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10/82/85  
INTERNAL REPORT  
(Limited distribution)

International Atomic Energy Agency

and

United Nations Educational Scientific and Cultural Organization

INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS

SURFACE PHOTOCONDUCTIVITY IN AMORPHOUS  $As_2Se_3$  \*

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## ABSTRACT

The presence of surface effects has been detected by noting changes in photoconductivity for the same sample under test in air at normal room temperature and pressure and in vacuum. For all samples, it has invariably been found that photo-sensitivity is increased by changing from an air to a vacuum surround. In order to investigate which of the atmospheric constituents was responsible for the change, the photoconductivity response spectrum of a thin film of amorphous  $As_2Se_3$  was measured at room temperature (297K) in a flow of dry  $N_2$ , dry  $O_2$  and  $H_2O$ . No change in the photoconductivity response spectrum was noted in the ambient of  $N_2$  and  $O_2$ . However, water vapour was found to decrease the photosensitivity of  $As_2Se_3$  and shift the photoreponse spectrum towards the lower wavelength side. These changes were found to be reversible in nature.

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July 1982

\* To be submitted for publication.

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## 1. INTRODUCTION

For the last two decades or so there has been a wide spread interest in amorphous semiconductors, partly due to a desire to confront fresh problems and partly due to an increasing awareness of their potential applications which range far beyond the optical and structural uses of the silica glasses (1,2). The photoconductive and electrical properties of these solids have been studied extensively. The chalcogenide glasses based on Se have attracted particular attention. Photoconductivity in amorphous chalcogenides has proven to be a valuable tool in understanding the nature of localized states and transport in these materials (3-8).

In spite of the fact that the physics of amorphous solids has developed into an extensive and ramified field, the nature of an amorphous semiconductor surface still remains unexplored. Attempts made to study the surface states in these semiconductors so far have generally been unsuccessful (9-10). To the author's knowledge, only two successful cases of field effect measurements in chalcogenide glasses (based on Te) have been reported (10-11); whereas similar investigations in the case of relatively simple amorphous semiconductors like Se and  $As_2Se_3$  have been reported to be unsuccessful (10).

Measuring the photoconductivity response spectrum of amorphous  $As_2Se_3$  specimens in air at normal room temperature and pressure and then in vacuum for the same specimen under test, we noted that not only did the photocurrent increase, but the maximum (and the long wavelength edge) of the photoconductivity response spectrum was also displaced toward the lower photon energies, keeping the incident photon flux, specimen temperature, and the electric field across the electrodes constant; thus indicating an increase of the photosensi-

tivity defined as the photocurrent per unit incident photon flux (13) and a decrease of the photoconductivity activation energy  $E_{ph}$ , defined as the photon energy corresponding to the maximum photoresponse (also defined as the red edge photon energy at which the photoconductivity has fallen to half its maximum value (23,24)). The increase in the photocurrent was noted to vary from sample to sample. For bulk samples, this change was relatively small compared with thin films. Under present experimental conditions, it changed, approximately from 30% to 55% of its value in air. A change in  $E_{ph}$  ranged from 0.03 eV to 0.05 eV. The rise and decay of the photocurrent was also affected. In vacuum it was comparatively slower. In vacuum the dark resistance of the specimens was also found to increase. As evacuation of the specimen was started, its resistance increased gradually, reaching a saturation value in about 12 hours degassing time at  $10^{-4}$  torr. On breaking the vacuum, the reverse occurred but now the saturation time limit was found to be considerably large (~24 hours). For thin films, an increase in the dark resistance ranged, approximately from 20 to 25%.

The Observed phenomenon was noted for amorphous  $As_2Se_3$  as well as its compounds with  $Tl_2Se$ . But in the present paper as a typical case, these measurements are presented for  $As_2Se_3$  thin films only. It may however, be mentioned here that the magnitude of the change was found to decrease with increase of  $Tl_2Se$  concentrations. To the author's knowledge, no such information is available in the literature at least for  $As_2Se_3$  and its compounds with  $Tl_2Se$ . For crystalline semiconductors, however, such measurements have been made quite often (12-20). Utsuni and Wada (21) have simply stated

that water vapour affected the storage phenomena in  $As_2Se_3$  thin films adversely. Kastner and Fritzsche have reported that water vapour decreases the surface conductance of amorphous Ge but by a very small amount, approximately 1%; whereas  $H_2$  and  $O_2$  etc, had no such effect (22).

In order to investigate the nature of the phenomenon, the photoconductivity response spectrum of a thin film (~1  $\mu$ m) of amorphous  $As_2Se_3$  was measured in the atmosphere of the following ambients:

- i) air at normal room temperature and pressure
- ii) vacuum of the order of  $10^{-6}$  torr
- iii) dry nitrogen
- iv) dry Oxygen
- v) water vapour<sup>\*</sup> carried by nitrogen

## 2. EXPERIMENTAL

### 2.1 Synthesis of Glasses and Specimen Preparation

These glasses were synthesized by the conventional method of heating the constituents (all of 99.999% purity) in evacuated and sealed off silica glass ampules to high temperatures above the melting temperature of the highest melting constituent, followed by quenching in water. During heating, the ampules were shaken for several times to ensure a thorough mixing of the constituents. The glasses thus prepared were completely amorphous. The amorphous state of the glasses was checked by examining their X-ray diffraction patterns. Thin films were prepared on freshly cleaved mica (muscovite) substrates by the thermal evaporation technique in vacuum ( $2 \times 10^{-6}$  torr). The substrate temperature during evaporation was maintained at 273 K.

## 2.2 Photoconductivity Measurements

The photoconductivity response spectrum measurements were made using evaporated gold electrodes in the coplanar geometry with an interplanar spacing of  $1.0 \pm 1\%$  nm. Also these measurements were made at room temperature (297K) and at 500 V/cm value of the electric field across the electrodes. Dry batteries were used for the d.c. voltage supply. To avoid any space-charge effects, the specimens were illuminated uniformly. The photoresponse spectra were corrected for the lamp output spectrum and were recorded at low photon flux ( $10^{11}$  photons/cm<sup>2</sup>sec.). The specimens before being subjected to the photoconductivity measurements were kept in the dark in the experimental chamber for an overnight period of about 20 hours. Amorphous  $As_2Se_3$  and its compounds with  $Tl_2Se$  were found to show the persistence of photoconductivity which tended to enhance in vacuum. Therefore, to avoid any error in the measurements, the photocurrent was allowed to decrease to its minimum value ( $\sim 1\%$  of its maximum value) after each reading and after the illumination of the specimen was ceased. Also during illumination, the photocurrent was allowed to reach its maximum (saturation) value before being recorded.

In order to ensure the absence of water vapour in nitrogen and oxygen, these gasses before their admittance into the experimental chamber, were made to pass through a coil having 6 turns (each turn of about 15 cm diameter) and kept immersed in a liquid air bath. The pressure inside the experimental chamber was maintained at atmospheric

level (760 torr) by allowing the gas to enter the chamber at one end and leave at the other, through a narrow orifice in the chamber base plate, made specially for the purpose. For water vapour nitrogen was used as the carrier. The amount of water vapour carried by nitrogen could be varied by varying the length of its path in the water bath which was constructed out of Buchner flasks and nitrogen gas was bubbled into it. These measurements were made at about 70% humidity level.

## 3. RESULTS

Results of these investigations are shown in Figs.1 to 3. Fig.1 shows the photoconductivity spectra of the film under test, in air (curve a), vacuum (curve b) and in the atmosphere of nitrogen (curve c). Fig.2 shows the photoconductivity spectrum of the same film in the atmosphere of oxygen (curve a), vacuum (curve b) and in water vapour (curve c). These photoresponse spectra were recorded after keeping the film in the atmosphere of these gases for about 48 hours.

The photoconductivity response spectrum maximum in air was found to occur at  $1.80 \pm 0.01$  eV. On evacuating the system to  $10^6$  torr and degassing it for about 20 hours, the photoconductivity spectrum maximum has shifted toward lower wavelength side (Fig.2, curve b) and now occurs at about 1.75 eV, suggesting a shift in the photoconductivity activation energy by about 0.05 eV. Also in vacuum the photocurrent was increased by, approximately 35%. As is obvious from Figs.1 and 2, no change in the photoconductivity response spectrum maximum was noted in dry  $N_2$  and  $O_2$

even upto 48 hours. The only change observed is the decrease in the photocurrent in both the cases by about 10%. This decrease in the photocurrent may be regarded as due to a decrease in the specimen temperature ( $\approx 6^\circ\text{C}$ ). This change in temperature does not shift the photoresponse spectrum but can decrease the photocurrent considerably (25). The change in the thermal exchange from the specimen in a nitrogen or oxygen atmosphere may also contribute towards the decrease in the photocurrent.

Curve c of Fig.2 shows the photoconductivity response spectrum measured in the atmosphere of water vapour carried by nitrogen, after about 23 hours keeping the film in this atmosphere. This curve shows that photoconductivity response spectrum has shifted toward higher energy side compared with that in vacuum (curve a). The photoconductivity maximum now occurs at 1.80 eV, suggesting that the water vapour has increased the photoconductivity activation energy by about 0.045 eV compared with its value in vacuum. Curve c' of Fig.2 also shows that the photocurrent has, approximately fallen to a value comparable with that in air. This establishes that water vapour is mainly responsible for the observed phenomenon.

Fig.3 shows the results obtained to determine nature of the reaction of water vapour with the specimen. After recording the curve c of Fig.2, the system was evacuated to  $10^{-6}$  torr and degassed for about 20 hours and under the same experimental conditions as above, the photoconductivity response spectrum of the specimen was measured. The photoresponse spectrum was shifted again toward the lower energy side by, approximately 0.045 eV, the same amount as before. This is shown in curve a.

After this, the film under test was exposed to air at normal room temperature and pressure, for about 50 hours and then its photoconductivity spectrum was measured. Curve b was obtained, which shows a maximum at about 1.80 eV. The photocurrent was also decreased as previously. Curve c was obtained on evacuating the system again. As is obvious, the photoresponse spectrum is again displaced and the photocurrent increased. This confirms that the reaction of water vapour with at least vitreous  $\text{As}_2\text{Se}_3$  is reversible in nature.

#### 4. DISCUSSION

These investigations certainly prove that the observed changes in the spectral distribution of photoconductivity of amorphous  $\text{As}_2\text{Se}_3$  are caused by water vapour in the air. The reversible nature of the changes suggests that these are caused due to adsorption of water molecules onto the surface of the specimen. On evacuation of the system, these adsorbed water molecules disappear gradually and change resistance of the specimen accordingly.

The observed decrease in the photosensitivity and shift of the photoconductivity response spectrum toward lower wavelength side in vacuum, may be explained in terms of b-and-bending of the semiconductor under the influence of a localized charged layer at the proper surface due mainly to surface states.

The surface of a finite solid possesses surface energy states caused due to abrupt termination of the lattice or as in covalent solids, due to the unsatisfied dangling bonds. The surface states may also originate due to the presence of contaminations like

gases ( $O_2$ ,  $NH_3$ ,  $O_3$  etc.) or liquids like  $H_2O$ , having an electric dipole moment (19, 20). In the presence of these states, the surface acquires a potential difference with respect to the bulk, and hence gives rise to a space-charge layer near the surface. Due to this space-charge layer the energy bands which are horizontal throughout the bulk may bend upwards or downwards as the surface is approached.

The surface of a semiconductor can be bent in three different ways viz, accumulation, depletion and inversion layers. In the case of an accumulation layer the body of the semiconductor is depleted of the majority carriers which are taken to the surface (19, 20, 27). The minority carriers are negligible both in the bulk and in the space-charge region. Hence, when an accumulation layer is formed the surface recombination decreases. The accumulation layers are, therefore, characterized by high conductance, which is due to the large number of majority carriers.

The formation of a depletion layer on a semiconductor corresponds to the situation in which the majority carriers (holes in the present case) are depleted from a region near the surface, but as yet the minority carriers' concentration does not exceed that of the majority carriers (19, 20, 27). Thus on the formation of a depletion layer, the sign of the current carriers does not change and the current is continued being carried by the majority carriers. Therefore, with the formation of a depletion region, the surface recombination is enhanced, and the surface conductance decreases (27 - 30) due to the reduced number of majority carriers available for conduction. A depletion region finally goes over to

an inversion region if the bands are continued bending in the same direction. Here the greatest contribution to the space-charge comes from the carriers of the opposite sign to those of the bulk (i.e. from electrons in a p-type semiconductor (19, 27, 30)). Thus we would have an n-type region on a p-type semiconductor and vice versa. The point where the minority carrier density equals the majority carrier bulk density, represents the limit of the depletion region. On further bending the bands in the same direction, the minority carriers (electrons on a p-type semiconductor) start accumulating on the surface. Therefore, the sign of the current carried changes from that of the holes (majority carriers) to that of the electrons (minority carriers). The inversion layer as well, like accumulation layers, are characterized by high conductance with the difference that in an inversion layer, it is due to the large number of electrons for a p-type semiconductor.

The band-bending and hence the expected thickness of the space-charge region into the semiconductor can be calculated from the knowledge of the Debye screening length  $L$  given by (27)

$$L = \left( \frac{\epsilon \epsilon_0 kT}{pe^2} \right)^{1/2}$$

where  $\epsilon$  is the dielectric constant of the semiconductor,  $\epsilon_0$  the permittivity of the free space,  $k$  is the Boltzmann's constant,  $p$  the hole concentration,  $e$  the electronic charge, and  $T$  the temperature. The space-charge region decays exponentially with  $L$  (27) which in turn varies as  $p^{-1/2}$ . This suggests that with increasing the bulk carrier density, the space-charge region tends to become thinner.

The Debye screening length for amorphous  $As_2Se_3$  ( $\rho \approx 10^{18} \text{ eV}^{-1} \text{ cm}^{-3}$ ,  $\epsilon \sim 9.5$ , (31) at room temperature (297K) is, approximately  $4 \times 10^{-7} \text{ cm}$ . This is considerably less than the thickness of the film ( $\sim 10^{-4} \text{ cm}$ ) used in the present investigations. This shows that the band-bending (and hence the depth of the space-charge region) is not large, it extends only upto a small distance into the bulk from the surface of the film.

As these thin films were deposited on mica (muscovite, room temperature resistivity greater  $> 10^{13} \text{ Ohm cm}$  (32)) substrates, therefore, because of the much smaller free carrier density of mica compared with that of amorphous  $As_2Se_3$ , the semi-conductor-substrate interface is not expected to affect the observed phenomenon.

Since in the presence of water vapour no increase in the conductivity was noted, the observed phenomenon does not seem to be due to the formation of an accumulation layer. The magnitude of the effect is small and no change in the direction of the current (due to sign of the carriers) was noted, at least upto 70% humidity level. Therefore, the possibility of the formation of an inversion layer, is also excluded. The observed phenomenon may, therefore, be explained in terms of the formation of a depletion layer by the water vapor as a result of which the majority carrier (hole) density near the surface is decreased and the number of recombination centres (near the surface) is increased. Water molecules on the surface create electrostatic field due to their large dipole moments which on an appropriate depth from the surface changes the electrical properties of the fast recombination centres. As a result of this, the surface recombination increases and hence the lifetime of the carrier decreases (33). Thus in the presence of water vapour the number of charge carriers (holes, in the present case) contributing

to the photocurrent, decreases.

Also in the presence of a space-charge region, the mobility of the carriers due to scattering by the surface, is decreased (27-30). The mobility of all the carriers which move close to the surface decreases more than that of those which move away from it (27). This reduction in mobility due to the presence of a space-charge layer becomes increasingly significant with the thickness of the specimen decreasing (27). Decrease in mobility due to scattering (physical) of carriers in the case of amorphous materials (the low mobility solids where the carrier mean free paths are small) is not expected to be very significant. However, we may expect deflection away from the space-charge layer, of the majority carriers (holes). In the coplanar arrangement of electrodes current flows parallel to the surface. The internal electric field (due to space-charge) transverse to the surface, may keep the holes away from the surface.

In vacuum in the absence of water-vapour the surface recombination decreases due to the decrease in the number of recombination centres and hence the increase of the carrier lifetime, (33). Therefore, the number and the mobility of the charge carriers contributing to the photocurrent, increases. This results in an increase in the photosensitivity of the semiconductor.

The observed increase in the photosensitivity in vacuum is evidently connected with the increase in the effective value of the product  $q\mu$  compared with its value in the ambient of water-vapour. Here  $q$  is the quantum efficiency and is a wavelength dependent parameter at least for  $As_2Se_3$  (34).  $\mu$  is the carrier mobility and

$\tau$  the recombination carrier lifetime. These quantities occur in the expression for photoconductivity (23). The quantum efficiency  $q$  may be written as  $q = 1/(1 + \tau_i/\tau)$ ,  $\tau_i$  is the ionization time(35). The product  $q\mu\tau$  does not seem to vary in the same way for all photon energies. The effect will be most prominent where the slope of the  $\ln q$  versus photon energy plot is the greatest i.e. at low photon energies. This manifests itself by displacing the photoconductivity response spectrum towards the lower wavelength side.

The results also show that oxygen and nitrogen gases do not affect the behaviour of the surface of amorphous  $As_2Se_3$ . This is probably due to the physical adsorption where no charge transfer like chemisorption takes place and impurity atoms/molecules are held on the surface by the weak forces like polarization or Van der Waals (36) of these gases on the surface of the semiconductor.

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**FIGURE CAPTIONS:**

- Fig. 1. Photoconductivity response spectra of a thin film of amorphous  $As_2Se_3$  in:
  - a) air; b) vacuum  $10^{-6}$  torr;
  - c) dry nitrogen at 760 torr
  
- Fig. 2. Photoconductivity response spectra of a thin film of amorphous  $As_2Se_3$  in:
  - a) vacuum  $10^{-6}$  torr; b) dry oxygen at 760 torr; c) water vapours carried by nitrogen
  
- Fig. 3. Photoconductivity response spectra of a thin film of amorphous  $As_2Se_3$  in:
  - a) vacuum  $10^{-6}$  torr (after the measurement of photoconductivity response spectrum in the presence of water vapour); b) air; c) vacuum  $10^{-6}$  torr



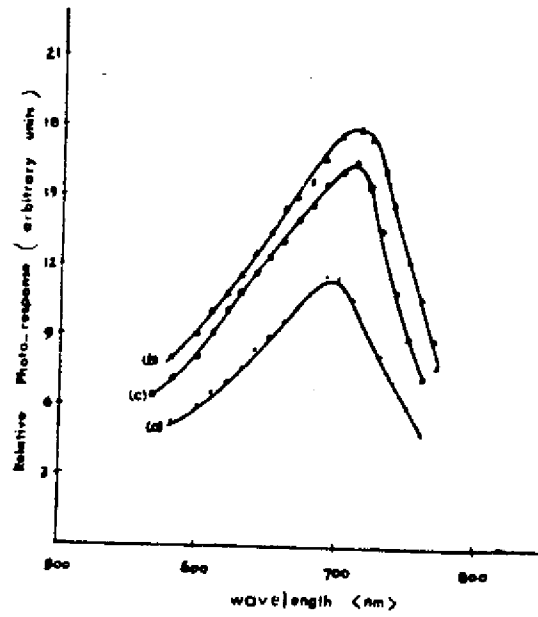


Fig.1

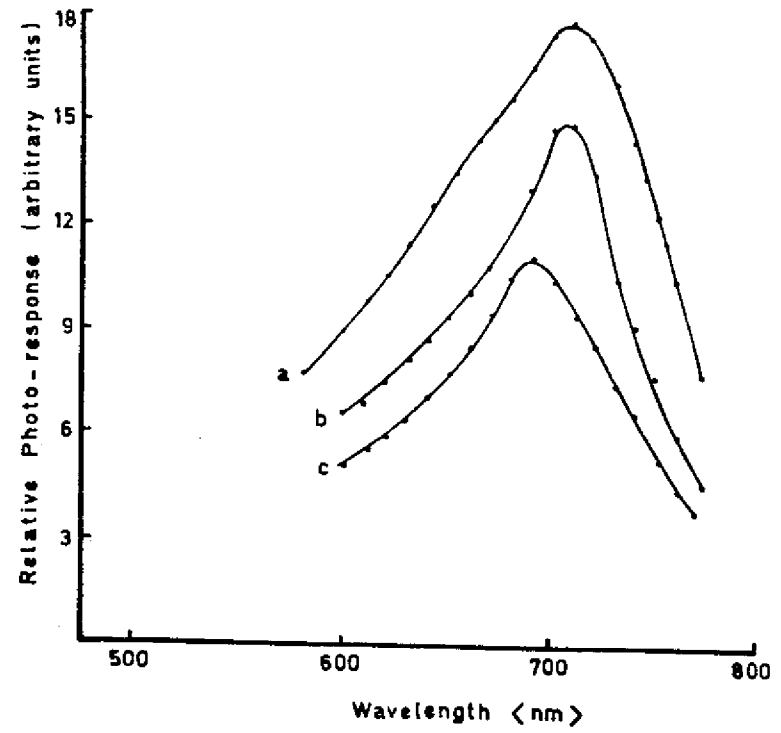


Fig.2

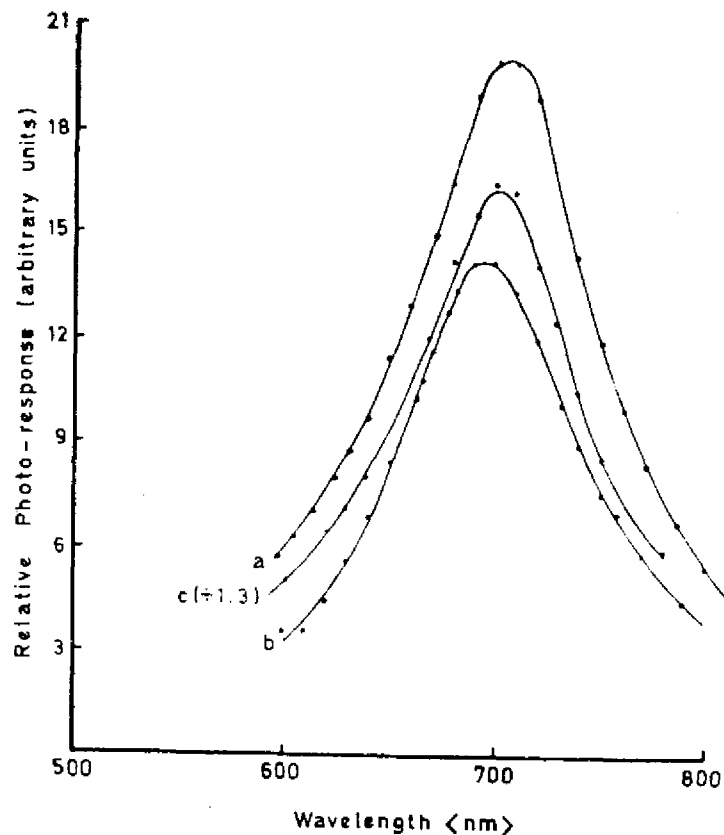


Fig.3

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