EFFECT OF LOW-DOSE-RATE IONIZING RADIATION ON THE COMPLEX DIELECTRIC PERMITTIVITY OF CdZnTe CRYSTALS

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For the first time, the change in the real and imaginary parts of $Cd_{1-x}Zn_xTe$ crystals complex dielectric permittivity when exposed to ionizing radiation with a small exposure dose rate (about hundreds of μ R/h) has been studied. Significant changes in the values of both parts of complex dielectric permittivity have been revealed. Regularities of specified changes have been established and explained taking into account a different radiation effect on free and bound charges. The basis of effect is the changes in the charge carrier localized states of point defect associates. Their appearance is due to a high concentration and a variety of intrinsic structural defects in the studied crystals as a consequence of the deviation of the crystal composition from the stoichiometric one.

Keywords: localized states, semiconductors, complex dielectric permittivity, cadmium zinc-telluride, ionizing radiation

1. Introduction

It is well known that changes in the electrophysical properties of crystalline semiconductors, as a result of ionizing radiation (IR) exposure, depend not only on the energy characteristics of radiation (energy of quanta or particles, radiation flux density, etc.), but also on the composition and structural perfection of specimens. In Ref. [1], changes in the low-frequency band of the real ε' and imaginary ε'' parts of the complex dielectric permittivity ε^* of Cd₁ Zn₂Te crystals (x = 0.12-0.18) with an increased polarizability due to the effects of ultrasmall exposure doses (5-55 R) of ⁶⁰Co radiation were reported. These changes had been irreversible and were explained by the substantially nonequilibrium state of the crystal intrinsic defects system, which arose due to the deviation of its composition from the stoichiometric one under conditions of growth from the melt. In this regard, it is of interest to study the change in the real and imaginary parts of ε^* of Cd_{1-x}Zn_xTe (or CZT) crystals of the same composition when exposed to IR with a small exposure dose rate (about hundreds of μ R/h). As far as we know, such experiments have not been carried out before. In a number of works (see, e.g. Refs. [2] and [3]), CZT crystals have been studied as ionizing radiation detectors operating in the current mode, i.e. when exposed to a constant electric field. Also, there are works (see, e.g. Ref. [4]) devoted to the effect of IR on the dielectric properties of crystals of different composition. However, in all these works, radiation with the exposure dose rate several orders of magnitude higher than the one specified above was used.

2. Specimens and the routine of experiment

The studied $Cd_{1-x}Zn_xTe$ (x = 0.12-0.18) crystals were grown by the Bridgman method from the melt under a high pressure of inert gas (Ar). Details of the crystal growth, as well as manufacturing of specimens and control of their composition, are given in Ref. [1]. Specimens with a total number of 30 items were manufactured from different parts of three crystal ingots grown under the same growth conditions from the same raw material. The values of ε' and ε'' were measured

by the capacitance method using an *Instek* LCR-819 High Precision LCR meter. An E6-13A teraohmmeter by *Punane RET* was used to measure the resistivity of the specimens to direct current. ¹³⁷Cs isotope was used to obtain ionizing radiation. The exposure dose rate of radiation applied to the specimen was only ~700 μ R/h. The energy spectrum of charge carrier localized states in the crystals was studied by the scanning photodielectric spectroscopy method (SPDS) [5].

3. Results and discussion

It was ascertained that the ¹³⁷Cs radiation of the specified exposure dose rate does not cause a noticeable effect on the electrical resistivity of CZT crystals in a constant electric field. At the same time, the same radiation exposure causes significant changes in the values of both parts of the complex dielectric permittivity ε^* . Moreover, only in 10% of the specimens, the response of these values to the IR exposure had a spasmodic nature (Fig. 1(a)). For the remaining part of the specimens more long-term changes in these quantities were observed both during the radiation exposure and after the exposure (Fig. 1(b)). Therefore, for each specimen, the change in the quantities of ε' and ε'' under IR exposure was characterized by averaging their 10 values measured initially, as well as under radiation exposure. The difference between the average values of these quantities was calculated:

$$\Delta \varepsilon_{i}^{\prime} = \langle \varepsilon_{yi}^{\prime} \rangle - \langle \varepsilon_{i}^{\prime} \rangle,$$

$$\Delta \varepsilon_{i}^{\prime\prime} = \langle \varepsilon_{yi}^{\prime\prime} \rangle - \langle \varepsilon_{i}^{\prime\prime} \rangle.$$
(1)

In this expression, the index γ is provided to the quantities measured under IR exposure. It is important to note that the parameters $\Delta \varepsilon'_i$ and $\Delta \varepsilon''_i$ for some specimens had a positive sign, while for the others they had a negative sign. For this reason, the change in the parts of ε^* for the entire set of specimens was characterized by averaging over it the absolute values of the specified parameters.



Fig. 1. Time dependences for the parts of the complex dielectric permittivity observed before and after the exposure of ionizing radiation (light circles) and during the exposure (dark circles): (a) spasmodic changes caused by radiation exposure, (b) long-term changes in the complex dielectric permittivity.

As a result, the parameters $\langle |\Delta \varepsilon_i'| \rangle$ and $\langle |\Delta \varepsilon_i''| \rangle$ were obtained. Their frequency dependences are shown in Fig. 2. As we see, on a double logarithmic scale, these dependences have a form of linear segments forming a negative angle with the frequency axis. This allows us to conclude that the studied specimens are characterized by a decrease in the absolute values of parameters $\Delta \varepsilon_i'$ and $\Delta \varepsilon_i''$ with the frequency under the law $\Delta \varepsilon_i'(\Delta \varepsilon_i'') \sim f^{\gamma}(\gamma \text{ is a constant})$.

To establish the relation between the changes in quantity ε^* under IR exposure and the point structure defects that had induced localized states of charge carriers in the band gap, the energy spectrum of these states has been measured by the SPDS method. Experiments have been carried out on the samples with the highest absolute values of the parameters $\Delta \varepsilon'_i$ and $\Delta \varepsilon''_i$. This choice of samples is dictated by the need to ensure the highest method sensitivity. Table 1 shows the typical values of the depths for the energy levels of localized states with regard to the top of the valence band in the initial state of the crystal, as well as under



Fig. 2. Frequency dependences for the absolute values of changes in real (a) and imaginary (b) parts of the complex dielectric permittivity, averaged over all samples.

SPDS method		TSC, TEES,	
Initial state	Under ionizing radia- tion exposition	PICTS methods [6]	Supposed origin of the states [6]
Energy, eV	Energy, eV	Energy, eV	
E_{V} + 0.10	$E_{V} + 0.10$		Complex impurity- V_{Cd}
E_{V} + 0.13		$E_{v} + 0.10 - 0.16$	A-centre
$E_{V} + 0.15$	$E_{V} + 0.15$		Impurity in Cd sites
$E_{V} + 0.17$	$E_{V} + 0.17$		
E_{V} + 0.22	E_{V} + 0.22	$E_v + 0.19 - 0.22$	$V_{ m Te}$
	$E_{V} + 0.24$		
E_{V} + 0.28	E_{V} + 0.28	$E_v + 0.27 - 0.29$	C, Si atoms
$E_{V} + 0.37$	$E_{V} + 0.37$	$E_v + 0.35 - 0.41$	Complex V_{Cd}
E_{V} + 0.47		E_v + 0.46–0.50	Structure defect connected with deformation
E_{V} + 0.52			
		$E_{v} + 0.60$	Zn atoms in a complex
		E_{v} + 0.75	Phonon impurities and vacancies

Table 1. Energy spectra of charge carriers localized states in $Cd_{1-x}Zn_xTe$ crystals, revealed by different methods.

IR exposure. Table 1 also contains the known data on the depth and supposed origin of the localized states in CZT crystals. As we can see, when exposed to radiation, the states with a depth of E_v + 0.13 eV, E_v + 0.47 eV and E_v + 0.52 eV disappeared, but the states with a depth of E_v + 0.24 eV appeared. Note that all the states that we observed agree with those known from the literature [6].

When explaining the described regularities of IR effect on the complex dielectric permittivity of CZT crystals, we assume that radiation causes excitation of both electronic [7] and elastic (see, e.g. Ref. [8]) crystal subsystems. In this case, various processes occur in the crystal, causing a change in the concentration of not only free charge carriers, but also a change in the characteristics of the subsystem of carriers localized on the states in the crystal band gap. It should be noted that while considering the electric polarization such localized charge carriers are usually called 'bound' (see, e.g. Ref. [9]).

Apparently, the main feature of the investigated CZT crystals is that, under the conditions of the experiments (room temperature, low radiation level and weak measuring electric field), the response of crystals to this field is determined by the behaviour of exact bound carriers. This assumption is consistent with the above-noted change in the energy spectrum of carrier localized states for the specimens with the greatest absolute values of the parameters $\Delta \varepsilon'_i$ and $\Delta \varepsilon''_i$ (see Table 1). In our opinion, this feature of the crystals is due to two factors.

The first factor is the high concentration (up to 10¹⁶ cm⁻³ [10]) and a variety of types [11] of point defects in the structure of crystals. This is due to the deviation of the crystal composition from the stoichiometric [12] one and the presence of background (uncontrolled) impurities.

The second factor is related to the diversity of point defects, namely, the presence of point defect associates of various types in crystals [13–15]. It is natural to assume that, under IR exposure, some of these defects pass through reconstruction, and, as a result, their polarizability changes. Moreover, depending on the type of associates, their reconstruction can be accompanied by both an increase and a decrease in their polarizability. In this case, the contribution of associates to the parts of ε^* changes in a similar way. Due to the compositional inhomogeneity inherent in the studied solid solu-

tions [12] and, correspondingly, to the difference in the sets of point defects in individual specimens, their parameters $\Delta \varepsilon'_i$ and $\Delta \varepsilon''_i$ are also different. The inertia of reconstruction process of the mentioned defects causes a weakening of the IR effect on the parameters $\langle |\Delta \varepsilon'_i| \rangle$ and $\langle |\Delta \varepsilon''_i| \rangle$ with the frequency (see Fig. 2).

4. Conclusions

In conclusion, the flux of ionizing radiation from the ¹³⁷Cs isotope with an essentially low exposure dose rate has a significant effect on the complex dielectric permittivity of $Cd_{1-x}Zn_xTe$ crystals in the low-frequency band. However, with such radiation exposure, no changes in the resistivity of crystals to direct current have been detected. For different specimens, the changes in the parts of dielectric permittivity could differ in signs. The absolute values of these changes averaged over the set of specimens decrease with the frequency of the electric field. This behaviour of the complex dielectric permittivity under radiation exposure is explained by a change in the state of point defect associates that have arisen during crystal growth from the melt.

References

- V.K. Komar, S.V. Sulima, O.N. Chugai, S.L. Abashin, O.T. Nikolov, S.V. Oleinik, V.M. Puzikov, I.S. Terzin, and Yu.A. Yatsina, Effect of ionizing radiation in ultrasmall doses on dielectric properties of CdZnTe crystals with anomalously high polarizability, Tech. Phys. Lett. **37**, 589 (2011), https://doi.org/10.1134/S106378501107008X
- [2] V.F. Dvoryankin, G.G. Dvoryankina, A.A. Kudryashov, A.G. Petrov, V.D. Golyshev, and S.V. Bykova, X-ray sensitivity of Cd_{0.9}Zn_{0.1}Te detectors, Tech. Phys. 55, 306–308 (2010), https://doi.org/10.1134/S1063784210020246
- [3] V.F. Dvoryankin, G.G. Dvoryankina, A.A. Kudryashov, A.G. Petrov, A.A. Davydov, N.V. Zhavoronkov, and D.V. Kapkin, X-ray detectors based on CdZnTe crystals grown from the vapor phase, Tech. Phys. 57, 1462–1464 (2012), https://doi.org/10.1134/S1063784212100040
- [4] A.U. Sheleg, K.V. Iodkovskaya, and N.F. Kurilovich, Effect of gamma radiation on the permittivity and electrical conductivity of TlGaS, crystals,

- [5] V.K. Komar, V.P. Migal, O.N. Chugai, V.M. Puzikov, D.P. Nalivaiko, and N.N. Grebenyuk, Investigation of localized states in cadmium zinc telluride crystals by scanning photodielectric spectroscopy, Appl. Phys. Lett. 81, 4195–4197 (2002), https://doi.org/10.1063/1.1525883
- [6] P. Fougeres, P. Siffert, M. Hageali, J.M. Koebel, and R. Regal, CdTe and Cd_{1-x}Zn_xTe for nuclear detectors: facts and fictions, Nucl. Instrum. Methods Phys. Res. A **428**, 38–44 (1999), https://doi.org/10.1016/S0168-9002(98)01578-2
- [7] E. Zarkadoula, G. Samolyuk, and W.J. Weber, Effects of electronic excitation in 150 keV Ni ion irradiation of metallic systems, AIP Adv. 8, 015121 (2018), https://doi.org/10.1063/1.5016536
- [8] Y. Zhang and W.J. Weber, Ion irradiation and modification: The role of coupled electronic and nuclear energy dissipation and subsequent nonequilibrium processes in materials, Appl. Phys. Rev. 7, 041307 (2020), https://doi.org/10.1063/5.0027462
- [9] A.K. Verma, Introduction to Modern Planar Transmission Lines: Physical, Analytical, and Circuit Models Approach, Ch. 6 (Wiley-IEEE Press, 2021) pp. 159–211, https://doi. org/10.1002/9781119632443.ch6
- [10]L. Xu, T. Feng, and W. Jie, Defect levels characterized by photoconductivity and thermally stimu-

lated current in CdZnTe crystals, J. Cryst. Growth **560**, 126050 (2021), https://doi.org/10.1016/j. jcrysgro.2021.126050

- [11]Y. Li, G. Zha, D. Wei, F. Yang, J. Dong, S. Xi, L. Xu, and W. Jie, Effect of deep-level defects on the performance of CdZnTe photon counting detectors, Sensors 20, 2032 (2020), https://doi.org/10.3390/ s20072032
- [12] T.E. Schlesinger, J.E. Toney, H. Yoon, E.Y. Lee, B.A. Brunett, L. Franks, and R.B. James, Cadmium zinc telluride and its use as a nuclear radiation detector material, Mater. Sci. Eng. R Rep. 32(4–5), 103–189 (2001), https://doi.org/10.1016/S0927-796X(01)00027-4
- [13] D.E. Onopko and A.I. Ryskin, Physical foundations of metastable impurity center reconstruction in semiconductors, Semiconductors 35, 1223– 1230 (2001), https://doi.org/10.1134/1.1418062
- [14]A.G. Nikitina and V.V. Zuev, Bistable amphoteric centers in semiconductors, Semiconductors 42, 142–148 (2008), https://doi.org/10.1134/S1063782608020048
- [15]T. Thio, J.W. Bennett, D.J. Chadi, R.A. Linke, and P. Becla, DX centres in CdZnTe: Cl and their applications, J. Cryst. Growth **159**(1–4), 345–349 (1996), https://doi.org/10.1016/0022-0248(95)00681-8

MAŽŲ DOZĖS GALIŲ JONIZUOJANČIOSIOS SPINDULIUOTĖS POVEIKIS CdZnTe KRISTALŲ KOMPLEKSINEI DIELEKTRINEI SKVARBAI

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