



EFFECT OF GAMMA IRRADIATION ON ELECTRICAL AND PHOTOELECTRICAL PROPERTIES OF $Cd_{1-x}Mn_xTe$ THIN FILMS

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Abstract: $A^{II}B^{VI}$ groups of compound semiconductors, their solid solutions, bulk and thin films on their base have wide application possibilities in modern microelectronics. Creation of photosensitive elements and structures on the base of these semiconductors and investigations of their usage potentials in various fields of optoelectronics is one of the most actual problems of modern materials science and engineering. Recently, $A^{II}(TM = Mn, Fe, Co, \text{etc})B^{VI}$ based semimagnetic semiconductors (SMSC) compounds has attracted increasing interest since they considered a promising material for the fabrication of high-performance room-temperature γ - and x -ray detectors. Such detectors offer great potential for nonproliferation applications, medical imaging, astrophysics research, and monitoring industrial processes, because they are able to operate at ambient temperatures, while providing a high detection efficiency and good energy resolution. The aim of the work is to determine of photosensitivity range of $Cd_{1-x}Mn_xTe$ SMSC thin films, to investigate their sensitivity to γ - irradiation and to study the creation possibility of γ - ray detectors on the base these thin films. In these respect, VAC and photoconductivity, as well as effect of γ - ray irradiation on these properties has been studied. Results of current investigation reveal that, $Cd_{1-x}Mn_xTe$ ($x=0.07$) epitaxial films of compounds are sensitive to γ -ray irradiation, indicating that these materials can be considered as a good candidate for γ -ray detector applications.

Keywords: $A^{II}B^{VI}$ compounds, $CdMnTe$, electrical and photoelectrical properties, γ -ray irradiation

Introduction.

In undertaking national security and nonproliferation inspections, radiation detectors mainly serve to locate and identify special radioactive nuclear materials. These applications especially demand portable, lightweight radiation detectors with high resolution. For more than four decades, CdTe has been known as a good candidate material for radiation detection. Detectors on the base of CdTe offer high conversion efficiency of photons to electronic charge carriers as compared to scintillators; consequently, for high-quality CdTe detectors, the energy resolution is expected to be considerably better than scintillators [1, 2]. More importantly, CdTe radiation detectors operate at room temperature, thereby obviating the need for a complicated cooling system and, accordingly, affording greatly enlarged application fields. However, a relatively high leakage current has proven to be a limiting factor during the development of large volume CdTe radiation detectors. In addition, CdTe detectors are relatively expensive, because it is difficult to grow large-volume CdTe crystals of acceptable quality, yield and difficult mechanism of crystal cutting [1,2]. On the basis of the utility of CdTe, the CdZnTe (CZT) compounds was explored carefully, because the introduction of Zn increases the band gap and reduces the leakage current (and noise) of radiation detectors [3-7]. It also is less expensive and easier to produce large-volume single crystals with CZT than with CdTe. Since the first practical CZT gamma-ray detector reported in 1992 [8], there have been many advances in

the performance of the devices. The high-resistivity CZT crystals required for radiation detection initially were grown using the high-pressure Bridgman method (HPB). In recent years, other methods were employed, such as the travelling heater method (THM) and the modified Bridgman method (MB). Today, large-volume CZT single crystals up to hundreds of cubic centimeters can be produced due to improvements in the growth techniques [1, 2, 9]. Such progress in bettering the quality of the CZT material for CZT radiation detectors and electron-transport-only device designs, as well as improving the related electronics greatly have enlarged their application fields and accelerated their commercial marketing [1,2].

The International Atomic Energy Agency (IAEA) has used CdTe and CZT detectors for over three decades [10, 11]. Recently, their usage significantly increased, stimulated by the improvement of material properties, their improved availability, and the development of improved detector-specific, low-noise integrated circuits for electronic readouts. Large-volume CZT detectors also are needed in nonproliferation inspections, because they potentially reduce the measurement time significantly. This is especially important in verifying unirradiated nuclear material where count rates are low and typical gamma lines of uranium or plutonium gamma spectra must be resolved. However, the large-volume CdTe/CZT detectors required for these inspections presently are limited by the lack of CZT single crystals whose active volume is more than about 500 mm³. The dearth of larger volume CdTe/CZT single crystals must be resolved before their widespread application in nonproliferation inspections [1,2]. Recently, scientists at Brookhaven National Laboratory (BNL) developed a hand-held gamma-ray spectrometer for nonproliferation inspections based on virtual Frisch-grid CZT detectors [1,2, 12-16]. The whole system achieves an effective detection volume of 19.2 cm³, that is, 10 times larger than commercial co-planar grid (CPG) CZT detectors. Consequently, detection efficiency is improved significantly. The system employs an 8x8 virtual Frisch-grid CZT detector array; each detector is 5x5x12 mm³. By using front-end application-specific integrated circuits (ASICs) developed at BNL, this spectrometer has a small profile and high energy-resolution. Further, its relatively simple configuration greatly lowers the cost. This achievement has allowed to build an inexpensive, large-volume detector array with high energy resolution and high detection efficiency, so affording wide application potentials in national security and nonproliferation inspections. Moreover, the detector modules are scalable to address a larger range of efficiency requirements [2].

The CdMnTe (CMT) SMSC compound is a relatively novel material for radiation detection. CMT is a diluted magnetic compound semiconductor, previously used as Faraday rotators, optical isolators, solar cells, lasers, magnetic field sensors, and infrared detectors [1,2]. In 1999, Burger et al. first investigated the potential application of this material as radiation detector [17]. They proposed that CMT has two main advantages over CZT, namely, better homogeneity and the lower amount of Mn needed to reach the desired band gap, so making it a good competitor to CZT. Most problems with CMT radiation detectors center on the poor quality of the crystals. Until now, it was difficult to obtain CMT single crystals with high resistivity and acceptable carrier transport properties [1,2]. Some recent progress suggests the possibility of a breakthrough in this field, leading to large improvement in the performance of CMT radiation detectors [8-26]. However, several material properties must be improved before CMT can be practically employed for X-ray and gamma-ray detections. First, compared to CZT, the bond ionicity of CMT is higher, entailing a greater tendency for crystallization into a hexagonal structure, but not in the expected zinc-blende structure [8-26]. Also, higher ionicity can generate twins easily in crystals. Second, the resistivity of CMT crystals must be improved. Normally, CMT crystals grown by the Bridgman methods are p-type materials

due to their high concentration of cadmium vacancies (V_{Cd}) the dominant acceptors, and the resistivity of as-grown crystals can be as low as $10-10^3 \Omega \text{ cm}$, thus not satisfying the resistivity requirement of X-ray and gamma-ray radiation detectors. Recently, detector-grade CMT crystals were grown, and the first CMT detector was fabricated Cd_{0.94}Mn_{0.06}Te doped with Vanadium $5 \times 10^{16} \text{ cm}^{-3}$. However, compared with the performance of CZT detector, there still is much room to improve CMT detectors before they can be applied as practical radiation detectors. The limited availability of CMT crystals to researchers also inhibits its development as a radiation detector. As a result, they remain at the stage of laboratory development and have not been incorporated practically in commercial detection systems [8-26].

In order to overcome the problems arising in the design and development of X- and gamma-ray detectors on the base of defect-free massive CMT single crystals we are offering, in this present study, fabrication of these detectors on the base of epitaxial thin films of Cd_{1-x}(TM)_xTe SMSC compounds for the first time. The development of molecular beam epitaxy (MBE) in the 1970s for arsenic III-V semiconductors has allowed to produce high-quality epitaxial layers with very abrupt interfaces, good control of thickness, doping, and composition. In recent years small-size semiconductors are successfully applied in microelectronics, spintronic, optoelectronics, integral optics, astrophysics, medicine and other fields. One of these semiconductor materials is Cd_{1-x}(TM)_xTe SMSC thin films which are considered to be very perspective and unique for making of detectors of γ - and x-ray [27-31]. Cd_{1-x}(TM)_xTe SMSC thin films have been studied less in comparison with other semiconductors, therefore obtaining of their perfect samples with high crystal perfection, clean and glossy surface, study of surface morphology and crystal structure and investigate of its application perspectives in device-building is one of the scientific problems and of great importance.

Therefore, the aim of the work is determination of photosensitivity range of Cd_{1-x}Mn_xTe SMSC thin films; investigation the photosensitivity of samples, irradiated by γ - ray; to study creation possibility of γ - ray detectors, operating at room temperature and without scintillation material.

In these respect, VAC and photoconductivity, effect of γ - ray irradiation on these properties, as well as possibilities of fabrication γ -ray detectors on the base of Cd_{1-x}Mn_xTe thin films has been studied.

Material and Methods. The powder of synthesized Cd_{1-x}Mn_xTe solid solutions were used to obtain thin films. The crystal structure of synthesized solid solutions has been studied by X-ray diffraction (XRD) method on Bruker D8 Advance XRD. Cd_{1-x}Mn_xTe ($x=0.07$) SMSC compounds thin films were grown on glass substrates by molecular beam condensation (MBC) technique in the vacuum evaporation equipment YBH-71П-3 with steam-oil pumping and nitrogen trap at working pressure of residual gas $(1 \div 2) \cdot 10^{-4} \text{ Pa}$. Glass substrates ($1 \times 1 \text{ cm}$) were cleaned with acetone, methanol and distilled water. The powder of Cd_{1-x}Mn_xTe solid solutions (purity 99.999%) was used to obtain thin films. The thickness of the obtained films was controlled with a micro interferometer microscope МИИ-4. The optical spectra were recorded on a UV-Visible SPECORD 210 PLUS spectrophotometer in the wavelength range $\lambda = 190-1100 \text{ nm}$. Samples irradiated by γ -quanta on Co⁶⁰ isotopes with energies $E = 1.17 \text{ MeV}$ and $E = 1.33 \text{ MeV}$.

Results and discussions. In order to grow Cd_{1-x}Mn_xTe ($x=0.07$) epitaxial films it has been used synthesized samples of Cd_{1-x}Mn_xTe solid solutions of corresponding compositions. The crystal structure of synthesized solid solutions has been studied by XRD method, Fig. 1. The spectra showed that solid solutions of Cd_{1-x}Mn_xTe crystallize on the zinc-blende type crystal lattice structure of CdTe (cubic, sphalerite) with lattice parameters of $a_1=6.4785$, $a_2=6.4775$ and $a_3= 6.4745$.

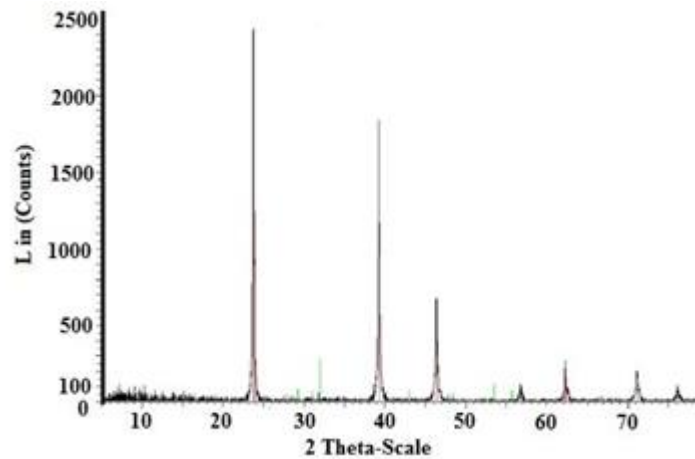
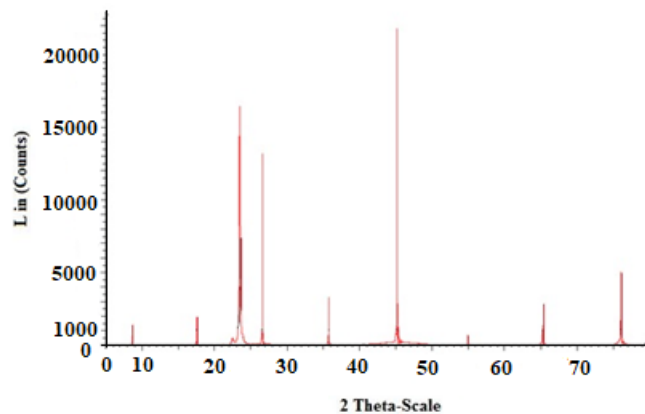
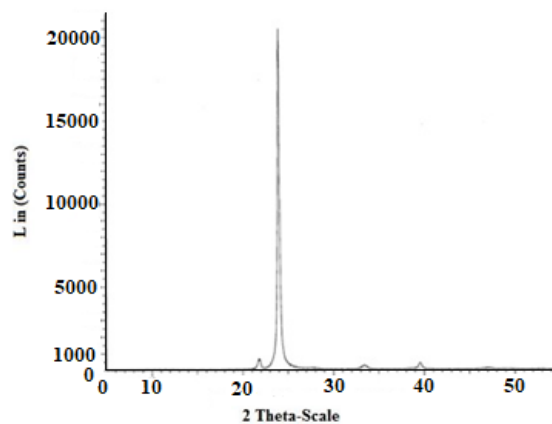


Figure 1. XRD spectrum of $Cd_{1-x}Mn_xTe$ ($x=0.07$) SMSC solid solution

$Cd_{1-x}Mn_xTe$ ($x=0.07$) epitaxial films were obtained on glass substrates by MBC method. By using additional source of Te vapor and controlling temperature, it has been determined the optimum conditions for obtaining $Cd_{1-x}Mn_xTe$ ($x=0.07$) epitaxial films with a perfect structure, clean and smooth surface and also without inclusion of the second phase. The temperature of substrate, temperature of source, condensation rate and film thickness were determined as: $T_{sub} = 300 \div 670$ K, $T_{sour} = 1000 \div 1100$ K, $v = 16 \div 19$ Å/s and $d = 0.5 \div 22$ μm, respectively.



a)



b)

Figure 2. XRD spectra of $Cd_{1-x}Mn_xTe$ ($x=0.07$) thin films obtained at substrate temperatures of
a) $T_{sub} = 300$ K and b) $T_{sub} = 675$ K

The crystal structure of films was studied by XRD method, which revealed that $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ epitaxial films grow on glass substrates at the (111) plane of face-centered cubic lattice having lattice parameter of $a=6.481\text{\AA}$ [32, 33]. It was determined that, at the substrate temperature of $T_{\text{sub}}=300\text{K}$, thin films of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ have an amorphous structure. Moreover, the increase in $T_{\text{sub}} (\geq 470\text{K})$ leads to the formation of polycrystalline films with a cubic crystal structure ($a=6,481\text{\AA}$) and only at substrate temperatures higher than 570 K epitaxial films starts to growth, Fig. 2.

The effect of γ -radiation on the electrophysical, photoelectrical and optical properties of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ thin films obtained on glass substrates were studied. VAC of the initial and γ -irradiated samples of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ epitaxial films has been studied. In $J=f(U)$ dependences of the initial samples of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ there was observed a linear part $J\sim U$ corresponding to Ohm's law and quadratic part $J\sim U^2$ [33]. After irradiation of samples by γ -quanta at a dose of $D_\gamma < 400\text{ Gy}$, a parallel shift of the curve occurs towards the decrease in current. The nature of the dependence does not change, an ohmic and quadratic trap region is observed. The observed pattern shows that when $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ films are irradiated by low doses, a number of deep levels are formed in the band gap, which accept part of the electrons and leads to a decrease in conductivity. At an irradiation dose of $D_\gamma = 400\text{ Gy}$, the conductivity increases, the ohmic part of the VAC lengthens, the quadratic part decreases, a rapid increase is observed behind it and a trapless quadratic part appears. Further irradiation at high doses $D_\gamma \geq 1.5\text{ kGy}$ leads to a decrease in conductivity, Fig. 3 [34-36].

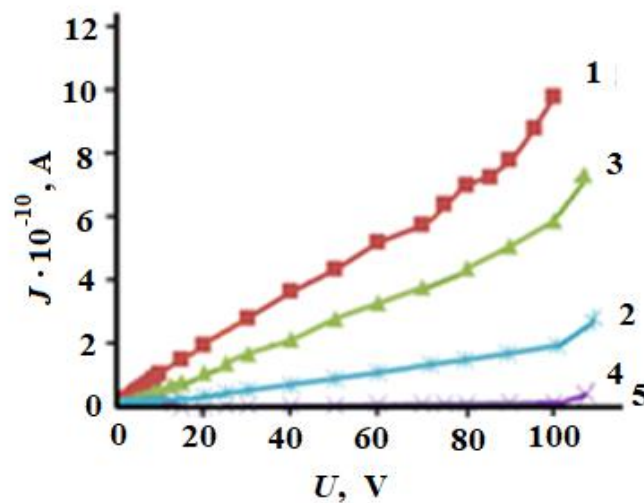


Figure 3. VAC of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ ($x=0.07$) epitaxial films on glass substrates having thickness of $d=15\mu\text{m}$:

1) $D_\gamma = 0$, 2) $D_\gamma = 100\text{ Gy}$, 3) $D_\gamma = 400\text{ Gy}$, 4) $D_\gamma = 1.5\text{ kGy}$ and 5) $D_\gamma = 25\text{ kGy}$

The effect of γ -radiation on the photoconductivity (PC) of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ ($x=0.07$) epitaxial films with a thickness of $d=1\text{ }\mu\text{m}$ and $d=15\text{ }\mu\text{m}$, on glass substrates has been studied. In the initial samples, the PC spectrum of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ epitaxial films has a wide band, the spectral range covers the wavelength region $\lambda=400\text{ nm}-1400\text{ nm}$. The PC maximum corresponds to $\lambda=780\text{ nm}$ for $x=0.05$, and $\lambda=768\text{ nm}$ for $x=0.07$. As the film thickness increases, the PC increases, Fig. 4. Irradiation of epitaxial $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ films at low doses $D_\gamma = 100\text{ Gy}$ leads to an increase in the PC, as well as a slight shift in the PC maximum. This behavior is associated with an increase in the concentration of slow recombination centers (r -centers), which include defects such as Te vacancies, and the influence of shallow-level centers on the lifetime of non-equilibrium holes. Electrons captured by fast recombination centers (s -centers) move to r -centers and increase the lifetime of holes, which leads to an increase in the photocurrent.

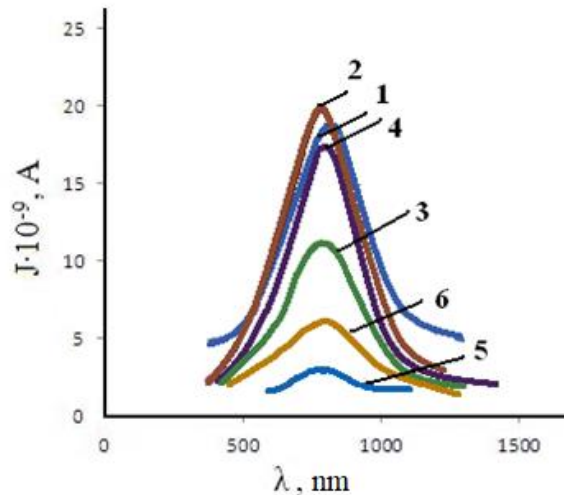


Figure 4. PC of Cd_{1-x}Mn_xTe (x = 0.07) epitaxial films on glass substrates, d = 15 μm, U = 4 V, T_{sub} = 653K, T_{sour} = 1173K, 1)D_γ = 0, 2)D_γ = 100 Gy, 3)D_γ = 200 Gy, 4)D_γ = 300 Gy, 5)D_γ = 2.5 kGy, 6)D_γ = 25 kGy

A further increase in the irradiation dose of γ -quanta up to $D_\gamma = 25$ kGy led to a decrease in the photosensitivity of epitaxial films. A decrease in photosensitivity at higher doses indicates to the emergence of a large number of recombination centers in the band gap with a large capture cross section for current carriers. This change in photosensitivity after irradiation with γ -rays is mainly associated with the restructuring of intrinsic defect levels in the band gap and a change in the degree of hole filling of sensitivity centers. When irradiated with a dose of $D_\gamma = 25$ kGy, the PC increases [34-36]. A significant change in the photoconductivity of Cd_{1-x}Mn_xTe epitaxial films at room temperature after irradiation with ionizing radiation makes it possible to create ionizing radiation detectors based on them.

Conclusions. VAC and photoconductivity, as well as effect of γ - ray irradiation on these properties for epitaxially grown thin films of Cd_{1-x}Mn_xTe (x=0.07) compound has been studied. Results of current investigation reveal that, epitaxial films of Cd_{1-x}Mn_xTe (x=0.07) compounds are sensitive to γ -ray irradiation, indicating that these materials can be considered as a good candidate for γ -ray detector applications.

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