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EVIDENCE FOR CHARGED DEFECTS IN CHALCOGENIDES FROM HIGH FIELD CONDUCTIVITY

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ABSTRACT. The non-linear current voltage behaviour of amorphous Ge$_4$Se$_5$Te thin film layers prepared by cooling from the melt, by thermal low temperature evaporation of the bulk material and by application of a flash evaporation technique is investigated, in order to study the influence of the density of localized intrinsic defect states on the Poole Frenkel effect. The experimental results are discussed in terms of a model of screened centres in the case of the flashed layers.

INTRODUCTION
Recently, the Poole Frenkel effect was shown to be consistent with the ideas of the model of charged dangling bonds [1,2]. The non-linear current-voltage characteristics of amorphous thin films of the composition Ge$_4$Se$_5$Te prepared by thermal vacuum evaporation could be interpreted in terms of the three-dimensional Poole Frenkel theory for single centres. The density of these centres was estimated to be in the order of $10^{16}$ cm$^{-3}$. In this paper amorphous thin films of the same composition prepared by flash evaporation are investigated, in order to study the influence of the variation of the defect concentration on the current-voltage behaviour. The observed field dependent conductivity of the flashed thin films is described by the model of the Poole Frenkel effect of screened centres. The dc conductivity of thin disks of the bulk glass prepared from the melt is measured for comparison, too.

THEORETICAL REMARKS
Considering only electrons the field increased carrier density related to the Poole Frenkel effect can be described by the general equation [3]

$$\frac{n(F)}{n(0)} = \frac{2}{\pi} \int_0^{\pi} \cos \frac{\Delta E_{F}\nu(F,\phi)}{kT} \sin \nu d\nu \quad (1)$$

$\Delta E_{F,n}(F,\phi)$ is the difference between the equilibrium Fermi energy
and the quasi Fermi energy $E_{F,n}$ describing the non-equilibrium 
ocite{cabri1974} occupation enhanced by the electric field. In the case of defects 
with negative effective correlation energy (Valence Alternation 
Pairs) $\Delta E_{F,n}$ was shown to be identical with the lowering of the 
ionization energy $\Delta E(F,\varphi)$ of Poole Frenkel centres \cite{2}. $\Delta E(F,\varphi)$ 
implicitly depends also on the density of charged defects, because 
in the case of higher defect concentrations screening effects have to 
be taken into consideration. Hence, the potential energy is given by the 
equation

$$E_{scr} = -\frac{e^2}{4\pi \varepsilon \varepsilon_r} \frac{1}{r} e^{-\frac{r}{r_0}} - e r F \cos \varphi$$ \hspace{1cm} (2)

The screening length $r_0$ increases with decreasing density of charged 
defects. Assuming random distribution of the defect centres $r_0$ is 
given by

$$r_0 = \left( \frac{kT \varepsilon \varepsilon_r}{e^2 D^+} \right)^{\frac{1}{2}}$$ \hspace{1cm} (3)

For isolated centres one obtains

$$E_{isol} = -\frac{e^2}{4\pi \varepsilon \varepsilon_r} \frac{1}{r} - e r F \cos \varphi$$ \hspace{1cm} (4)

In the case of screened centres $\Delta E(F,\varphi)$ is determined from equation (2) by

$$\Delta E_{scr}(F,\varphi) = -\left( \frac{r_m r_0}{r_m + r_0} - r_m \right) e r F \cos \varphi$$ \hspace{1cm} (5)

with $r_m(F,\varphi)$ being the coordinate of the maximum value of equation 
(2) estimated numerically. For isolated centres $\Delta E(F,\varphi)$ is usually 
determined by

$$\Delta E_{isol}(F,\varphi) = \beta F^{\frac{7}{2}} \cos^{\frac{7}{2}} \varphi \hspace{1cm} \beta = \sqrt{\frac{e^3}{4\pi \varepsilon \varepsilon_r}}$$ \hspace{1cm} (6)

Combining (1) and (6) one obtains equation

$$\frac{n(F)}{n(0)} = \frac{2}{\alpha^2} \left( 1 + \alpha \sinh \alpha - \cosh \alpha \right), \hspace{0.5cm} \alpha = \frac{\beta F^{\frac{7}{2}}}{kT}$$ \hspace{1cm} (7)

which is identical with the expression for the Poole Frenkel effect 
deduced by Connell et al. \cite{4}. In the case of screened centres the 
field dependent carrier density is calculated from equation (1) and 
(5) by numerical integration. Fig. 1 shows the influence of the 
screening length $r_0$ on the field enhanced carrier concentration. 
Provided the mobility is independent of the field strength a field 
dependent conductivity should be observed by changing the density of
charged intrinsic defects (VAPs) in a sufficiently large range for a suitable material as predicted above (fig. 1).

EXPERIMENTAL

Samples of the bulk glass of the composition Ge₄Se₂₅Te were prepared by cutting and polishing the material up to a thickness of about 0.1 mm. Because of the chemically ordered network structure of such a glass formed from the thermal equilibrium of the melt the intrinsic disorder should be lower than in the case of thin amorphous layers prepared by vacuum evaporation which was already described [5]. In the flash evaporation technique small particles of the glass were successively evaporated at 1000 °C and at a pressure of 1.3 · 10⁻³ Pa: substrate temperature 8 - 12 °C, deposition rate 2.5 nm · s⁻¹. Before evaporating the glass a bottom contact was deposited upon the substrate. The sandwich arrangement was completed by deposition of a top contact without interruption of the vacuum process. The composition of the layers was proved by chemical analyses. All of the samples showed the identical composition Ge₄Se₂₅Te. They were free from concentration gradients [6].

RESULTS AND DISCUSSION

The field dependent conductivity of the amorphous Ge₄Se₂₅Te layers prepared by different methods is shown in fig. 2. The experimental data of the bulk material and of the films obtained by the thermal evaporation technique at lower temperatures are excellently described by the theoretical curve derived from the model of the Poole Frenkel effect of isolated centres corresponding to equation (7). At room temperature and at higher temperatures the observed curves of the field dependent conductivity of the flashed thin films are in accordance with the model of the Poole Frenkel effect of screened centres (equations 1 and 5) assuming a screening length r of about 2.5 nm. From equation (3) a density D₄ of 2 - 8 · 10¹⁶ cm⁻³ is calculated in the limits of an estimated temperature interval of 280 - 1270 K for the freezing point in the formation of intrinsic defect centres in the deposition process of the layers by flash evaporation. The higher defect concentration is an intrinsic effect of the amorphous material caused by the preparation method and cannot
be attributed to foreign additives incorporated during the film processing as in the case of sputtering or CVJ). The experimental results are in accordance with the model of VAPs within the limits of a model of screened Poole-Frenkel active centres. Consequently, the intrinsic defects are charged. Independently defect states doubly occupied and unoccupied result from the absence of paramagnetism in such an amorphous material. The Poole-Frenkel active behaviour of such centres requires imperatively the assumption of double excitation. In such a process the $D^-$-centres $T^-$ or $C_1^-$ are transformed into $D^+$ centres $C_1^+$ and reversed as shown in Fig. 3.

Fig. 3 Intraconversion of charged defect states $C_1^+$ and $T_1^-$ or $C_1^-$ respectively, by double excitation

REFERENCES