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Study of Photoconductivity in Mixed Composite of $MgTiO_3$ and ZnO

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Abstract- The photoconductivity of mixed composite of magnesium titanate ($MgTiO_3$) and zinc oxide ZnO has been reported with respect to the number of parameters. The sample is prepared by heating the homogeneous mixture of $MgTiO_3$ and ZnO in a cylindrical furnace at 600 - 800^0 C for 45 minute. For the measurement purpose, the cell is fabricated in the form of sandwich. In this type of cell the electrodes are in direct contact with the material. The effect of voltage, temperature and field intensity on photoconductivity have been investigated.

***Index Terms* -** ZnO, Photocurrent, Temperature, Sub-linear, Super-linear, Intensity, Voltage

I. Introduction

The photoconductivity study provides us the substantial information about the electronic transition in semiconductor [1, 2]. Photoconductivity of material is considered to be an important tool for providing information, regarding the nature of the photo-excitations. In last few years most of the researchers have shown their great interest in the study of photoconductive properties of inorganic nanoparticles. A number of markers have investigated photo conducting properties of various materials [3]. Recently the optical and electrical characteristics of Al doped zinc oxide thin film prepared by solgel technique, property of gallium doped zinc oxide deposited on glass by spray analysis and applications of inorganic nano crystal in polymer-network have also been studied. It has been observed that zinc oxide (ZnO) exhibits good photoconductivity and high transparency in the visible region. Therefore zinc oxide may be used as transparent electrodes for solar cells [4]. A number of researchers have reported the measurement of photoconductivity in ZnO thin films [5]. It has been observed that in many materials the electrical conductivity is increases by absorption of radiation. If we consider the case of homogeneous material, the density of holes and electrons are uniform throughout the materials.

We know that the rise and decay curves of photocurrent are governed by the trapping states or recombination centers which lie in the forbidden energy zone of a photoconductor. So that using these curves we can understand the nature and distribution of traps and recombination centre. In case of semiconductor materials, the photoconductivity is caused due to photo−generation of electron−hole pairs in the material after absorption of photons which further increases the carrier density, resulting increase in conductivity of material. The response time is also an important tool of photoconducting materials. A good photosensitive material exhibits large change in conductivity and shows fast response. If the trapping centres are destroyed then response time becomes slow. The trapping centers may also increase the decay time if they slowly release the trapped carriers after the removal of excitation source. The number of electron and holes in insulators may be much larger than the corresponding free charge carrier density in the dark. In the steady state, generation rate of electrons and holes must be equal to the rate of recombination of charge carriers and the rate of trapping must be equal to the rate of de-trapping (or re-excitation). To analyze carrier transport problems involving these processes, it is important to set a quantitative criterion to separate trapping and recombination centers. To separate trapping and recombination centers Rose [6] has used demarcation. The demarcation level for electron traps is EDn, may be considered as the level at which a captured electron has an equal probability of being excited into the conduction band and of recombining with a hole from the valence band. Similarly, the demarcation level for hole traps is EDp, may be considered as the level at which a captured hole has an equal probability of being excited into the valence band and of recombining with an electron from the conduction band. But in the case of semiconductor the inverse is often true and the influence of radiation may be understood as a small perturbation on a large dark carrier density. The discussion of photoconductivity is mainly base on two assumptions:

(1) The conductivity is dominated by one of the carriers so that the contribution of the other may be effectively neglected.
(2) During the photoconductivity process, the crystal remains neutral without a build-up of appreciable space charge in the crystal.

There are a number of possible processes for the generation of intrinsic electron–hole pairs, such as singlet exciton–singlet exciton collision ionization and singlet exciton–triplet exiton collision ionization.
Two-photon carrier generation processes are directly related with the generation of two photo-excited states (excitons), which interacts and resulting in a direct transition of an electron from the valence band to the conduction band or to an auto ionization state above Ec [7,8]. The conductivity is mainly dominated by one of the carriers therefore the contribution of the other carrier

may be neglected. A number of investigators have also observed the dependence of light intensity on photoconductivity which is primarily based on the charge neutrality condition with the gap states assumed to be distributed in energy but not necessary exponentially or based on the assumption that each localized gap state has three possible charge values such as neutral, negative, or positive[9,10]. Kao and Schellenberg [11] have generalized the Rose's original expression and explained in two ways:

(1) By extending the contribution of gap states to recombination from those above the mid gap states to any states between electron and hole quasi–Fermi levels.

(2) By including the effect of the asymmetry of gap state distributions and the charge neutrality condition.

This analysis showed that the odd values of the exponent may occur in solids with gap states distributed discretely or continually.
Investigation of photocurrent with respect to number of parameters such as voltage, light intensity, energy of illumination temperature provides us substantial information about the material. The variation of photocurrent with light intensity provides information regarding the charge trapping and recombination process inside the material. The temperature dependence of photocurrent gives substantial information about the energy depth of Fermi-level and the localized defect states at a given temperatures.

II. MATERIALS AND METHODS

For the preparation of the mixed system, high purity Mg, TiO_3 and ZnO are taken in different proportions by weight. The three base material pure magnesium (Mg), pure titanium oxide (TiO_3) and pure zinc oxide (ZnO) are taken in the powder form. These proportions are ground properly in order to get homogeneous mixture. The homogeneous mixture is then fired in a ceramic tube in cylindrical furnace at 600^0C to 800^0C in controlled atmosphere for about 45 minutes. The heated material is then suddenly quenched at room temperature and again ground in order to get microcrystalline form. The sample is preserved in desiccators in order to avoid moisture. For firing process Muffle furnace or a high temperature wire bounded tubular furnace was used. The cell is fabricated in the form of parallel plate capacitors by embedding the sensitive material in polystyrene binder and sandwiching it between the two conducting glass plates. All the measurements are taken with the help of parallel plate capacitor having thickness varying from 0.3 cm to 0.5 cm with the electrode area 1.5 cm to 2.5 cm. For the measurement purpose of photoconductivity, the cell is placed in complete dark chamber and the radiations from a 2500 Watt mercury lamp is allowed to fall through a

window over the upper surface of the cell. The intensity of light over the upper surface of cell can be varied by changing the slit width or changing the distance between the lamp and hole. In our experiment, we have prepared two samples having different proportions. The first sample is made by taking 40% Mg, 40% TiO_3 and 20% ZnO named as MTZ1 and second sample is made by taking 46% Mg, 46% TiO_3 and 8% ZnO named as MTZ2.

III. RESULT & DISCUSSION

A. Effect of Voltage

The figure (1.1 and 1.2) shows variation between photocurrent (I_p) and voltage (V). The variation of photocurrent (I_{pc}) and applied voltage (V) is plotted on ln-ln scale which can be expressed by power law ($I_{pc} \alpha V^s$) , where s represents slope of the line[14]. The behavior of graph can be explained on the basis of value of slope. For s < 1, the variation of photocurrent versus voltage is sub-linear. For s =1, the variation of photocurrent versus voltage shows linear (ohmic) behavior, and for s >1, the behavior of photocurrent with voltage is super-linear.

Sample MTZ1

In the presence of light, the photocurrent increases slowly with applied voltage upto 4.5 volt and with further increase in voltage, the photocurrent increases rapidly and shows a nonlinear (I – V) characteristic. A non linear curve normally exhibits the existence of different kind of conduction mechanism. For lower value of applied voltage, the value of slope is smaller than that at higher voltage. Thus the slope changes its behavior with the variations of applied voltage. With the help of graph, we can conclude that photocurrent- voltage (I-V) curve shows two regions of conduction:

(1) Shows sub-linear behavior upto 4.5 volt
(2) Shows super-linear behavior after 4.5 volt

The above conduction processes can be explained in such a way that for low voltages the density of injected charge carrier is lower than the density of thermally generated charge carriers which in turn leads to the ohmic behavior. For high voltages, the value of slope is greater than that of at low voltages, which suggests that at higher voltage the conduction is primarily dominated by the trap limited space charge limited conduction (SCLC) mechanism [12]. The trap limited space charge limited current varies with voltage. The voltage at which the curves changes its slope i.e. from sub-linear to super-linear is known as transition voltage (V_t = 4.5 Volt). The transition of sub-linear to super-linear variation is ascribed to flow of trap limited as well as space charge limited current inside the material. The super-linear behavior of the photocurrent shows that some of the charge carriers are being injected into the material from the electrode [13, 14].

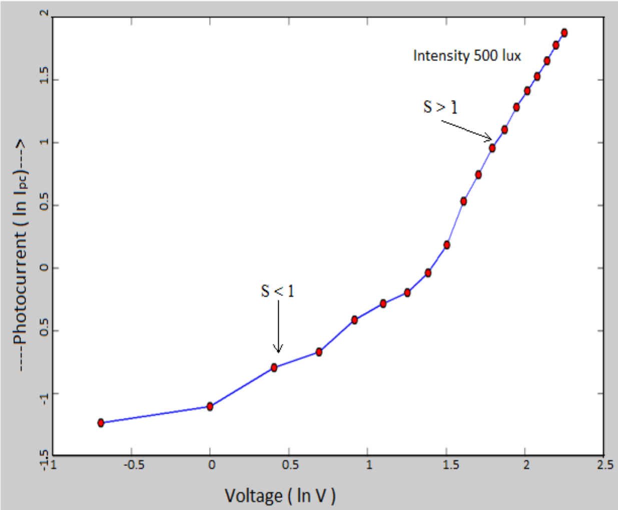

Figure 1.1: Variation of Photocurrent with Voltage on ln-ln scale for sample MTZ1

Sample MTZ1

From the graph it is clear that, at low voltage (nearly 2 volt), the photocurrent increases sub-linearly with the increase in temperature. It is observed that with the increase in temperature there is rapid and incremental change in photocurrent. At 4 volt photocurrent first increases rapidly with increasing temperature and shows a hump in the curve. Further increase in temperature shows decrement in the photocurrent which further increases sub-linearly with increasing temperature. Similar variation of temperature versus photocurrent is observed at 6 volt. The points at which photocurrent increased rapidly, thermal quenching of photocurrent occurs. Thermal quenching of photocurrent can be explained on the basis of Rose Model.
According to Rose model, the steady state Fermi levels are shifted towards their respective band edge with an increase in the intensity of illumination. During this shift of Fermi levels, a large number of traps are converted into recombination centers. Thus, life time of the electron increases and the photoconductivity is sensitized. Fermi levels are shifted towards the middle of the gap with an increase in temperature.

Sample MTZ2

From the figure, it can be observed that sample MTZ2 shows approximately similar behavior as observed for MTZ1. This shows that voltage- current characteristics are in good agreement for both the samples having different compositions of material.

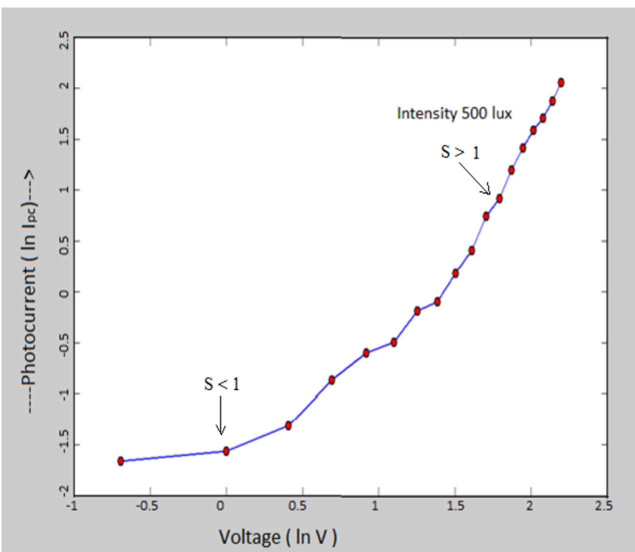

Figure 1.2: Variation of Photocurrent with Voltage on ln-ln scale for sample MTZ2

B. *Effect of Temperature*

The figure (2.1 and 2.2) shows the variation of photocurrent with temperature for sample MTZ1 and MTZ2. These curves are plotted for three discrete values of voltage i.e. 2V, 4V and 6V. The graphs plotted are non linear in nature.

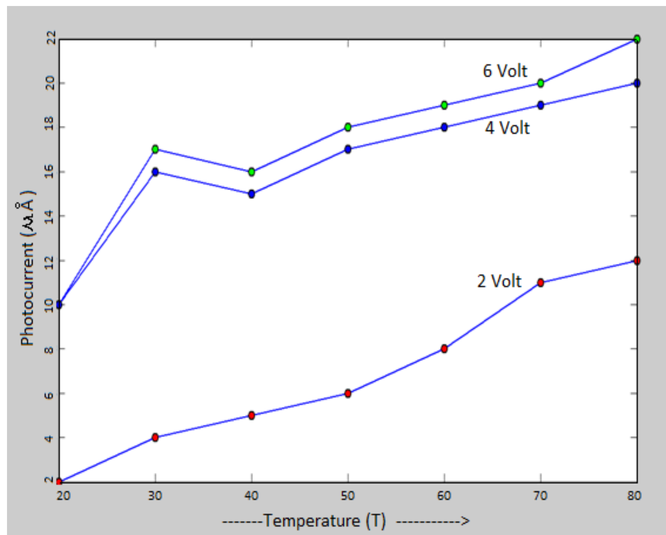

Figure 2.1: Variation of Photocurrent with Temperature for sample MTZ1

Sample MTZ2

It has been observed from the curve that photocurrent increases sub-linearly with increasing temperature at 2 volt. For 4 volt and 6 volts graph depicts approximately similar pattern. It is observed from the curve that it has small value of slope. It is observed that at 2 volt there is minimal impact of temperature on photocurrent. Similar patent is observed at 4 and 6 volt. On comparing MTZ1 and MTZ2, it is concluded that higher the concentration of ZnO, larger will be the photocurrent. Hence MTZ1 demonstrate great impact on photocurrent.

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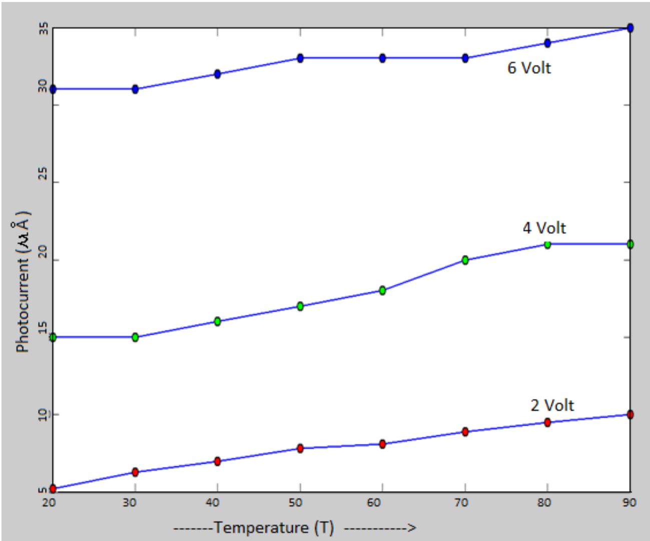

Figure 2.2: Variation of Photocurrent with Temperature for sample MTZ2

C. Effect of Intensity

The figure (3.1 and 3.2) shows the variation of photocurrent with intensity at fixed voltage (5 Volt) for the sample MTZ1 and MTZ2. The variation of photocurrent (I_{pc}) and applied intensity of illumination (L) is plotted on ln-ln scale which can be expressed by power law (I_{pc} α L^s), where s represents slope of the curve. The variation of intensity is measured in Lux and photocurrent is measured in micro-ampere. The curves plotted between photocurrent (I_p) and intensity (L) for sample MTZ1 and MTZ2 are non-linear in nature having different slopes in different regions of lower and higher intensity of illumination.

Sample MTZ1

For lower intensity region curve shows sub-linear nature of photocurrent whereas for higher intensity region, it depicts super-linear nature of photocurrent. The sub-linear ($s < 1$) and super-linear ($s > 2$) nature of photocurrent can be explained on the basis of class I and class II states. Class I consist of states which have roughly similar cross section for electron and holes while the class II states have a higher capture cross section for holes than for electrons and lie close to the valence band. The sub-linear behavior of photocurrent illustrates that the class I centres are distributed continuously between the conduction band and Fermi level [15]. On further increasing the intensity of illumination, it has been observed that separation between conduction band and electron Fermi levels is highly increased, due to which large number of class I centres are converted into recombination centres. Thus the life time of electron decreases gradually. This decrement in life time of electron reduces the rate of increase of photocurrent which in turn, is responsible for sub-linearity in the photocurrent. On further increasing the intensity of illumination, the holes Fermi level are shifted towards valence bands and electron Fermi levels are shifted towards conduction bands. Due to this class II states are converted in recombination centres, which characterize the super-linear behavior of photocurrent.

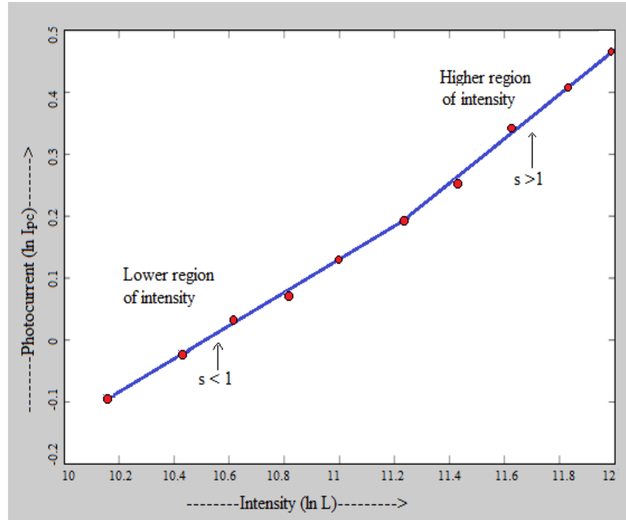

Figure 3.1: Variation of Photocurrent with intensity on ln-ln scale for sample MTZ1

Sample MTZ2

In lower intensity region, the photocurrent increases sub-linearly with increased intensity of illumination whereas in higher intensity region, increase in intensity of illumination further increases the photocurrent but it has been observed that slope of the curve is reduced to a lower value as compared to the lower intensity region.

On comparing both the samples it has been observed that both the curve depicts overall increase in photocurrent with increase in intensity of illumination. MTZ1 shows sub-linear to super-linear increment in photocurrent with enhancing slope of the curve whereas MTZ2 portrays the approximate similar behavior but with reduced slope. The comparative study of both the samples MTZ1 and MTZ2 concluded that variation of photocurrent with intensity of illumination depends on the compositions of the material. We can also confer that higher will be concentration of ZnO, higher will be the photocurrent produced.

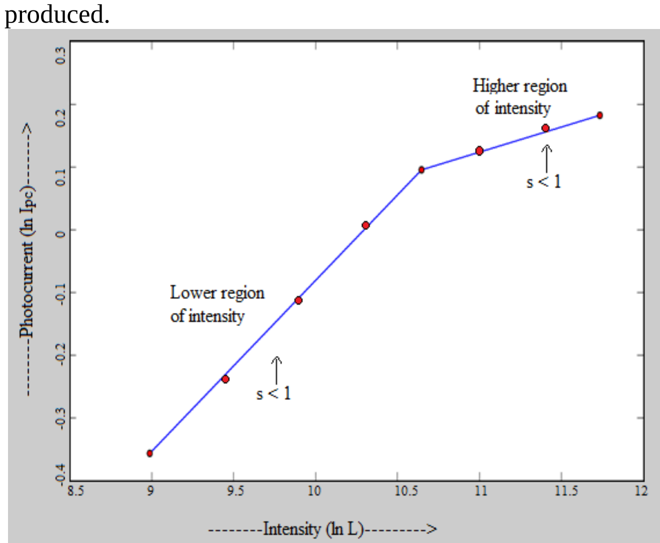

Figure 3.2: Variation of Photocurrent with intensity on ln-ln scale for sample MTZ2

IV. CONCLUSION:

We have studied effect of voltage, intensity of illumination and temperature on the photoconductivity of mixed composite of $MgTiO_3$ and ZnO in different compositions. The photocurrent-voltage (I-V) curve shows two regions of conduction: for s<1, photocurrent shows sub-linear behavior and for s>1, the photocurrent shows super-linear behavior. The transition of sub-linear to super-linear variation is ascribed to flow of trap limited as well as space charge limited current inside the material. The voltage- current characteristics are in good agreement for both the samples having different compositions of material. On increasing the temperature, the Fermi levels are shifted towards the middle of the gap. Due to this shift of Fermi levels, a large number of traps are converted into recombination centers. Thus, life time of the electron increases and the photoconductivity is sensitized. The curves plotted between photocurrent (I_p) and intensity (L) for both sample MTZ1 and MTZ2 are non-linear in nature having different slopes in different regions of lower and higher intensity of illumination. The comparative study of both the samples MTZ1 and MTZ2 concluded that variation of photocurrent with intensity of illumination depends on the compositions of the material. We can also confer that higher will be concentration of ZnO, higher will be the photocurrent produced.

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