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Effect of photo-crystallization on the photoconductivity of a-Se₈₅Te₁₅ (*)

A. Kumar, R. Misra, S. Goel and S. K. Tripathi

Department of Physics, Harcourt Butler Technological Institute, Kanpur-208 002, India

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Résumé. — Cet article traite de la photoconductivité en régime permanent et transitoire de films minces de l'alliage amorphe Se₈₅Te₁₅ avant et après photocristallisation. Celle-ci est induite par illumination sous lumière blanche (3 000 Lux) pendant environ 18 heures. Les résultats indiquent que la conductivité à l'obscurité (σ_d) et la photoconductivité en régime permanent (σ_{ph}) augmentent de plusieurs ordres de grandeur après cristallisation. Le régime transitoire est aussi affecté par la photocristallisation. Les résultats sont interprétés en termes d'états de défauts dans le matériau.

Abstract. — The present paper reports the steady state and transient photoconductivity in amorphous Se₈₅Te₁₅ thin films before and after photo-crystallization which is induced by shining white light (3 000 Lux) for about 18 hours. The results indicate that the dark conductivity (σ_d) and steady state photoconductivity (σ_{ph}) increase by many orders of magnitude on crystallization. The transient behaviour is also affected on photo-crystallization. The results are interpreted in terms of the defect states in this material.

Photo-crystallization in amorphous solids was first observed in amorphous selenium by Dresner and Stringfellow [1] and then by de Neufville [2], Clement *et al.* [3] and Kim and Turnbull [4]. These authors showed that photo-crystallization effects result from the production of hole-electron pairs in the vitreous phase. Kotkata *et al.* [5] have studied the effect of light on crystallization kinetics in amorphous Se doped with sulphur and showed that the effect of light shining is to destroy the Se-Se bonds, reducing the Se chain length and opposing the crystal growth. The photo-crystallization in amorphous Ge_xSe_{1-x} ($0 \leq x \leq 0.2$) has been investigated by Matsushita *et al.* [6] and they have shown that only for Ge_{0.5}Se_{0.95} films, the photo-crystallization effect is enhanced over thermal effects. However, in the case of Se_{1-x}Te_x films, Okuda *et al.* [7] have shown that photocrystallization is suppressed for $x = 0.04$ whereas for the other compositions ($x > 0.1$), the photo-crystallization was certainly enhanced as compared to pure Se films.

In most of the above cases, light was shone on the samples at temperatures near the crystallization

temperature. The light increased or decreased the rate of crystallization as compared to purely thermal effects. However, in a recent study [8], we have observed photo-crystallization in a-Se₈₅Te₁₅ films at room temperature where the possibility of thermally induced crystallization is negligible.

The present paper reports the effect of photo-crystallization on the photoconducting behaviour of amorphous Se₈₅Te₁₅ thin films prepared by vacuum evaporation technique described earlier [9]. This particular composition is chosen because of its higher photosensitivity ($\sigma_{ph}/\sigma_d = 69$ at 370 Lux). The photocrystallization is achieved by shining white light at room temperature for 18 hours in a vacuum $\sim 10^{-3}$ Torr. Such an exposure is capable of changing dc conductivity permanently by 3-4 orders of magni-

The steady state and transient photoconductivity measurements have been made in two states (i) before crystallization (state A) (ii) after photo-crystallization (state B). Results indicate that the transient photoconductivity behaviour is quite different in these two states. The method of measurements is same as reported earlier [9].

Temperature dependence of the steady state photoconductivity is studied at various levels of illumination (30 Lux to 370 Lux). The results of these measurements for a particular intensity

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(370 Lux) in states A and B have been shown in figure 1. The same figure also contains the plots of the temperature dependence of the dark conductivity (σ_d) in both the states. The activation energy for the dc conduction (ΔE) is calculated from the slopes of $\ln \sigma_d$ vs. $1000/T$ curves and the values of ΔE for states A and B are inserted in table I. It is clear from this table that ΔE decreases and σ_d increases quite appreciably after exposing the film with white light for about 18 hours (state B). This is due to photo-crystallization of the amorphous film after shining the white light [8]. The values of σ_{ph} at 295 K in states A and B are also given in table I. σ_{ph} increases on photo-crystallization which indicates that the material is becoming more photoconductive in absolute physical terms. σ_{ph} , in general, is proportional to the product of the carrier mobility by its lifetime. The observed increase of σ_{ph} may be related to the increase in carrier mobility due to crystallization. However, the band gap and optical absorption

coefficient may also change during crystallization which may also influence the photoconductivity. Moreover, the influence of grain boundaries and tissue structures should also be taken into account as structural changes are quite likely on crystallization. In the absence of these details, the exact reasons for the increase in photoconductivity on crystallization cannot be predicted.

Figure 2 shows the rise and decay of the photo-current at room temperature and at an intensity of 370 Lux for both the states A and B. It is clear from this figure that the rise and decay curves are quite different in two states (A and B).

A peak in the rise curve is observed in state A and the decay is quite fast in this state. This anomalous behaviour in the rise curve was also observed [9] by us in $\text{Se}_{80}\text{Te}_{20}$ alloy and explained in terms of non-equilibrium recombination as suggested by Andriesh *et al.* [10]. According to them, such a maximum occurs in high intensity region where bimolecular

Fig. 1. — Temperature dependence of dark and photoconductivity (at 370 Lux) in states A and B.

Fig. 2. — Transient photoconductivity (at 370 Lux) at room temperature in states A and B.

Table I. — Electrical parameters in amorphous thin films of $\text{Se}_{85}\text{Te}_{15}$ in states A and B.

Parameters	State A	State B
σ_d at 295 K	$1.25 \times 10^{-9} \Omega^{-1} \text{cm}^{-1}$	$1.76 \times 10^{-6} \Omega^{-1} \text{cm}^{-1}$
ΔE	0.39 eV	0.20 eV
σ_{ph} at 295 K	$8.61 \times 10^{-8} \Omega^{-1} \text{cm}^{-1}$	$8.61 \times 10^{-6} \Omega^{-1} \text{cm}^{-1}$
σ_{ph}/σ_d at 295 K	68.8	4.9

recombination takes place. At lower intensities where monomolecular recombination takes place, this maximum may be absent and the photo-current rises monotonically to the steady state. Maan *et al.* [11] have observed similar maximum in case of $\alpha\text{-In}_{20}\text{Se}_{80}$ and the decay of photocurrent during illumination has been attributed to recombination between holes in the valence band with electrons at recombination centres. The possibility of light induced defects as a possible cause for such a behaviour may also not be ruled out.

It is interesting to note that such a maximum in the rise of photocurrent disappears in the crystallized state (see Fig. 2). This indicates that the anomalous behaviour of the rise of photocurrent may be a characteristic of amorphous state. The decay of photocurrent after the cessation of light is also quite slow in crystallized state (state B). A persistent photocurrent (asymptotic value of the photocurrent in the decay curve) also exists in this state. The persistent photocurrent is observed in many chalcogenide glasses [12-15]. The exact reasons for such a persistent current are not yet clear. However, it is believed [14] that this current may not be simply due to the carriers trapped in the localized states. To simplify the analysis, we subtracted the persistent photocurrent from the measured current during the decay process. It is found that $\ln I_{\text{ph}}$ vs. time curves do not yield straight lines even after subtracting the persistent photocurrent. This indicates that the decay process is non-exponential.

To study the decay rate in case of non-exponential decay, we prefer to use the differential lifetime concept as suggested by Fuhs and Stuke [16] and is given by

$$\tau_d = - \left[\frac{1}{I_{\text{ph}}} \frac{dI_{\text{ph}}}{dt} \right]^{-1}. \quad (1)$$

In the case of an exponential decay the differential lifetime will be equal to the carrier lifetime. However, in the case of a non-exponential decay, τ_d will increase with time and only the value at $t = 0$ will correspond to the carrier lifetime.

From the slopes of I_{ph} vs. time curves, we have

calculated the values of τ_d using equation (1) at various times for a decay curve of figure 2. Corrected values of photocurrent are used for these calculations. The results have been plotted in figure 3. It is clear that τ_d increases with the increase of time. This confirms the non-exponential decay in the present case as, for an exponential decay, τ_d should be constant with time.

It is clear from figure 3 that τ_d at a particular time is more in state B as compared to state A. This indicates that the decay of photocurrent becomes slower after photo-crystallization. A slower decay of photocurrent in state B indicates that the density of defect states increases after photo-crystallization in the present case.

Fig. 3. — Differential lifetime as a function of time during photo-conductive decay in states A and B.

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