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Photoelectrical properties of In/n-$CuIn_5Se_8$ Schottky barriers

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Abstract

$CuIn_5Se_8$ homogeneous crystals of n-type conductivity have been grown. Donor centers activation energy has been estimated. In/n-$CuIn_5Se_8$ Schottky barriers have been created and the first spectral dependencies of quantum efficiency of photoconversion of these structures have been derived. The nature of interband optical transitions has been interpreted and the band gap values for direct and indirect transitions in $CuIn_5Se_8$ crystals have been determined on the results of analysis of the Schottky barriers photoactive edge absorption. A possibility of utilization of $CuIn_5Se_8$ crystals in wide-band photoconverters of the optical radiation has been established.
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1. Introduction

Characteristic properties of interatomic interaction in the system of I-III-VI elements have opened the possibilities of creating a family of new formation concept of diamond-like phases, which properties are determined by an atomic composition and a character of positional ordering of the atoms involved in their composition [1–4]. Such interaction have opened new ways in the management of compound properties with the general formulae I III_{2n+1} VI_{3n+2} (n=0,1,2...) and stimulated the necessity to study the interconnections of the physical properties of this class of complex semiconductors with the atomic composition, which is defined in fact by the value of n in the general formulae of such substances. Already the first investigations of such type of interatomic interactions have shown, that the $CuIn_3Se_5$ compound is formed at n=1 [2], which differs on physical properties from the earlier investigated compound $CuInSe_2$ [5]. Information on growth and first investigations of bulk crystals of the $CuIn_5Se_8$ ternary compound to which is corresponded n=2 and also creation and investigation of the first photosensitive structures on the base of this compound in I-III-VI ternary system has been presented in this work.

2. Experimental

$CuIn_5Se_8$ crystals have been grown by the directional crystallization of the melt close to stoichiometry with vertical arrangement of a quartz crucible. The large-block ingots had the average dimensions: ~12 mm in diameter and up to 40 mm in length. In contrast to $CuIn_3Se_5$ a pronounced cleavage is characteristic for $CuIn_5Se_8$ ingots, which gives an opportunity to make plane-parallel plates with mirror-line perfect planes by a simple splitting.

The investigation of X-ray diffraction patterns of $CuIn_5Se_8$ grown crystals has shown, that this ternary compound in contrast to $CuIn_3Se_5$ crystallizes in hexagonal structure. The unit cell parameters calculated by the least squares method from the lines with angles $2\Theta > 60°$ are equal to a=4.038±0.002 Å and c=32.782±0.005 Å at T=300 K. Hence, a change in atomic composition in the row of $CuIn_{2n+1}Se_{3n+2}$ compounds is accompanied by a change of the type of crystal structure. $CuInSe_2$ reveals chalcopyrite structure at n=0 [2], but at n=2

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$CuIn_5Se_8$ compound with hexagonal structure arise already. So, a change in atomic composition in the Cu-In-Se system is accompanied by a reconstruction in the crystal structure of the substance.

According to the determination of the thermovoltage sign all $CuIn_5Se_8$ crystals obtained in the given work reveal an electron type of conductivity, what has not been observed earlier for representatives of compounds of this class like $CuInSe_2$ and $CuIn_3Se_5$ [4–6]. Hence, changes in composition in a row of $CuIn_{2n+1}Se_{3n+2}$ compounds cause conversion of the type of conductivity from p to n. This circumstance takes into consideration the same degree of purity of the components used for the preparation of the given compounds, and that the main type of lattice defects in such crystals at the indicated changes of the atomic composition undergoes the essential changes. The resistivity ρ for n-$CuIn_5Se_8$ crystals, as it can be seen from the Table 1, was found to be some orders of magnitude smaller than in the case of $CuIn_3Se_5$. A typical temperature dependency $\rho(T)$ for one of the n-$CuIn_5Se_8$ samples has been presented in Fig. 1. The samples for measuring were prepared by splitting from the ingots and had mean dimensions $0.1 \times 2 \times 6$ mm^3. Ohmic contacts were formed by an electrical discharge between silver conductors (50 μm in diameter) and the necessary spots in the samples. Probe measurements of ρ testify that the difference in resistivity on various pairs of the probe of the same sample do not exceed ±3%. This circumstance is the base for the conclusion, that the developed method of growth of $CuIn_5Se_8$ crystals allows to produce electrically homogeneous crystals of the given ternary compound.

3. Results and discussion

It is seen from Fig. 1 that the exponential dependency of resistivity on temperature displays clearly in the interval of temperatures under investigation T=300–400 K [7]:

$$\rho = \rho_0 \exp\left(\frac{E_d}{kT}\right), \quad (1)$$

where k–Boltzmann constant, E_D=0.28 eV–activation energy of donor centers, determined in an assumption of a high degree of their compensation by acceptors. It is seen from Table 1, that the transition of crystals from $CuIn_3Se_5$ to $CuIn_5Se_8$ leads to an the essential drop of the activation energy of the dominated centers have taken place at the same time with the decrease of the resistivity of the substance. One should bear in mind in this case, that the measurement results presented here concern a current transfer in the direction, which is perpendicular to the hexagonal axis c and therefore they can be connected with $\rho\perp$ component. Unfortunately, for the present we couldn't be able to prepare samples for current transfer investigation in n-$CuIn_5Se_8$ single crystals along c axis and therefore to discuss the possible current transfer anisotropy is not possible so far. It should be indicated also, that the problem of making a complete set of crystallographic orientations of samples from layered crystals has not been solved yet even for more investigated and simpler in composition crystals, for example, InSe.

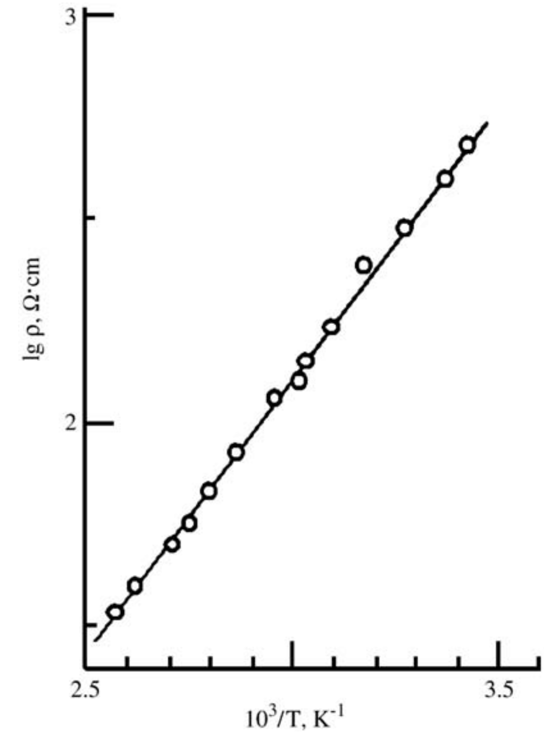

Fig. 1. Temperature dependency of the resistivity of n-$CuIn_5Se_8$ crystals.

Fresh-cleaved mirror-like (001) $CuIn_5Se_8$ planes have been used for making of Schottky barriers by vacuum deposition of thin (d=0.05–0.1 mm) films of pure indium. A possibility of making such cleavages is determined by anisotropy of interatomic bounds in $CuIn_5Se_8$ and, naturally, allows to exclude from the technology of making barriers such important operations, as a mechanical and chemical treatment of a samples surface [8]. The typical thicknesses of the cleaved plates are about from 0.05 mm to 12 mm in diameter.

The investigation of the firstly made In/n-$CuIn_5Se_8$ Schottky barriers have shown, that the structures made by us display a pronounced rectification. The usual ratio of a direct current to reverse current at bias $U \approx 2$ V did not exceed 2, what is associated hypothetically with structure periphery imperfections. The forward direction in these barriers have corresponded to negative polarity of the external bias on the semiconductor. Photovoltage is always arisen at lighting and it predominates in

Table 1
Photoelectrical properties of n-$CuIn_5Se_8$, p-$CuIn_3Se_5$ crystals and In/n-$CuIn_5Se_8$, In/p-$CuIn_3Se_5$ Schottky barriers at T=300 K

Crystal	ρ, Ω cm	$E_{D,A}$, eV	Structure	$\hbar\omega^m$, eV	$\delta_{1/2}$, eV	S_U^m, V/W	E_G^{ind}, eV	E_G^d, eV
n-$CuIn_5Se_8$	10^3–10^4	0.28	In/n-$CuIn_5Se_8$	1.64	1.6	100	0.99	1.07
p-$CuIn_3Se_5$	10^7	0.4–0.7	In/p-$CuIn_3Se_5$	1.25	0.9	1250	1.04	1.12

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