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**LETTER TO THE EDITOR**

# Anomalous DC dark conductivity behaviour in a-Se films

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**Abstract**
Thin films of amorphous selenium have been prepared by thermal evaporation. DC conductivity measurements were carried out on these films in the temperature range between 208 and 322 K. Above room temperature, the dark conductivity is thermally activated with activation energy $E_\sigma$ = $1.05 \pm 0.08$ eV. For temperatures below 285 K, an increase in the dark current is observed, which is interpreted in terms of a shift of the Fermi level that makes more states available for a hopping process.

## 1. Introduction

For a considerable length of time, amorphous selenium, a-Se, was one of the most widely used amorphous semiconductors. It has been used as a photoconductor in photocopiers and also in x-ray imaging technique known as xero-radiography for decades. Recently it is being considered for digital x-ray imaging [1]. Its electrical properties, especially its low dark current, render it suitable for x-ray imaging use. However, until now many of its properties have not been fully explored or understood. Its low glass transition temperature $T_g$ of about 40 °C and the care this necessitates in collecting experimental data have, undoubtedly, played a role in this matter [2, 3]. We, therefore, decided to study some of the unresolved issues concerning the a-Se properties [4]. In the course of that work, we noticed some very unusual temperature dependence of the DC conductivity in our samples. We have since carried out further series of DC conductivity measurements on numerous a-Se bulk and film samples, and report our observations in this contribution.

## 2. Experimental details

Amorphous selenium films of varying thickness in the micrometre range were evaporated onto Corning 7059 glass substrates from 99.995% pure Se (Alfa Aesar) and from stabilised Se (a-Se with 0.2–0.5% As and a few ppm of Cl, provided by Kasap, University of Saskatchewan). It was found that alloying a-Se with small amounts of As increases the glass transition temperature $T_g$

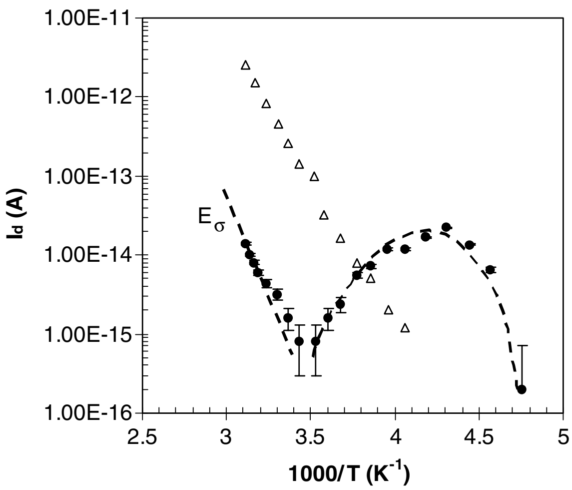


**Figure 1.** The temperature dependence of the dark current $I_d$ from an a-Se film (●) deposited on a Corning 7059 substrate, and from a standard microscope slide with the same electrode geometry (△).

and prevents crystallisation of a-Se over time [3]. For electrical measurements, gold electrodes were evaporated in a gap configuration onto the top of the films and on some bulk Se glasses. Typical gap cell dimensions were $0.5 \times 10$ mm$^2$. The dark currents were monitored over long times by connecting the output of a Keithley current amplifier to a pen recorder. The current–voltage characteristic was measured to assure Ohmic contact behaviour. DC conductivity measurements were made in vacuum at 50 V applied across the gap cell, and in the range of 208–322 K.

## 3. Results

Steady-state dark conductivity measurements were performed on many pure and stabilised a-Se bulk and film samples. Figure 1 shows a representative data set for the steady-state dark current measured as a function of temperature in pure and stabilised a-Se films (full symbols in figure 1). On pure Se films, measurements above 313 K ($10^3/T < 3.2$) were taken during short excursions above $T_g$; no subsequent evidence of crystallisation was seen. For temperatures above 285 K (i.e., $10^3/T < 3.5$), the a-Se data show the expected thermally activated behaviour, as indicated by the more or less constant slope of the log $I_d$ versus $1/T$ plot. As an average slope over all measured samples, we found $E_\sigma = 1.05 \pm 0.08$ eV, which corresponds well with literature values for this quantity as well as with the notion that this activation energy—indicating the position of the Fermi level to first approximation—should be roughly half the optical gap [5]. An unusual departure from standard semiconductor behaviour is observed for temperatures below 285 K: the current in the a-Se films actually increases with decreasing temperature to show a maximum near 240 K. To make sure that our equipment was still measuring the intended low-level a-Se currents, and not some spurious leakage, we duplicated our sample configuration on a standard microscope slide and measured its low-temperature conductivity. The open triangles in figure 1 represent those data. Only the expected featureless decrease of the current with decreasing temperature is seen. The 'anomalous' peak in the temperature dependence of the dark current that we observed in all measured a-Se

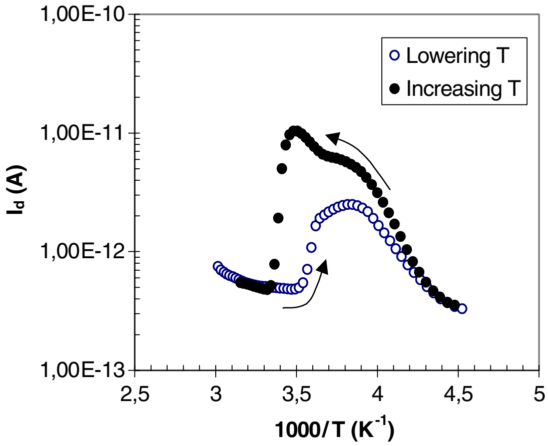


**Figure 2.** The temperature dependence of the DC dark conductivity from a pure a-Se film sample upon continuous cooling (open circles) and subsequent heating (full circles) at a rate of 0.04 K $s^{-1}$. (This figure is in colour only in the electronic version)

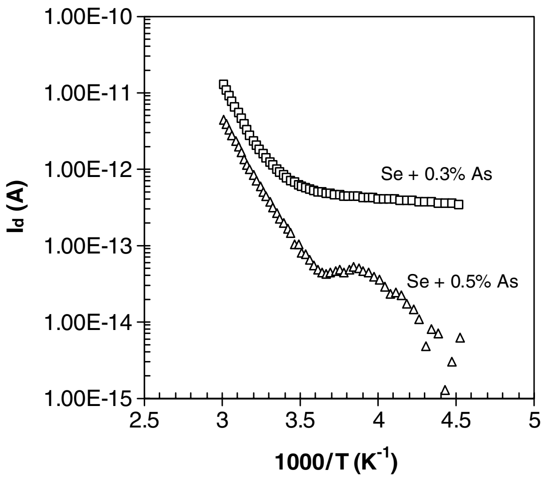


**Figure 3.** The temperature dependence of the dark current $I_d$ from bulk samples of a-Se + 0.3% As (□) and a-Se + 0.5% As (△).

films below 285 K does show temperature hysteresis when examined in successive cooling and heating cycles. Figure 2 shows one such cycle for a continuous temperature change of 0.04 K $s^{-1}$.

The observed anomalous dark conductivity behaviour is much weaker, but not fully absent, in the bulk samples. Figure 3 shows the currents for two different selenium samples measured under continuous cooling of the sample at a rate of 0.04 K $s^{-1}$. In both cases the dark current deviates from the thermally activated process at temperatures below 278 K. While the current through the sample with 0.5% As mimics the current maximum seen in the films at the corresponding temperatures, for the a-Se + 0.3% As sample it looks more like just a change of slope. When this last set of data is replotted in a $\ln(I_d T^{1/2})$ versus $T^{-1/4}$ diagram, to check for the possible occurrence of variable-range hopping, the low-temperature data lie on a straight line of the same quality as the corresponding one in the ln $I_d$ versus $T^{-1}$ diagram of figure 3.

## 4. Discussion

Activated temperature dependences like the ones seen for $1000/T < 3.5\ \mathrm{K}^{-1}$ ($T > 285$ K) in figures 1 and 3 are frequently observed in amorphous semiconductors in general, and in earlier reports on a-Se in particular. The data can be well described by the standard expression for the DC dark conductivity,

$$\sigma = \sigma_0 \exp(-E_\sigma/kT),$$

with a material-related pre-exponential factor $\sigma_0$ and an activation energy $E_\sigma$ that marks the approximate position of the Fermi level, $E_F$, with respect to the mobility edge [6]. In intrinsic semiconductors and negative-$U$ materials like the chalcogenides, $E_F$ is located near the middle of the gap (i.e. $E_F \approx E_\sigma \approx E_g/2$) [5, 6]. This agrees with our measured values of the activation energy, $1.05 \pm 0.08$ eV, which is roughly half the reported values for the optical gap of amorphous selenium [5].

Below the activated region, the temperature dependence of the dark current and the observed maximum about 240 K are quite surprising, but their appearance has been verified by numerous measurements on many different selenium films. The occurrence of such a maximum has also been confirmed by independent measurements of Benkhedir at the University of Tebessa, Algeria [7]. In the existing literature, essentially no conductivity data are available for the temperature region below 270 K.

The change of slope observed for the conductivity in the sample with 0.3% As suggests a changeover to a hopping regime, dominated by either a defect band or variable-range optimisation at $E_F$. Such a hopping process also offers a possible interpretation for the observed current maximum in terms of the energy distribution of localised states deep in the gap of a-Se that was reported by Abkowitz [8] on the basis of xerographic cycled-up residual voltage decay and xerographic dark discharge measurements. There is ample evidence that in chalcogenide materials the Fermi level is being pinned by defect states [9]. On the other hand, changes in the temperature will cause observable displacements of the Fermi level and will change the occupation of states deep in the gap (the so-called 'statistical shift of $E_F$'). If $E_F$ were to shift through one of the deep defect bands, making more states available for the hopping process, a current rise with decreasing temperature could be explained. Using the statistical shift expression $\Delta E_\sigma = \gamma T$ and the estimates that $\gamma$ will be of the order of five to ten times the Boltzmann constant as developed by Overhof and Thomas [10], a shift of some 30–60 meV can be expected over the range $3.5 < 10^3/T < 4.6$ where the current maximum is observed. Such a shift would cover a good part of any defect band. Changing the occupation of states deep in the gap will be a slow process and would account for the observed thermal hysteresis of the dark conductivity.

## 5. Conclusion

Above 285 K, the DC conductivity of a-Se is thermally activated in line with the expected pattern for a chalcogenide amorphous semiconductor. On the other hand, below 285 K a different mechanism determines the conductivity and leads to the appearance in thin films of a maximum near 240 K. The statistical shift of the Fermi level, combined with a hopping conduction mechanism in a band of deep defects, may account for the observations.

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