parameters on the base of the reflection coefficient are the tasks of the given work.

### 2. EXPERIMENT TECHNIQUE

The cleavage of Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) single crystal having the mirror smooth surface is used for measurement of  $R(E)$  reflection coefficient. The reflection coefficient is measured by method of double-beam spectroscopy. The crystals are doped by Cl impurities having  $n$ - type conduction and Tb impurities which have  $p$ - type conduction.

The technology of single crystal Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) and films obtaining is described in works [8, 9, 10]. The single crystals are obtained by Bridgman method as in [3] and films by thickness 0,3  $\mu$ m of polycrystalline Bi<sub>2</sub>Te<sub>3</sub> (Ni, Cu, Zn) on cleavage surface of rock salt are obtained by its in sublimation vacuum.

The definition methods of optical parameters are given in work [11] and this procedure is described in [12].

As usual, the special computer programs are applied for calculation of optical parameters. The optical parameters of investigated materials are calculated by programs written by author of work [13]. These programs are checked at calculation of optical parameters of some materials in works [14 – 18].

### 3. RESULTS AND THEIR DISCUSSION

$R(E)$  reflection coefficients of single-crystalline  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) of  $n$ - and  $p$ - types parallel and perpendicular to  $C$  axis and also its film samples of  $n$ - and  $p$ - types are measured in the work; the spectra of their optical parameters:  $\alpha$  absorption coefficient,  $\varepsilon_1$  real and  $\varepsilon_2$  imaginary parts of dielectric constant, indexes of  $\kappa$  absorption and  $n$  refraction,  $n_{\text{ef}}(E)$  effective number of valence electrons taking part in transitions up to the given energy  $E$ ,  $\varepsilon_{0,\text{eff}}(E)$  effective static dielectric constant, functions of character  $-Img \varepsilon^{-1}$  volume and  $-Img(\varepsilon+1)^{-1}$  surface electron losses, phase of  $\theta$  reflected light,  $\varepsilon_2 E$  optical conduction, integral function of  $\varepsilon_2 E^2$  bound state density and  $(\alpha, \beta)$  electrooptical differential functions are defined.

Only spectra of  $R(E)$  reflection coefficients,  $\varepsilon_1$  real and  $\varepsilon_2$  imaginary parts of dielectric constant, functions of character  $-Img \varepsilon^{-1}$  volume and  $-Img(\varepsilon+1)^{-1}$  surface electron losses,  $(\alpha, \beta)$  electrooptical differential functions, spectra of  $\sigma$  optical conduction of massive and film samples are presented in fig. 1-12 for brevity and the values of interband optical transitions defined by maximums of  $\sigma$  optical conduction are presented in table 1.

The presented data give the possibility to compare optical spectra, transitions of single crystals and film samples  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn).

The authors of work [5] for the case of high energies for single-crystalline  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) have found the transitions 1,4 eV and 1,1 eV. As it is seen from table 1 the same transitions are observed for  $n$ - and  $p$ - types correspondingly. As it is seen from table 1 the transitions corresponding to corresponding energies 1,4 eV and 1,15 eV for film samples  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) of  $n$ - and  $p$ -types. From this it is concluded that the values of some optical transitions  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) at transition of material from crystalline to non-crystalline state for non-crystalline samples are saved.

As it is mentioned in [18] the study of absorbing transitions in materials is impossible because of absorption big value in region of interband transition energies  $E > E_g$  ( $E_g$  is forbidden band thickness). The reflection is the unique method.

The analytical singularities of imaginary part of  $\varepsilon_2(E)$  complex dielectric constant and functions bound by  $dN/dE$  state densities are always coincide and the gradient of interband states makes the main contribution into  $dN/dE$  functions:

$$\frac{dN_{ij}}{dE} \sim \int \frac{dS_k}{|\nabla_k E_{ij}|}, \quad (1)$$

where  $E_{ij}(k) = E_j(k) - E_i(k)$  is between conduction band and valence band.  $dN/dE$  values near critical points in  $k$ -space defined by  $|\nabla_k E_{ij}| = 0$  expression and also the position of critical points and transition type can be theoretically calculated from band structure

The analysis of  $\varepsilon_2(E)$ ,  $dN/dE$  functions and  $R(E)$  reflection coefficient show that disposition and

character of maximums in their spectra are similar ones or very close to each other. That's why the values of corresponding interband gaps and band nature can be defined with the help of direct comparison of experimental data in  $E > E_g$  region with theoretical calculations of  $dN/dE$  function. As it is mentioned in [18]  $E_0$  resonance frequency presents itself that frequency at which  $2nk \cdot E$  conduction achieves to maximum by which the interband transitions are defined.

The high transparency in wide energy region  $E < E_g$  is character for non-crystalline materials and several methods of  $E_g$  definition are known. Its evaluation by the level of  $\alpha(E)$  absorption coefficient of long-wave edge is the one of them.  $E_g$  exact value for non-crystalline semiconductors is discussion one and as usual the discussion of  $\alpha(E)$  spectrum character in Urbakh and Tauc models is carried out without  $E_g$  evaluations [20,21].  $E_g$  is defined for  $\alpha(E) = 10^3 \text{cm}^{-1}$  value by Tauc model.

As it is mentioned in [20] the state density  $N(E)$  is the similar useful one for crystalline and non-crystalline substances. The state density motion in non-crystalline substance doesn't essentially differ from corresponding one in crystal. The thin structure in 1<sup>st</sup> case can be blurred and local states can appear in forbidden band. The band structure is saved as it is defined by atom short-range order in materials.

The author of work [22] mentions that it is impossible to make the principal boundary between single-crystalline, polycrystalline and amorphous substance states. The presence of the band structure i.e. the forbidden band and conduction band can be derived from the fact of atom short-range existence and it is no necessity to require the periodic atom disposition for such derivation.

As authors [23-25] show that the structure of indirect binary compound can be considered as collection of different basis clusters presented in structural matrix with different static weight and put into effective medium. As authors [26] mention the atom short-range order makes the main contribution in electronic state density. However, as atom short-range order in binary compounds can significantly change from node to node in structural matrix of amorphous substance, especially statistics of basis clusters defines the final electronic state density. Thus, the low-molecular structural configurations that are accompanied by significant benefit in energy can appear at melt cooling or in the process of amorphous substance formation by another way. Such formations can serve neither as crystal germs nor as the centers of solid amorphous phase growth because of the fact that they are characterized by special symmetry.

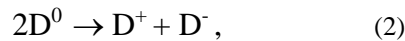
The ambiguity of structural order in compositional amorphous solid states by  $A_x B_{1-x}$  type can be considered as their general property. The peculiarities of atom short-range order near each node are caused firstly by physical methods of  $A_x B_{1-x}$  system preparation and secondly by chemical order principle, taking under consideration «rule 8-N» [20] and value of a quantity of bond energy of neighbor atoms.

The decomposition of ideal structure in non-crystalline solid state takes place in systems including the atoms with not- divided electron couples. It is very often that at the breakage of electron couple is on the one of fragments, i.e. the bond heterolytic breakage. The one positive and one negative charged defect centers in regions of atom short-range order appear. The energy necessary for bond breakage is partly compensated because of existence of non- divided electron couple of atoms being surrounding and number of chemical bonds doesn't change. Thus, the structural disorder exists in homogeneous non - crystalline materials of stoichiometric composition along with density oscillations and existing topological disorder of different types. It is revealed in the form of positive and negative charged defect centers as in the case of point defects in crystals. As a result, the reaction defects, the formations of which are characterized by lowest change of free energy, dominate.

The idea of atom short-range order at formation of electron energy bands is the one of the fundamental conceptions in physics of disordered system. This idea has the experimental and theoretical confirmation on the example of many non-crystalline solid and liquid semiconductors [27]. The mechanism of formation of valence and conduction bands in non-crystalline semiconductors is mainly formed by authors [28, 29]. The similarity of the main peculiarities of spectral dependence of imaginary part dielectric conduction  $\varepsilon_2(E)$  is emphasized in the works of these authors for non-crystalline semiconductors and their crystal analogues. This similarity is shown by author [28] on the example of selenium. The analogous conclusion is made in the relation to  $\alpha\text{-As}_2\text{S}_3$  and  $\alpha\text{-As}_2\text{Se}_3$  in works [28, 29].

Only maximum smoothing takes place in their  $\varepsilon_2(E)$  for the cases of amorphous materials and similarity of the curves for amorphous and crystalline samples is preserved.

Nowadays, it is established that one can directly change optical, photoelectric and electric properties of non-crystalline semiconductors with the change of chemical composition and also by impurity introduction. The concentration changes of charged defect centers  $D^+$  и  $D^-$  ( $U^{-1}$  are centers) takes place in them and these defects form from initial neutral defects  $D^0$  by reaction:



which can be eigen or impurity and mixed defects giving the possibility to control by their physical properties.

Thus, reflection coefficients  $R(E)$  of single crystals and film samples  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) of  $n$ - and  $p$ -types in beam energy interval 1÷6 eV falling normally to the surface are measured in the work. The measurements are carried out parallel and perpendicular to  $C$  axis for the case of  $\text{Bi}_2\text{Te}_3$  single crystals. It is shown that values of some optical

transitions  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) from crystalline to non-crystalline state are preserved for the non-crystalline samples.

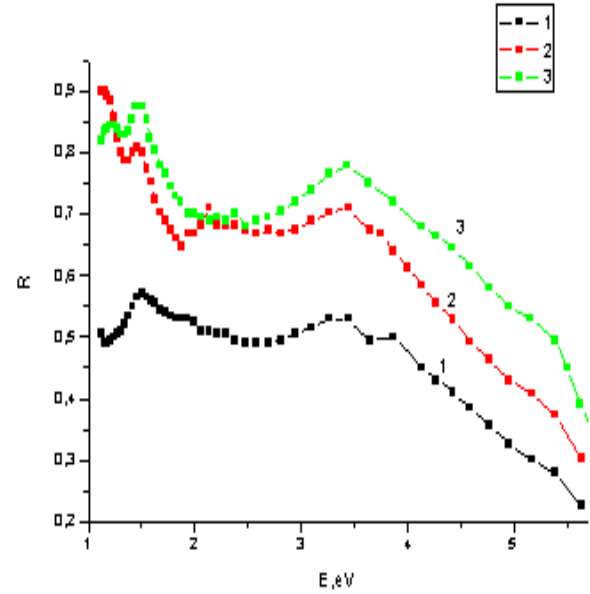


Fig. 1. Reflection spectra ( $R$ ) of single crystals: 1 -  $\text{Bi}_2\text{Te}_3$  (Cu), 2 -  $\text{Bi}_2\text{Te}_3$  (Ni), 3 -  $\text{Bi}_2\text{Te}_3$  (Zn).

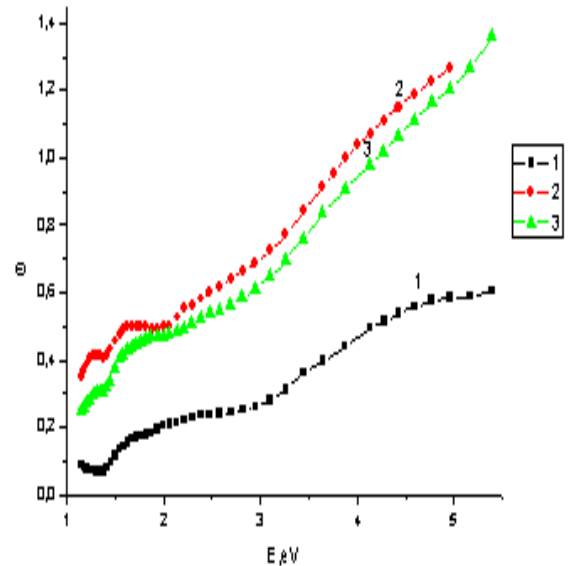


Fig. 2. Reflection phase spectra ( $\theta$ ) of single crystals: 1 -  $\text{Bi}_2\text{Te}_3$  (Cu), 2 -  $\text{Bi}_2\text{Te}_3$  (Ni), 3 -  $\text{Bi}_2\text{Te}_3$  (Zn).

Only reflection coefficient spectra,  $\varepsilon_1$  real and  $\varepsilon_2$  imaginary parts of dielectric constant, functions of character  $-\text{Im}g\varepsilon^{-1}$  volume and  $-\text{Im}g(\varepsilon+1)^{-1}$  surface electron losses, the spectra of  $(\alpha, \beta)$  electrooptical differential functions, the spectra of optical conduction of  $\sigma$  massive and film samples correspondingly are shown for brevity in fig. 1 -12 and the values of interband optical transitions defined by  $\sigma$  optical conduction maximums are shown in table.

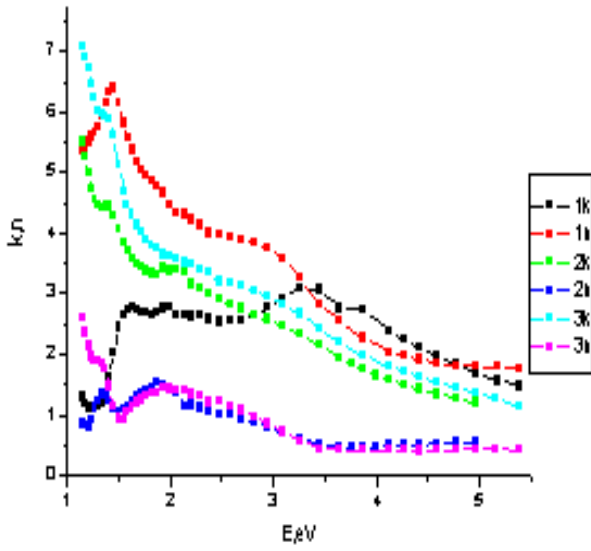


Fig. 3.  $k$  and  $n$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3(\text{Cu})$ , 2 -  $\text{Bi}_2\text{Te}_3(\text{Ni})$ , 3 -  $\text{Bi}_2\text{Te}_3(\text{Zn})$ .

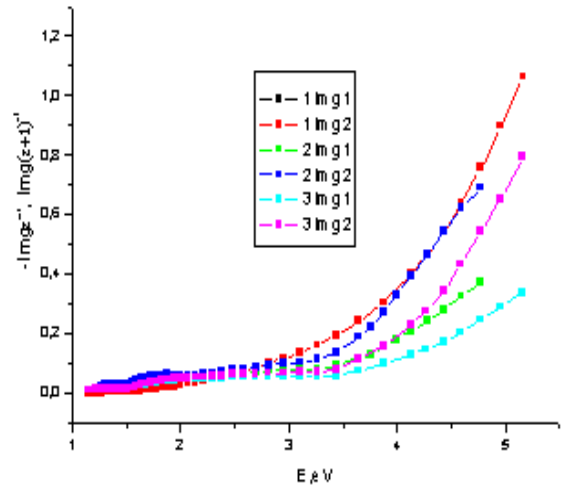


Fig. 6.  $-\text{Im}g\epsilon-1$  and  $-\text{Im}g(\epsilon+1)^{-1}$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3(\text{Cu})$ , 2 -  $\text{Bi}_2\text{Te}_3(\text{Ni})$ , 3 -  $\text{Bi}_2\text{Te}_3(\text{Zn})$ .

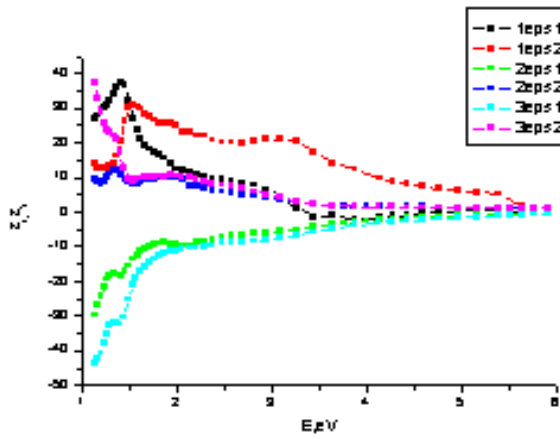


Fig. 4.  $\epsilon_1$  and  $\epsilon_2$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3(\text{Cu})$ , 2 -  $\text{Bi}_2\text{Te}_3(\text{Ni})$ , 3 -  $\text{Bi}_2\text{Te}_3(\text{Zn})$ .

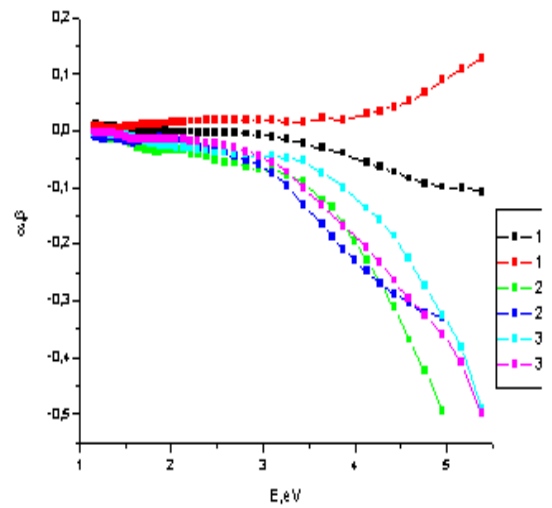


Fig. 7.  $\alpha$  and  $\beta$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3(\text{Cu})$ , 2 -  $\text{Bi}_2\text{Te}_3(\text{Ni})$ , 3 -  $\text{Bi}_2\text{Te}_3(\text{Zn})$ .

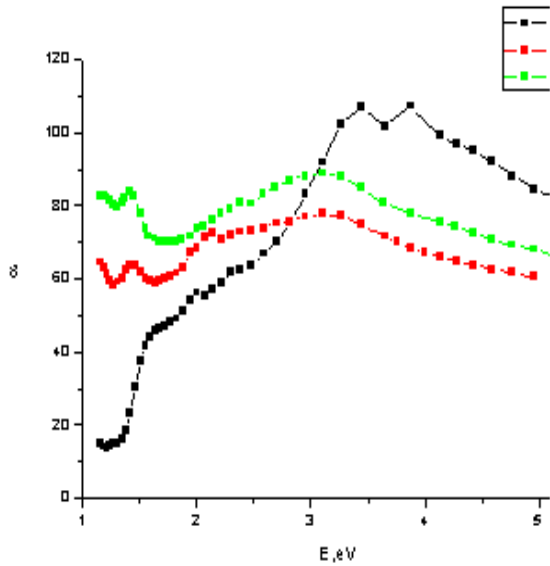


Fig. 5.  $\alpha$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3(\text{Cu})$ , 2 -  $\text{Bi}_2\text{Te}_3(\text{Ni})$ , 3 -  $\text{Bi}_2\text{Te}_3(\text{Zn})$ .

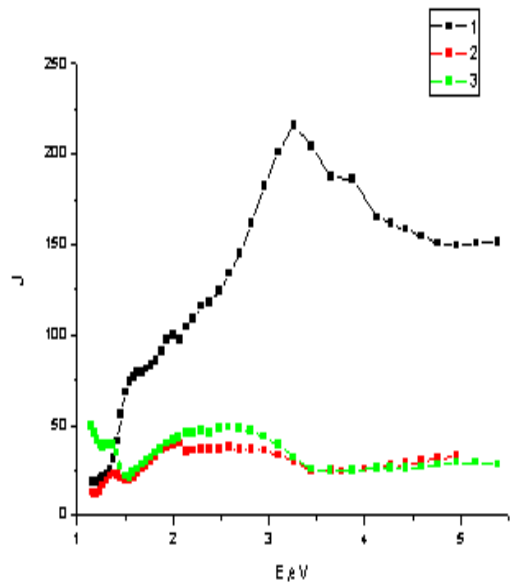


Fig. 8. The state density spectra ( $J$ ) of single crystals: 1 -  $\text{Bi}_2\text{Te}_3(\text{Cu})$ , 2 -  $\text{Bi}_2\text{Te}_3(\text{Ni})$ , 3 -  $\text{Bi}_2\text{Te}_3(\text{Zn})$ .

**OPTICAL PARAMETER SPECTRA OF  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) SINGLE CRYSTALS**

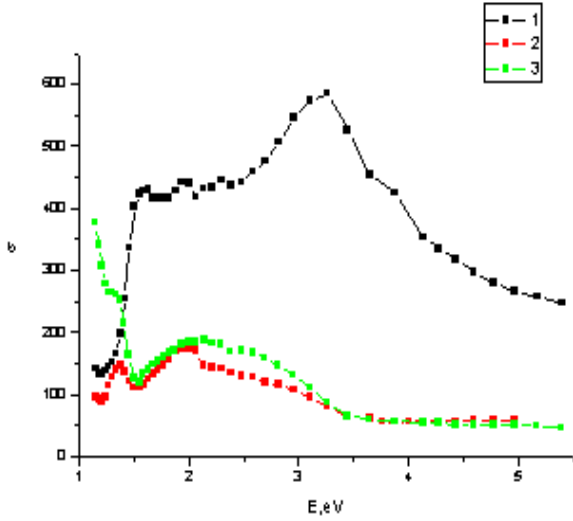


Fig. 9. Optical conduction ( $\sigma$ ) of single crystals: 1 -  $\text{Bi}_2\text{Te}_3$  (Cu), 2 -  $\text{Bi}_2\text{Te}_3$  (Ni), 3 -  $\text{Bi}_2\text{Te}_3$  (Zn).

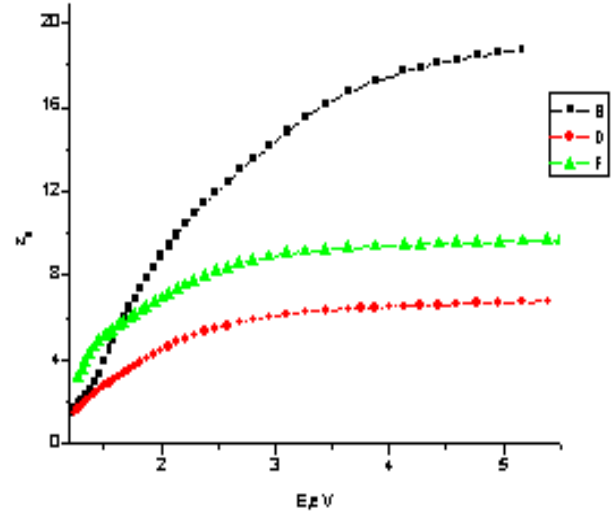


Fig. 11.  $\epsilon_0$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3$  (Cu), 2 -  $\text{Bi}_2\text{Te}_3$  (Ni), 3 -  $\text{Bi}_2\text{Te}_3$  (Zn).

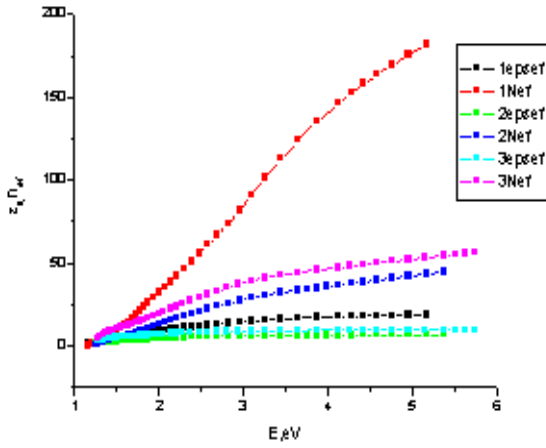


Fig. 10.  $\epsilon_0$  and  $n_{\text{eff}}$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3$  (Cu), 2 -  $\text{Bi}_2\text{Te}_3$  (Ni), 3 -  $\text{Bi}_2\text{Te}_3$  (Zn).

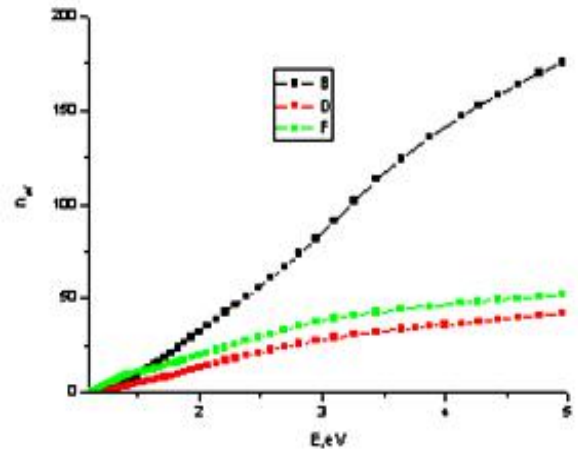


Fig. 12.  $n_{\text{eff}}$  spectra of single crystals: 1 -  $\text{Bi}_2\text{Te}_3$  (Cu), 2 -  $\text{Bi}_2\text{Te}_3$  (Ni), 3 -  $\text{Bi}_2\text{Te}_3$  (Zn).

Table

The optical transitions in  $\text{Bi}_2\text{Te}_3$  (Ni, Cu, Zn) single crystals in 1 ÷ 6 eV energy interval of  $n$ - and  $p$ - types parallel and perpendicular to  $C$  axis defined by optical conduction.

$n$ - type parallel to $C$ axis	$n$ - type perpendicular to $C$ axis	$p$ - type parallel to $C$ axis	$p$ - type perpendicular to $C$ axis
1.01	0.70	1.41	1.19
1.10	0.79	2.02	1.27
1.14	1.29	2.29	1.37
1.36	1.86	2.96	1.81
1.94	2.07	3.18	1.92
2.16	2.17	7.21	2.10
2.28	2.31	7.41	2.23
2.44	2.58	-	2.42
2.61	2.71	-	2.99
3.02	3.09	-	4.95
4.24	3.95	-	6.98
5.06	4.61	-	7.10
7.08	6.45	-	-
7.22	6.81	-	-
7.38	6.97	-	-
7.69	7.12	-	-
8.10	7.41	-	-
-	8.08	-	-
-	8.32	-	-

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