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M.K.Jangid
Department of Physics, Vivekananda Global University

Sharma, Sumit
Department of Mechanical Engineering, Poornima College of Engineering

Ved Prakash Meena
Department of Physics, Vivekananda Global University

Ved Prakash Arya
Department of Education in Science and Mathematics, Regional Institute of Education, NCERT

他

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Impact of Annealing and Hydrogenation on the Optical Characteristics of ZnSe/Mn and ZnSe/Co DMS Thin Films Prepared via Thermal Evaporation Method

M.K.Jangid[1*], Sumit Sharma[2], Ved Prakash Meena[1], Ved Prakash Arya[3], S.S. Sharma[4*]

[1]Department of Physics, Vivekananda Global University, Jaipur-303012, India

[2]Department of Mechanical Engineering, Poornima College of Engineering, Jaipur-302022, India

[3]Department of Education in Science and Mathematics, Regional Institute of Education, NCERT, Ajmer- 305004, India

[4]Department of Physics (H & S), Govt. Mahila Engineering College, Ajmer-305002, India

*Author to whom correspondence should be addressed:
E-mail: mahesh.jangid@vgu.ac.in(MKJ), shyam@gweca.ac.in (SSS)

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Abstract: This article reports that the annealing and hydrogenation effect on the transmission, reflection and bandgap of ZnSe/Mn and ZnSe/Co diluted magnetic semiconductors (DMS). The diluted magnetic semiconductor multilayers thin films of ZnSe/Mn and ZnSe/Co were physically deposited by thermal evaporation technique at 10^{-5} torr vacuum pressure onto glass substrates. To achieve the inter-diffusion and homogeneous structures, the prepared samples were annealed at 473K for one hour in vacuum. Hydrogenation of annealed samples was done at pressure 20 and 30 psi H_2 to observe the effect of hydrogenation on optical properties of thin films. The transmission and reflection of the samples were recorded using UV-Vis. spectrophotometer in the visible range. It was observed that the transmittance as well as energy bandgap of the samples varies with annealing temperature and hydrogen pressure. The opposite impact of annealing and hydrogenation on optical properties of ZnSe/Mn and ZnSe/Co thin films were observed that is Mn and Co interlayer with ZnSe reverse the effect of hydrogenation. The reverse effect of hydrogenation creates new possibilities to use ZnSe/Mn and ZnSe/Co diluted magnetic semiconductors in the field of photovoltaics, energy storage, spintronics and sensors.

Keywords: Thin film; DMS; Annealing; Hydrogenation; Transmission; Reflection; Bandgap

1. Introduction

II-VI group DMS thin film materials have attracted a lot of attention in recent years in both fundamental studies and technical worldwide applications due to their unique size dependent properties that are different from their corresponding bulk characteristics. In addition, a significant decrease in the cost of manufacturing of these DMS thin films as compared to single crystals has been reported. The mostly used alternatives for present semiconductor technology are wide band gap semiconductors. A new generation of photodiodes based on wide bandgap semiconductors is the result of many efforts done in the present decade. A novel family of semiconductor materials called DMSs has been created by alloying II-VI group compounds with the transition metal elements[1)]. Because of the unique magnetic and optical properties revealed by this novel family of DMSs, it is possible to manufacture new functional semiconducting devices for various applications[2)]. A potentially useful non-toxic II-VI semiconducting material, zinc slenide has admirable properties including (i) broad direct energy bandgap (2.7eV)[3)],. (ii) good exciting binding energy (22MeV[4)]), (ii) high luminescence efficiency,. (iv) high refractive index and (v) high transparency in the visible range[5)]. Zinc selenide has good quantum efficiency and more stability[6),7)]. ZnSe-based nano-structured materials represent promising substances for optoelectronic as well as photovoltaic devices[8),9)] like blue lasers, thin semiconductors, photo detectors, and solar cells due to their characteristics[10),11)]. Numerous optical studies have also been conducted on various wide bandgap materials and found the bandgap in the range from 3.00 eV to 3.6 eV[12),13)]. The tendency of hydrogen to form compounds with the majority of crystal defects and impurities makes it a significant contaminant in many semiconductors.

Hydrogen has attracted interest from both the basic and technology domains due to its presence throughout crystal formation and processing[14),15)]. The electronic performance of H_2 in an array of materials is adequately described by a "universal alignment" model[16),17)]. Several studies have been reported into how hydrogen affects properties of semiconductor thin films[18),19)]. Atomic impact of hydrogen on the magnetic characteristics of (Ga,Mn)N and it may have a significant impact if it passivates the deep Mn acceptors[20),21)]. Recently novel type of semiconductor magneto electronic spintronic devices, such as spin LEDs or transistors, are emerging as a result of the advantages of employing electronic spins for these purposes[22),23)]. Diluted magnetic semiconductors (DMS) combine semiconductor versatility with magnetic ordering[24)]. Future spintronics applications show considerable potential for the diluted magnetic semiconductors. Their magnetic and semiconducting characteristics open up intriguing prospects for cutting-edge technologies. Only (In-Mn)As is DMS to display both electric field control magnetism[25)] and commercial production cost of the solar cell could be optimized ferromagnetism[26),27)]. DMSs are alloy semiconducting materials that contain a significant amount of magnetic ions (Mn^{+2}, Cr^{+2}, Fe^{+2}, and Co^{+2}). It is due to the ease with group II cations can be replaced by such +2 magnetic ions in the host II-VI crystals. Magneto-optic characteristics of II-VI group supported DMSs have been well researched and optically isolators have been made use due to their large Faraday effect[28)]. Metals or transition metals (Al, Co, Sr, Mn etc.) doping with ZnSe has improved the characteristics of these materials[29)], The doped ZnSe thin films have been effectively used in a variety of applications[30),31)]. Sr-doped ZnSe and Ni-doped ZnSe synthesized thin films have been studied for optical and vibration properties and reported the identification of three new vibrational bands by FTIR and Raman analysis[32),33)]. The optical studies indicate the doped samples to have a red shifted absorption spectrum and a decreased bandgap about 3.3eV. It has also been investigated how doping of Mg transition metal affected the characteristics of ZnSe film and reported that the energy bandgap of film was 3.48eV, the particles was 11 nm in dimension, and the Mg was entirely bonded in the ZnSe lattice[34),35)]. Due to the fact that most II-VI based dilute magnetic semiconductors typically exhibit anti ferromagnetic or spin glass ordering, the II-VI semiconductor doped with a magnetic element has also been researched but has only found a limited number of uses. Exploring its electron charge and spin properties have enabled the tremendous advancements being currently achieved in the realms of electronics and information technologies[36)].The present study reports how hydrogen pressure and annealing temperature impacts transmission, reflection and bandgap of ZnSe/Mn and ZnSe/Co multilayered thin films in the visible range which were prepared using the vacuum thermal evaporation method under high (10^{-5}torr) vacuum pressure.

2. Experimental

The materials (ZnSe, Mn& Co) having 99.99% purity were purchased (Alfa Aesar, Jonson Matthay Company, U.S.A) for the current study. The Vacuum coating unit (Hind High Vacuum Co. (P) ltd. Bangalore-58) was used to deposit thin films. To prepare the samples, above compounds were put into the two different boats inside the vacuum chamber. The distance between boats and substrate was kept at 15 cm in each case. The evaporation of materials was carried out in a vacuum by gradually raising the electrodes current to heat the boat. Using the stacked layer approach, multilayered DMS thin films were produced. A Hindhivac thickness monitor model DTM-101was used to evaluate the thickness of prepared multilayers. The measured thickness were recorded 500nm for both ZnSe/Mn 500 nm and ZnSe/Co i.e. through co-doping ZnSe/Mn(500nm) and ZnSe/Co(500nm) DMS thin films were developed onto the glass substrates. Before deposition the substrates, the vacuum chamber were cleaned properly to minimize oxidation. All samples were vacuum packaged within 30 minutes of being removed from the unit. The prepared thin films were vacuum annealed at 473K in vacuum (10^{-5}torr) for one hour to establish the inter-diffusion and homogeneous structure of the film. Hydrogenation of annealed samples was done at pressure 20 and 30 psi H_2 to observe the effect of hydrogen on optical properties. The transmission and reflection of the samples were recorded using UV-Vis. spectrophotometer (UV-1800 Shimadzu spectrophotometer).

3. Results and Discussion

3.1 Annealing effect on ZnSe/Mn and ZnSe/Co DMS thin films

Annealing is a widely used technique to modify the properties of thin films. In this study, we investigated the annealing effect on the transmission spectra of ZnSe/Mn and ZnSe/Co diluted magnetic semiconductors thin films. Figure 1 shows the transmission spectra of ZnSe/Mn before and after annealing. It was noted that the transmission decreases after annealing of samples compare to as-deposited samples. After annealing transmission decreases because of annealing can introduce defects or impurities in the thin film, creating absorbing centers that can contribute to the absorption of light. These defects may arise due to the diffusion of atoms or molecules within the film during annealing. Figure 2 shows the transmission spectra of ZnSe/Co thin films. According to this graph, transmission increases after annealing as compared with the as-deposited samples. It reveals that annealing enhance the crystallinity of thin films by reducing defects and dislocations. A more ordered crystal structure can result in fewer scattering events and less absorption, leading to increased transmission. The findings revealed that annealing caused a considerable variation in the transmission of DMS films. This rise in transmission is observed because of elimination of defects and the improvement in crystal quality that occurs as a result of annealing[37)].

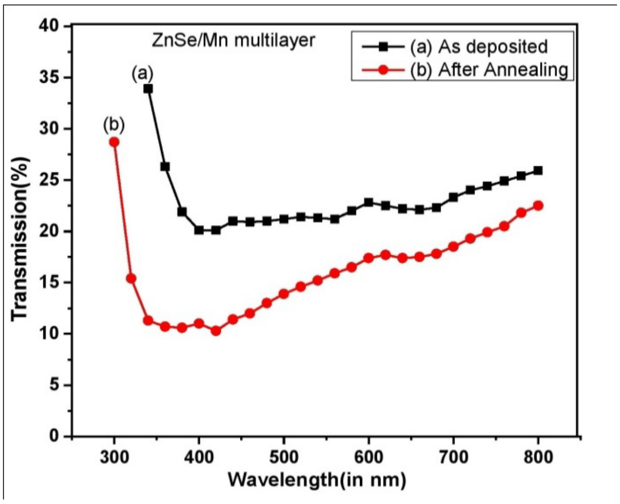

Fig. 1: Transmission spectra of as-deposited and annealed ZnSe/Mn DMS thin films.

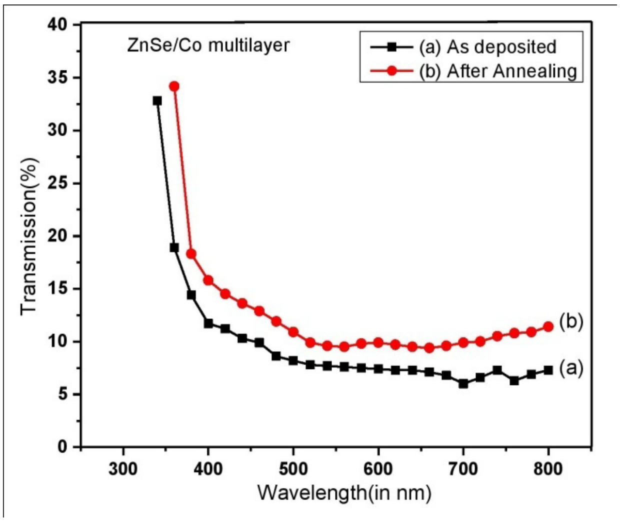

Fig. 2: Transmission spectra of as-deposited and annealed ZnSe/Co DMS thin films.

Using the Tauc relation given below, the energy bandgap of samples were estimated[38)].

$$(\alpha h\nu)^{1/n} = A(h\nu - E_g)$$

Taking n=0.5 (for direct bandgap) plotted the curve between $(\alpha h\nu)^2$ and photon energy (hν) to find out the energy band gap (Eg). Figure 3 and Figure 4 show the Tauc spectra of ZnSe/Mn and ZnSe/Co DMS thin films, respectively. The band gap of annealed ZnSe/Mn thin film is found to change and increases after annealing (2.34eV to3.13eV) (Table 1). It may be due to tailoring of band gap by mixing of Mn in ZnSe interface which creates a new phase that is responsible for changing the bandgap. Ikhioya et al reported a variation in the band gap of the synthesized Co/ZnSe films[39)]. The band gap found to reduce in case the of annealed ZnSe/Co films (3.19eV to 2.87eV) (Table 1). It is due to the improved crystallinity of the thin film owing to annealing. This can lead to a more uniform and ordered structure, influencing the electronic properties and potentially causing a decrease in the bandgap.

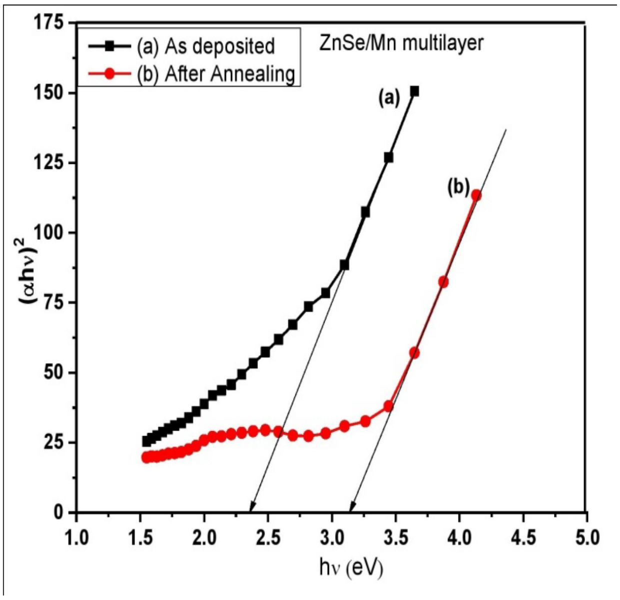

Fig. 3: Tauc spectra of as-deposited and annealed ZnSe/Mn DMS thin films.

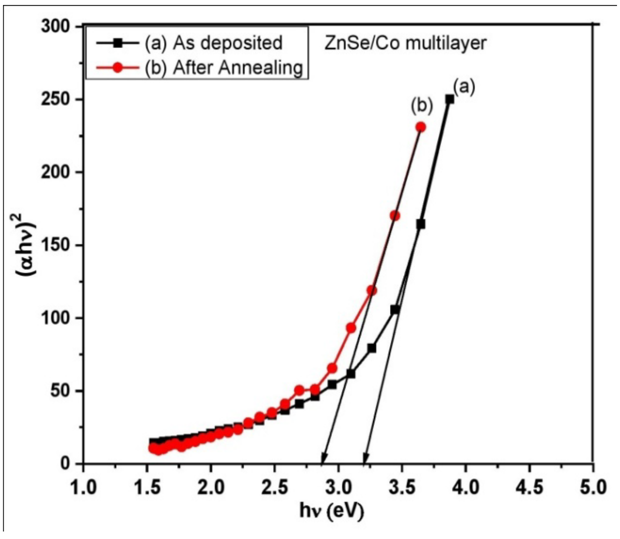

Fig. 4: Tauc spectra of as-deposited and annealed ZnSe/Co DMS thin films.

Table 1. Variation in energy bandgap after annealing of ZnSe/Mn and ZnSe/Co DMS thin films.

Sample Type	Optical Bandgap (in eV)	
	ZnSe/Mn Multilayer	**ZnSe/Co Multilayer**
As-deposited	2.34	3.19
After Annealing at 473K	3.13	2.87

Annealing of ZnSe/Mn and ZnSe/Co DMS thin films can significantly affect their reflectance in the visible range. Several factors, including annealing temperature, annealing time and film thickness influence the reflectance of thin films. Figure 5 illustrates reflectance versus wavelength curve of as-deposited and annealed ZnSe/Mn DMS thin films. The reflectance of annealed ZnSe/Mn DMS thin films is found to increase. This is because of the crystalline quality has improved, and the number of flaws and contaminants in the films has decreased. However, excessive annealing can also cause faults like surface roughness and other flaws to emerge, which can reduce reflectivity. The concentration of flaws and impurities, modifications in the film microstructure, and the creation of secondary phases containing Mn are all factors that contribute to how annealing affects reflectance. The reflectance of the annealed Mn/ZnSe thin film in the visible range has been studied and reported an increase in the reflectance [40),41)].

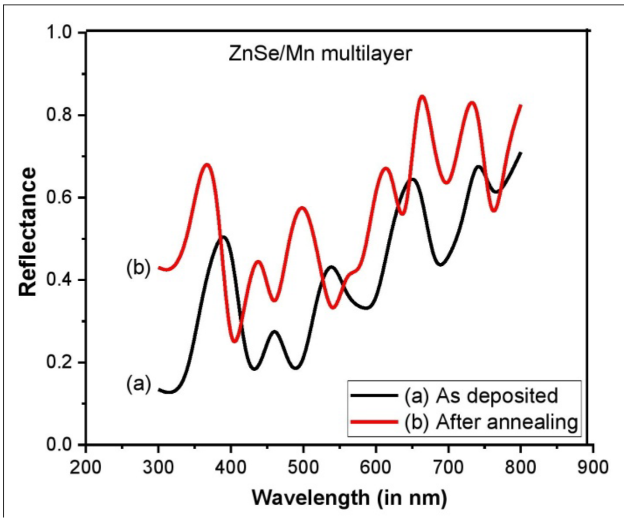

Fig. 5: Reflectance of as-deposited and annealed ZnSe/Mn DMS thin films.

Figure 6 represents the reflectance of the ZnSe/Co DMS thin films against wavelength. The annealing of the thin film has the opposite impact on ZnSe/Co diluted magnetic semiconductors as compared to ZnSe/Mn DMS thin films. A number of variables including temperature, annealing time, Co doping concentration and film thickness, impact how annealing changes the reflectance of ZnSe/Co thin films in the visible range. Annealing may induce the absorbing centers within the thin film, leading to increased absorption of incident light and, consequently, a decrease in reflectance [42),43)].

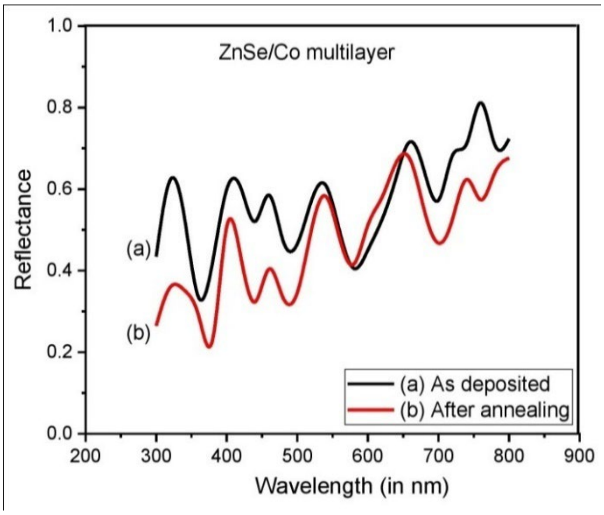

Fig. 6: Reflectance of as-deposited and annealed ZnSe/Co DMS thin films.

3.2 Hydrogenation effect on ZnSe/Mn and ZnSe/Co DMS thin films

Figure 7 represents the variation in transmittance with wavelength at different hydrogen pressure for annealed ZnSe/Mn DMS thin films. The variation in optical transmittance was measured in the wavelength range 300nm to 800nm for all the samples. A higher transmittance has been observed after hydrogenation of thin films. Hydrogenation may induce the defects within the thin film. These defects can alter the electronic and optical properties of the material, potentially leading to a decrease in absorption and an increase in transmission. Thus, films show a continually increasing transmittance (reduced absorbance) across the visible region, allowing for the use of this material in photo electrochemical cells[44)].

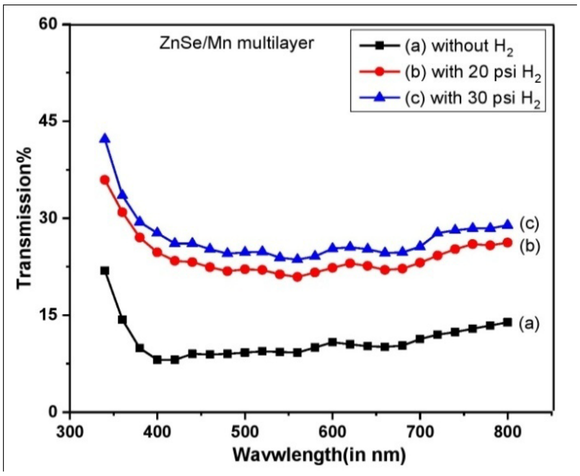

Fig. 7: Effect of hydrogenation on transmission of annealed ZnSe/Mn DMS thin films.

Figure 8 shows variation in transmittance with wavelength at different hydrogen pressure for annealed ZnSe/Co DMS thin films. A lower transmittance was seen after hydrogenation of thin films. Thus, films show a continually decreasing transmittance across the visible region after hydrogenation. The blemish passivation by H_2 absorption at the film interface is the cause of these changes in the transmission spectra[45),46)].

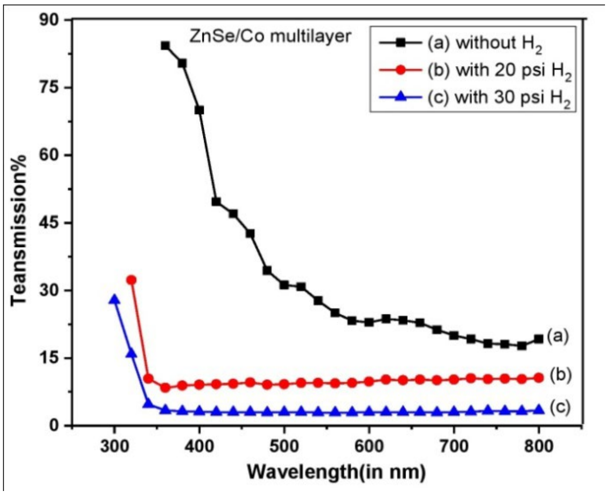

Fig. 8: Effect of hydrogenation on transmission of annealed ZnSe/Co DMS thin films.

Again using the Tauc relation, the energy bandgap of samples were estimated by plotting the $(\alpha h\nu)^2$ versus photon energy (hν) curves. Figure 9 and Figure 10 show the Tauc spectra of annealed ZnSe/Mn and ZnSe/Co DMS thin films, respectively. In each case, a variation in the bandgap with pressure of hydrogen has been recorded. In case of ZnSe/Mn DMS thin films, a decrease in bandgap with hydrogen from 2.43eV to 2.25eV whereas for ZnSe/Co, an increase from 2.09eV to 3.31eV has observed (Table 2). In general, the way that hydrogenation impacts on energy bandgap of a material can vary depending on a number of factors including the specific semiconductor material, the type of defects and the hydrogenation process. Some defects can be passivated, which can cause the band gap increase; yet, other interactions or changes to the electronic structure can cause the band gap to decrease. The variation in the optical bandgap is caused by the transition of the structure from a metallic to a semiconductor phase as the hydrogen pressure increases[47)]. High electrical charge regions, such as semiconductors, contain hydrogen, with the strongest semiconductor being an example. Thin film hydride systems greatly benefit from the ability of hydrogen to absorb electrons from thin films, which aligns with the anionic theory of hydrogen-metal interactions with semiconductors[48),49)].The gathering of hydrogen at interface could be the cause of the changed energy bandgap[50)]. Upon hydrogenation of thin films, variation in optical band gaps were noted; these variations are the result of a band-filling phenomenon called the Burstein-Moss shift. Similar results have also been reported when examining the impact of hydrogen on the electrical and optical characteristics of Al/ZnO thin films[51),52)].

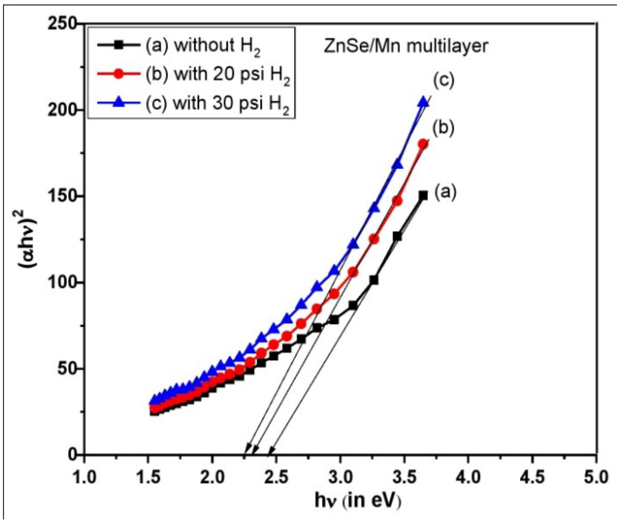

Fig. 9: Effect of hydrogenation on energy bandgap of annealed ZnSe/Mn DMS thin films.

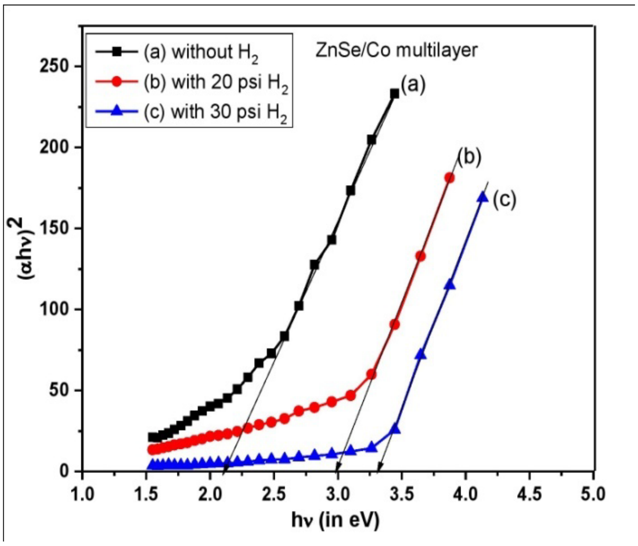

Fig. 10: Effect of hydrogenation on energy bandgap of annealed ZnSe/Co DMS thin films.

Table 2.Variation in energy bandgap with hydrogenation of annealed ZnSe/Mn and ZnSe/Co DMS thin films.

Sample Type	**Optical Bandgap (in eV)**	
	ZnSe/Mn Multilayer	**ZnSe/Co Multilayer**
Without H_2	2.43	2.09
With 20 psi H_2	2.31	2.98
With 30 psi H_2	2.25	3.31

4. Conclusion

Hydrogen is a vital impurity in many semiconductors, due to its ability to interact with the majority of crystal impurities and defects to form complexes. Hydrogen has drawn both basic and technological interest due to its significance in the development of crystals and processing. The ZnSe/Mn and ZnSe/Co diluted magnetic semiconductor multilayers have deposited by the vacuum thermal evaporation technique and confirmed the effects of hydrogen on the film structures as well as the inter-diffusion. The results obtained unequivocally show that hydrogenation can be utilized to change the optical characteristics of films. Although the energy bandgap of ZnSe/Mn was found to decrease with hydrogen pressure (2.43eV to 2.25eV), whereas the energy bandgap of ZnSe/Co was found to increase with hydrogen pressure (2.09eV to 3.31eV). It is due to defects that have been hydrogen passivated in these DMS thin films. It means Mn and Co interlayer with ZnSe reverse the effect of hydrogenation and also tailored the bandgap of the prepared dilute magnetic semiconductors. The reverse effects of hydrogenation on ZnSe/Mn and ZnSe/Co thin films offer opportunities for tailoring their optical and magnetic properties, opening up avenues for applications in various fields, including spintronics, sensors, photovoltaics, and energy storage. The ability to control these properties through hydrogenation provides a versatile approach to optimize the performance of devices based on diluted magnetic semiconductors.

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Nomenclature

DMS	*Diluted Magnetic Semiconductors*
UV-Vis	*Ultraviolet-Visible*
ZnSe	*Zinc selenide*
Mn	*Manganese*
Co	*Cobalt*
H_2	*Hydrogen*

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