# Electron structure and optical properties of $Cd_{1-X}Mn_XTe$ thin films

M. A. Mehrabova, I. R. Nuriyev, H. S. Orujov

***Abstract*—** Band structure and DOS have been calculated for $Cd_{1-x}Mn_xTe$ (x=0.0625, x=0.0312, x=0.0156) semimagnetic semiconductors by ab-initio method using the Atomistix Toolkit program. It has been determined that with an increase in Mn quantity, the energy gap increases and lattice parameter decreases. The calculated magnetic moment for Mn atom is found to be equal to $5\mu_B$. It was revealed that the antiferromagnetic phase is stable in $Cd_{1-x}Mn_xTe$. The theoretical results are in good agreement with the experimental data obtained by us. It has been synthesized $Cd_{1-x}Mn_xTe$ solid solutions (x=0.01, 0.03, 0.05). Thin films of the given solid solutions were obtained on the glass and mica substrates. Lattice parameters and compositions of the obtained samples were defined with X-ray diffraction method. It has been defined the optimum conditions for obtaining of $Cd_{1-x}Mn_xTe$ (x=0.01, 0.03, 0.05) solid solutions' thin films on the glass and mica substrates. With increasing of substrate temperature, films obtained on the glass substrates are crystallized, and the films obtained on the mica substrates became more perfect. The homogeneity of the composition of synthesized solid solutions have been established by EPR method.

***Keywords*—** ab-initio method, Atomistix Toolkit program, band structure, DOS, EPR, semimagnetic semiconductor, molecular beam condensation, X-ray diffraction method, UVS.

## I. Introduction

Recently, there has been an increasing interest to study wide bandgap II–VI semiconductors such as CdTe and $Cd_{1-x}Mn_xTe$. Among several reasons for this interest, the most prominent is the requirement of these materials for their applications in optoelectronic devices, mobile communication, and cellular automotive. Revolutionary display technology breakthroughs are achieved by them in overhead illumination, in picture transmission and in visualization techniques along with the short wavelength emitting LD and LEDs. In addition, exploitation of these wide-gap semiconductors holds promise for revolutionary improvements in the cost, size, weight, and performance of a broad range of military and commercial microelectronic and optoelectronic systems [1,2].

$Cd_{1-x}Mn_xTe$ is a well-studied semimagnetic semiconductors (SMS) used in Faraday rotators, optical isolators, solar cells, lasers, and infrared detectors. Recently, it was proposed as potential material for gamma-ray and x-ray detectors. These materials being of radiation resistance makes their use perspective in space and space telescopes. Recent years, the interest has increased for reducing the value of solar modules in cosmic applications. The solar cells made on base of CdTe thin films have potential advantages for its low value, high production capacity, high efficiency, its resistance to ionizing radiation and operating stability [3-5].

To study use potentials of these materials in above mentioned fields and improve their physical parameters it should be studied their physical properties and improve their obtain technology. For solving such problems it is necessary a comprehensive approach, i.e. simultaneously theoretical and experimental researches are demanded.

It should be mentioned that theoretical researches, applied in modern material science, are not limited only with explanation of experimental results, but also can forecast them. Today by means of thorough theoretical calculations it can be defined a number of physical properties of the material, having information only about its composition and structure. On one hand it is related with the development of possibilities of computer engineering and computer aids, and on the other hand with rapid improvement of theoretical research methods.

Our aim is the study of obtain technology of $Cd_{1-x}Mn_xTe$ thin films, theoretically and experimentally definition of optimal value of physical parameters and their improvement for effective work of devices on their base.

## II. Electronic structure and DOS

In this section band structure, DOS and PDOS have been calculated for CdTe and $Cd_{1-x}Mn_xTe$. We carried out first principles calculations in the framework of density functional theory (DFT) using Atomistix ToolKit (ATK) programme [6]. It has been carried out the optimization of crystal structure and atom relaxation. It should be noticed, that in the article Te atoms are given in brown, Cd atoms in grey and Mn atoms in violet color in the figures.

It has been given the results of calculations of energy band structure (BS) and density of states (DOS) of CdTe

This work was supported in part by the Science Development Foundation under the President of the Republic of Azerbaijan Grant № EIF- 2012-2(6)-39/06/1-M-25.

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within the spin-polarized density-functional theory (DFT) and the Local Spin Density Approximation (LSDA) on double zeta double polarized (DZDP) basis with regard to Hubbard-U correction (Fig.1, Fig.2, Fig.3). In the result of DOS spectrum analysis (Fig.3) it has been determined that upper levels of valence band located in the range of [-5; 0] eV, mainly comes from p-states of Te atoms, and lower zones of conductivity - from s-states of Cd atoms. Considering that contribution of s-states of Cd atoms to forming of fundamental gap is insignificant, we neglected this correction in further calculations. For obtaining the experimental value $E_g$ it has been taken the value of Hubbard-U parameter for 5p-states of Te, which was 3.7 eV. The calculated band gap width which is 1.53 eV, corresponds to experimental values.

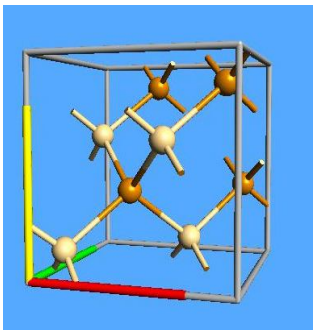


Fig.1 Bulk configuration of CdTe

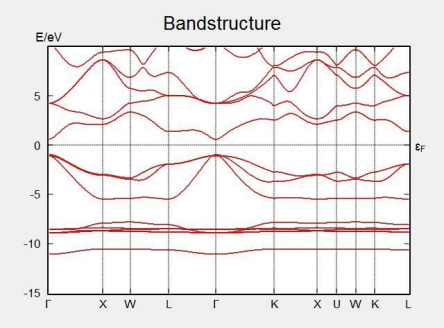



Fig.2. Energy band structure of CdTe

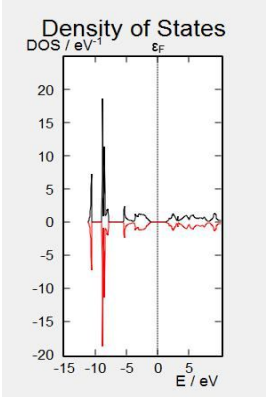
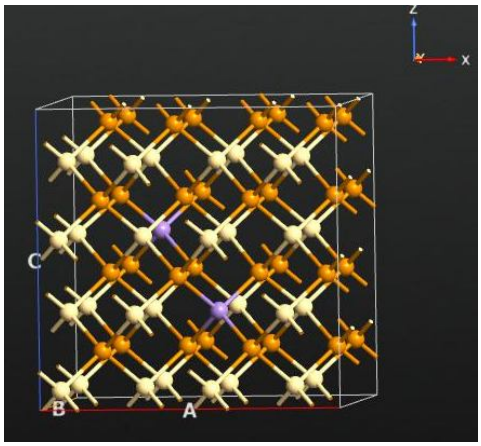



Fig.3. DOS spectrum of CdTe

We have examined spin-polarized electronic band structure, DOS as well as magnetism of $Cd_{1-x}Mn_xTe$ SMS in antiferromagnetic phase for the systems x=0.0625 (64 atoms), x=0.0312 (128 atoms) and x=0.0156 (256 atoms). In our previous works we have investigated these parameters for other systems [7-12].

We modelled the bulk configuration (Fig.4) and have calculated band structure (Fig.5), DOS (Fig.6) for $Cd_{1-x}Mn_xTe$.

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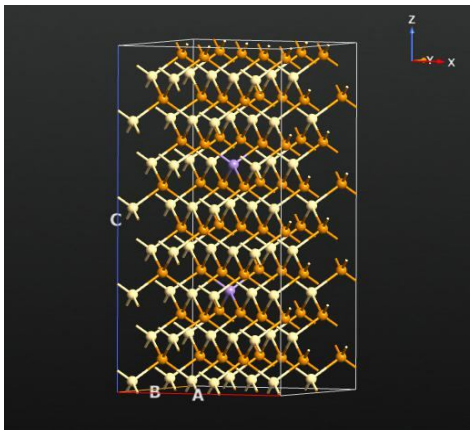


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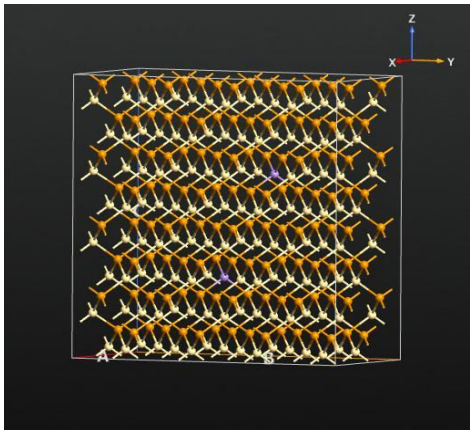
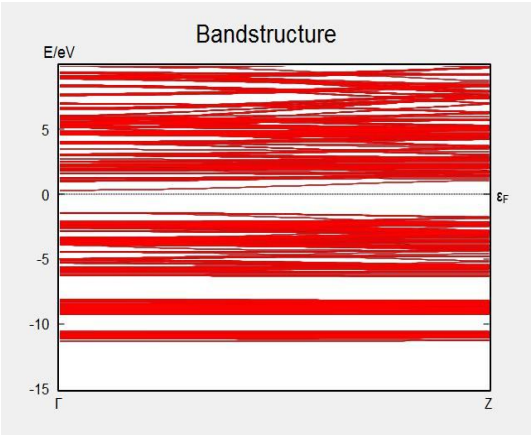



c)

Fig.4 Bulk configuration of $Cd_{1-x}Mn_xTe$:
a) x=0.0625, b) x=0.0312, c) x=0.0156

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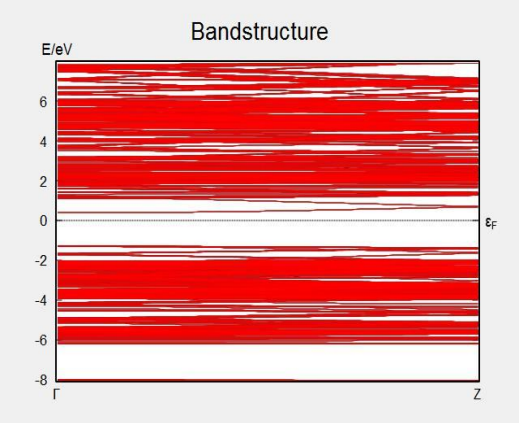


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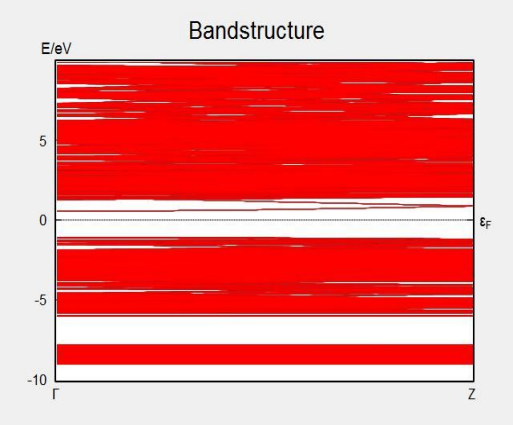



c)

Fig.5. Energy band structure of $Cd_{1-x}Mn_xTe$: a) x=0.0625, b) x=0.0312, c) x=0.0156

The calculations for $Cd_{1-x}Mn_xTe$ are performed within the spin- polarized DFT and the LSDA+U on double zeta double polarized (DZDP) basis with regard to Hubbard-U correction. The value $U_{Mn}$ = 3.59 eV for 3d states of Mn is the literature data, and the value $U_{Te}$ = 3.7 eV for 5p states of Te atoms was taken as in CdTe. We have determined that the energy gap for $Cd_{1-x}Mn_xTe$ with x=0.0625, x=0.0312 and x=0.0156 is correspondently equal to $E_g$=1.7319 eV, $E_g$=1.682 eV, $E_g$=1.652 eV.

It has been determined that lattice parameter decreases insignificantly and the energy gap increases with an increase in Mn quantity in $Cd_{1-x}Mn_xTe$. Value of Mn concentration for large compositions x=0.25is not subject to the general law, so in our studies, we chose to work with the composition $0.01 \leq x \leq 0.07$ (Fig.6).

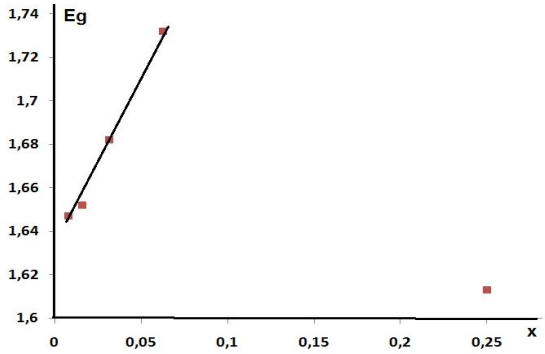
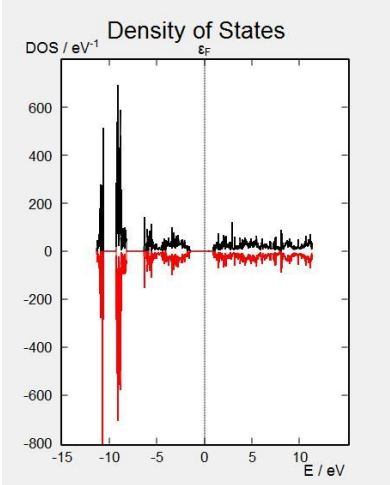



Fig.6 Dependence band gap $E_g$ on Mn concentration for $Cd_{1-x}Mn_xTe$

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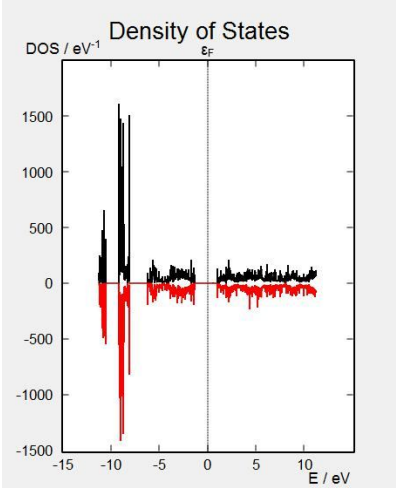


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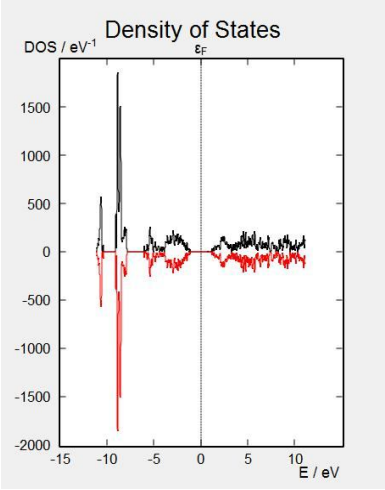
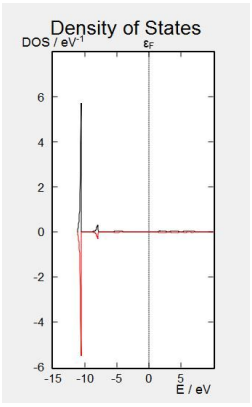


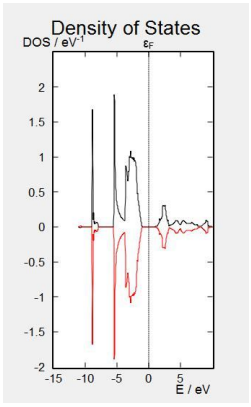

c)

Fig.7 DOS spectrum of $Cd_{1-x}Mn_xTe$:
a) x=0.0625, b) x=0.0312, c) x=0.0156

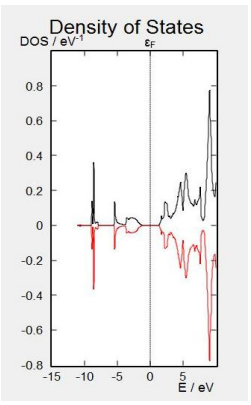
The DOS curves bring out two characteristic features in the valence band: a core-like peak, at ~10 eV below the valence band maximum, originating mainly from Te s states, and structure at the top of the valence band formed mainly from Te p states and Mn d states form the sharp peak at ~6 eV below the valence band maximum with weak participation of Te d states (Fig.8).



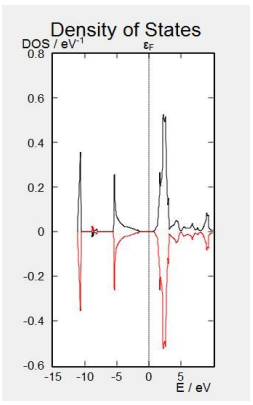

Te s satates




Te p satates

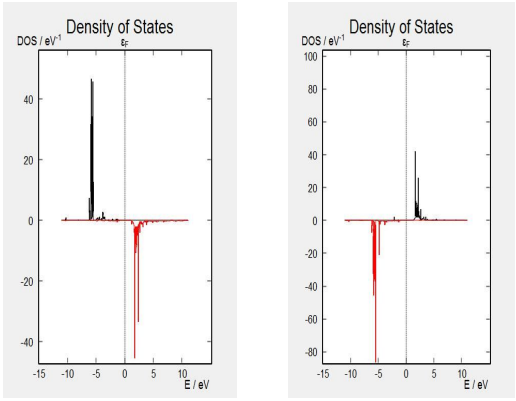



Te s satates




Te p satates

The structure at the bottom of the conductivity band formed from Cd s states and Te p states and peak at ~2eV above the conductivity band minimum, originating mainly from Mn d states.




Mn d satates

Fig.8 PDOS spectrum of $Cd_{1-x}Mn_xTe$

Because of the coupling of host *s* and *p* states with localized Mn 3*d* states, Mn-doped II-VI DMS exhibit interesting properties that combine semiconductor electronics with magnetism. Most of the Mn DMS, such as $Cd_{1-x}Mn_xTe$, shows antiferromagnetic behavior caused by the super exchange interaction, where occupied Mn *d* states couple with the neighboring unoccupied Mn *d* states, thus lowering the total energy. We have considered super cells with 2 Mn atoms having opposite spins. The calculated magnetic moment for Mn atoms is found to be equal to $5\mu_B$ ($\mu_B$ is the Bohr magneton) for $Cd_{1-x}Mn_xTe$.

## III. Obtain Technology of Thin Films

$Cd_{1-x}Mn_xTe$ solid solutions (x=0.01, 0.03, 0.05) have been synthesized and lattice parameters and compositions of the obtained samples defined with X-ray diffraction method (X-ray diffractometer Broker, Germany D8 ADVANGE). These researches showed that the synthesized samples of $Cd_{1-x}Mn_xTe$ (x=0.01, 0.03, 0.05) are crystallized on the base of cubic lattice of CdTe and the lattice parameter decreases insignificantly with an increase in Mn quantity in synthesized samples (Fig.9).

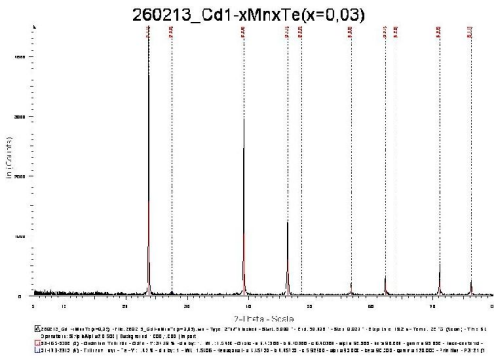



Fig.9 X-Ray diffraction pattern of the synthesized $Cd_{1-x}Mn_xTe$ solid solution, x=0.03

Thin films of the given solid solutions were obtained at standard vacuum installation УВН-71П3 with molecular beam condensation method. It is known that one of the key conditions in obtain technology of films with perfect structure with the mentioned parameters is the use of synthesized materials with the established specific composition as an evaporation source. In the work as an evaporation source it has been used synthesized solid solutions with corresponding composition. The evaporation process of the synthesized samples occurred from graphite Knudsen cells.

Materials from glass and mica monocrystals served as a substrate. The films were obtained on substrates at temperature (T=300 K, 400 K)

Crystalline structures of the obtained films were also studied with X-ray diffraction method. It has been established that at T=300 K the films obtained on substrates of glass have amorphous structure, and the films obtained on monocrystalline mica substrates have the polycrystalline structure (Fig.10).

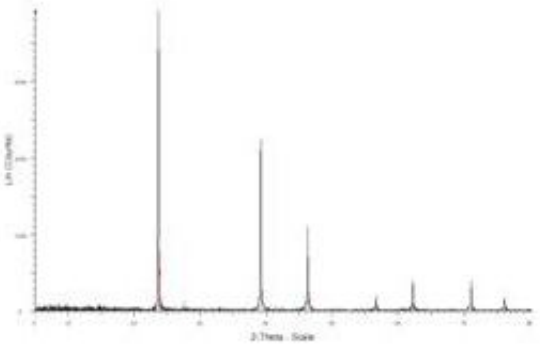


Fig.10 X-Ray diffraction pattern of $Cd_{1-x}Mn_xTe$, x=0.03 thin films on substrates of monocrystalline mica at T=300 K

It has been defined that with increase in substrate temperature (T=400K), there occurs crystallization of films, obtained on substrates of glass (films become polycrystalline, (Fig.11), but films on mica substrates become more perfect.

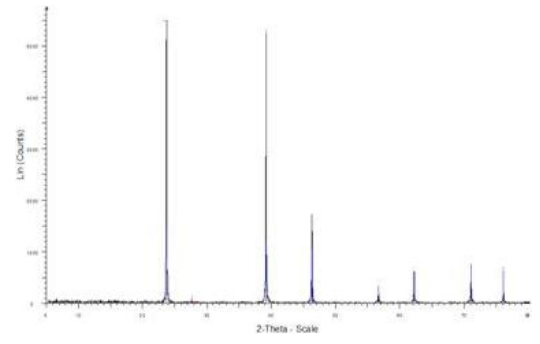


Fig.11 X-Ray diffraction pattern of $Cd_{1-x}Mn_xTe$ (x=0.03) thin films on glass substrates at T=400K

Hence, it has been established that using synthesized solid solution samples of $Cd_{1-x}Mn_xTe$ of different composition as a source of evaporation, it is possible to obtain $Cd_{1-x}Mn_xTe$ thin films of corresponding compositions. With an increase in substrate temperature, there occurs crystallization of films obtained on substrates of glass, and the films grown on mica substrates become more perfect.

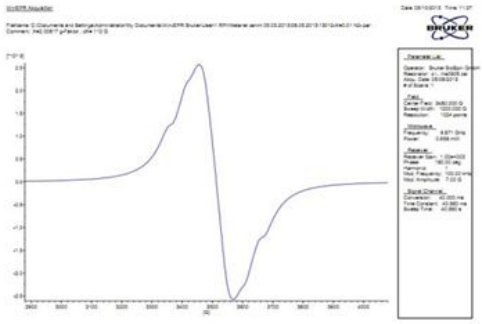
Thus, with increasing of substrate temperature, films obtained on the glass substrate crystallized, and the films grown on mica substrates become more perfect.

## IV. EPR ANALYSIS

It has been studied the homogeneity on the composition of the synthesized solid solutions of $Cd_{1-x}Mn_xTe$ (x=0.01, 0.03, 0.05) by EPR method. EPR spectra have been taken at room temperature in EPR spectrometer of the company "Bruker" EMX/lus (in the range - x, λ=3.2 sm-1). The parameters of EPR spectra obtained from three compositions are as following:

1) x=0.01, g1=2.0082, ΔH=112 Gs,
2) x=0.03, g2=2.0082, ΔH=93 Gs,
3) x=0.05, g3=2.0083, ΔH=101Gs.

The spectra consist of simple symmetrical singlet lines. Complementary spectra are not observed at little and medium values of magnetic field. It testifies that $Mn^{2+}$ ions have uniformly penetrated into matrix of compound. EPR spectra emerge from paramagnetic centre with a single chemical nature. The equal values of g-factor (g=2.0082) for three compositions asserts this fact. The main reason of different ΔH- values of parameters for these compositions can be explained in the following way. With an increase in the quantity of $Mn^{2+}$ ions the distance between ions decreases and unpaired electron shells overlap each other. Therefore the spectrum width changes depending on the quantity of $Mn^{2+}$ ions. It should be mentioned that with a decrease in the quantity of $Mn^{2+}$ ions the distance between ions increases, and they approach isolated state, in the consequence of which the magnetic interaction with ions decreases. As spin moment in $Mn^{2+}$ ion atom is I=5/2, then in isolated ion it should be observed the quantity n=2I+1=6 of hyperfine structure line. For composition x=0.01 it is observed a part of these lines. The constant of hyperfine structure is A≈64 Gs (Fig.12).



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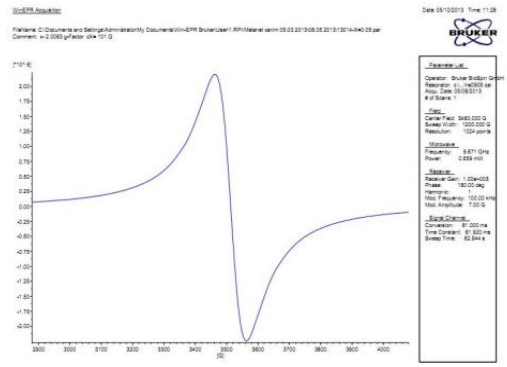

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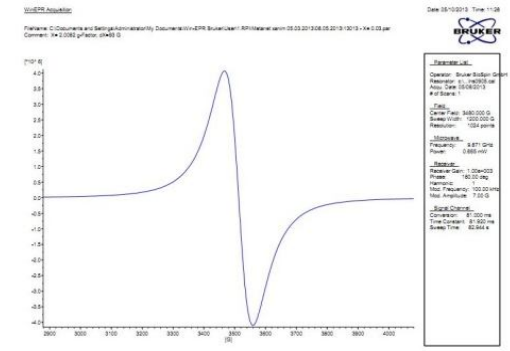


c)

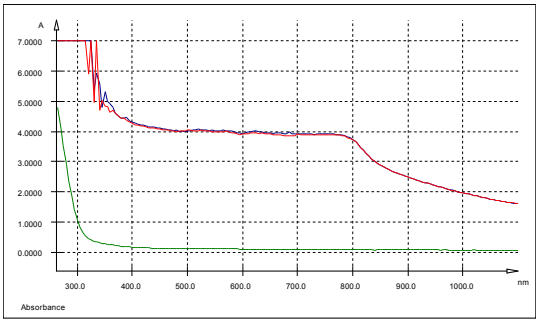
Fig.12 EPR spectra of solid solutions of $Cd_{1-x}Mn_xTe$: a) x=0.01, б) x=0.03, c) x=0.05

Thus, it has been established that $Mn^{2+}$ ions penetrated uniformly into the lattice of the synthesized solid solution.

## V. Optical properties

For defining the band gap width it has been studied the optical properties of the given solid solutions. The spectra of absorption and transmission of $Cd_{1-x}Mn_xTe$ thin films, obtained at room temperature on substrates of mica and glass have been studied. The spectra have been registered in UV-Visible SPECORD 250 PLUS - 223G1020 spectrometer at wave length λ=300-1000nm. Mn concentration varied at the range of x=0.01-0.05.

The absorption and transmission spectra of $Cd_{1-x}Mn_xTe$ thin films on the mica and glass substrates have been depicted in Fig.13 and Fig.14 respectively (Spectra of mica-air and glass-air have also been given).

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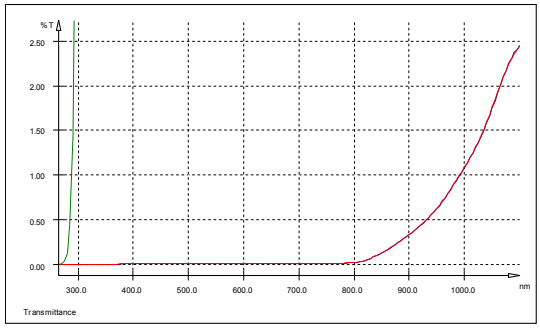
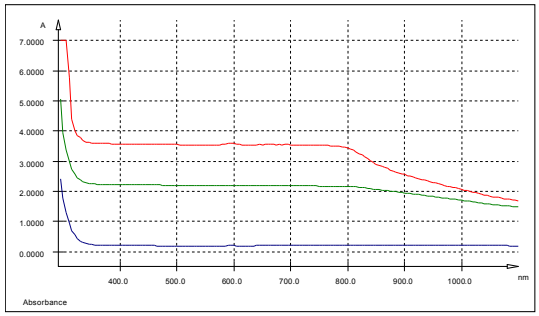



b)

Fig. 13 UV-Visible spectra: spectra of mica-air are depicted with green color (mica relatively to air), mica + $Cd_{1-x}Mn_xTe$ (air) with red, mica+$Cd_{1-x}Mn_xTe$ (mica) with blue a) absorption, b) transmission

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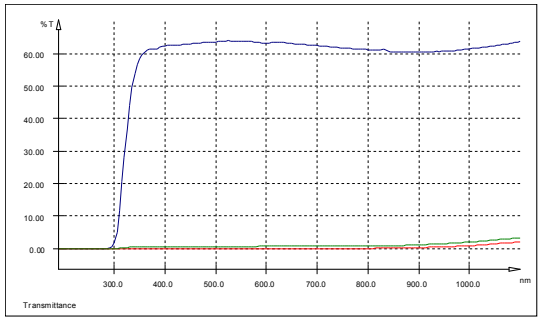


b)

Fig. 14 UV-Visible spectra: glass-air spectra are given with blue color (glass relatively to air), glass + $Cd_{1-x}Mn_xTe$ (air) with green, glass + $Cd_{1-x}Mn_xTe$ (glass) with red a) absorption, b) transmission

On the base of comparative analysis of optical spectra of absorption and transmission the following results have been obtained:

1. It has been revealed that change in Mn concentration (from 0.01 to 0.05) in the studied films leads to a change in band gap width $E_g$ from 1.53 eV to 1.59 eV (Δλ=30 nm). Depending on the type of substrates the absorption range for mica-air 784 nm, (Fig.13), and for glass-air was 833.5 nm, Δλ=49.5 nm (Fig.14). The obtained values of $E_g$, satisfactorily corresponds to the calculated data obtained by us.
2. Films, obtained on the substrates of mica and glass at medium optical range λ=300-800 nm are not transparent and

transmits at λ=800-1000 nm. It points to usage efficiency of these substrates for obtain of thin films in creation of radiation converters, functioning at near IR range.

## VI. Conclusion

It was revealed that SMS $Cd_{1-x}Mn_xTe$ are stable in the antiferromagnetic phase. It was obtained that with an increase in Mn quantity in $Cd_{1-x}Mn_xTe$ the energy gap increases and lattice parameters decrease insignificantly. The calculated total magnetic moment is found to be equal to 5μB for $Cd_{1-x}Mn_xTe$.

It has been defined the optimum conditions for obtaining thin films of $Cd_{1-x}Mn_xTe$ (x=0.01, 0.03, 0.05) solid solutions on substrates of glass and mica. The homogeneity on the composition of the synthesized solid solutions has been established by EPR method. It has been defined the range of transmission for thin films of $Cd_{1-x}Mn_xTe$ (x=0.01, 0.03, 0.05) near IR range. It has been revealed the difference of values of band gap width depending on the composition of the studied solid solutions.

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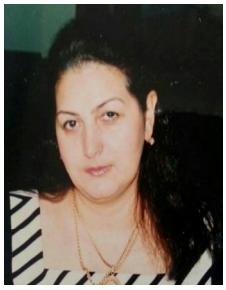


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Associate professor (2011),
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**Scientific activity**: Physics of solid state, theoretical study of physical effects in solid states, semiconductors, radiation physics, radioecology, biophysics.

**M. Mehrabova became a member of the**: V Euroasian Conference on Nuclear Science and İts Application in Turkey (2008), International Conference on Structural Analysis of Advanced Materials in France (2009), Polymer Networks Group20th Conference in Germany (2010), Senior Member of the WSEAS Intern. Conferences in Greece (2010), Barselona, Spain (2011), Prague, Czech.(2011), Catania, Italy (2011), Montreux, Switzerland (2011), Rovaniemi, Finland (2012), Porto, Portugal (2012), plenary speaker in the WSEAS Intern. Conf. in Cambridge, UK, (2013), plenary speaker in the WSEAS Intern. Conf.in Tenerife, Spain (2014).

**Career/Employment:**
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1. International Diploma №085 of "YOUNG SCIENTISTS OF XXI CENTURE", Winner of the International Project of "YOUNG SCIENTISTS OF AZERBAIJAN", 2004
2. Scientific Technology Centre of Ukraine – «Technological Management: commercialization of R & D results in scientific research institutes», 2007
3. Azerbaijan Bank Training Centre – « Technology Transfer Management», 2007, December
4. Civilian Research Development Foundation –"FROM IDEA TO MARKET" A workshop series for technology entrepreneurs, 2008
5. Civilian Research Development Foundation, Science and Technology Entrepreneurship Program, STEP Venture Forum, 2008
6. Participation in the V Eurasian Conference "Nuclear Science and its Application", Turkey, Ankara, 2008, October
7. Participation in the International Conference of Structural Analysis and Advance Materials, France, Tarbes, 2009, September
8. GRBS SERTIFICATE, Intermediate level of English language, 2009, July
9. SRDF STEP Technology Entrepreneurship Program (Workshop), Baku, Azerbaijan, 2010, July 1-2
10. Specialization course on "Information and innovation: the international experience and modern approach" ANAS, Scientific Innovation Center, 2010, December 15-17
11. Participation in the 20-th International Research Project Exibitions , Turkey, Istanbul, 2011, May.

**Consultant and participant of the projects**:
1. BII- Project BII-F-249, 2006
2. STCU project: 4798 - "Biochemicals indicators of venom of Vipera lebetina obtusa (biotic and abiotic factors)" (2008-2010)
3. SRDF Science &Technology Entrepreneurship Program, STEP Venture Forum (2009), Preparation of Aggressive-Resistant Polymer Elastic Materials for Oil and Mechanical Engineering Industries
4. STCU project: 3913 – "Influence of mineral oil, heavy metals and radiation on biodiversity of spiders", (2009-2012)
5. MEF, 20th International research projects exhibition, Turkey, Istanbul, 2011, "Influence of heavy metals and radiation on biodiversity of coccinellidae", head of the project.
6. MEF, 21th International Research Projects Contest, Turkey, Istanbul, 2012, "Obtaining Of Perfect $Pb_{1-x}Mn_xTe(Se,S)$ Nanofilms And Analysis Of Their Use Potentials in Spintronics", head of the project

7. STCU project: 5816 - Project Title: Obtaining of Cd1-xMnxTe nanofilms and study of their potential applications in x- and gamma-ray detection. Project Manager: Mehrabova Matanat Ahmad (2013-2014)
8. SDF (Azerbaijan)- Technology development of obtaining $Cd_{1-x}Mn_xTe$ epitaxial thin films and study of their use potentials in devices with different purpose (2013-2015)
9. SRDF, 2013, "Creation of new generation solar cells on the base of CdMnTe thin films" Project manager M. A. Mehrabova
10. SOKAR - State Oil Company of Azerbaijan Republic-2013, "Creation of high-efficiency thin film solar elements on the base of CdTe/CdMnTe"

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Ph.D in physics-mathematics (1971), Azerbaijan
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**Scientific activity**: Specialized on IR photoelectronics, crystal growth, physics of solid state, semiconductors, nanomaterials.

**H. R. Nuriyev became a member of the:** International Scientific and Engineering Conference on Photoelectronics and Night Vision Devices in Moscow (1992-2010), 9th International Symposium "High-purity Metallic and Semiconducting Materials" in Kharkov, Ukraine (2003), National Conferences of growth of crystals in Moscow (2004-2008), 3rd International Conference on Technical and Physical Problems in Power Engineering in Ankara, Turkey (TPE-2006), Russian Symposium on Scanning Microscopy, Probe Microscopy and analytic research Technique, Chernogolovka settlement in Moscow (2001-2011), 17th International Conference on Advanced Laser Technologies in Antalya, Turkey (2009), National Conference on application X-ray, Synchrotron radiations, Neutrons and Electrons for research of materials in Moscow (2003-2009), International Conference "Micro- and nanoelectronics 2005" in Moscow, Second International Conference on Technical and Physical Problems in Power Engineering (TPE-2004)in Tabriz, Iran (2004), Photovoltaic and Photoactive Materials-Properties, Technology and Applications, NATO Advanced Study Institute in Sozopol, Bulgaria (2001), Senior Member of the WSEAS –Intern. Conferences in Prague, Czech.(2011), Catania, Italy (2011), Montreux, Switzerland (2011), Rovaniemi, Finland (2012), Porto, Portugal (2012), Cambridge, UK, (2013), Tenerife, Spain (2014).

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**Consultant and participant of the projects**:
1. STCU Project № 3237, 01.07.2006-01.07.2008, "Make photoreceivers on the base of the epitaxial films of GaSe, GaTe, InSe lamellar semiconductors and monocrystals".
2. INTAS Project 01-0190, 01.01.2002 - 01.01.2004, "$Pb_{1-x}Mn_xTe$ epitaxial films and photosensitive homo- and heterostructures on their base".
3. ANAS project, 01.01.2010-31.12.2010, "Complex researches in the direction of making renewable energy sources with high efficiency on the base of nanostructure materials".
4. STCU project: 5816 - Project Title: Obtaining of Cd1-xMnxTe nanofilms and study of their potential applications in x- and gamma-ray detection. Project Manager: Mehrabova Matanat Ahmad (2013-2014)
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**This work was supported by the Science Development Foundation under the President of the Republic of Azerbaijan Grant № EİF- 2012-2(6)-39/06/1-M-25**