A.c. conduction in evaporated MoO$_3$/SiO amorphous thin films

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The dielectric properties of vacuum-deposited MoO$_3$/SiO films of different compositions studied in the frequency range $10^2$ to $10^6$ Hz at various temperatures (193 to 393 K) are reported. The properties of the film capacitor are found to be temperature and frequency dependent. The decrease in a.c. conductance with increasing concentration of SiO in MoO$_3$ may be attributed to the increasing number of trapping centres generated in MoO$_3$/SiO films during the evaporation process.

1. Introduction

A.c. measurements are an important means for studying the dynamic properties (capacitance, conductance, permittivity and loss factor) of a dielectric. The advantage of a.c. measurements over the d.c. measurements is that internal time-dependent processes in the insulator can be investigated with a.c. measurements. Moreover, the a.c. voltage bias need never exceed a few hundred millivolts. Thus the maximum field within the insulator film is kept to a minimum and there is little danger of more than one conduction process being active. This measurement also helps to distinguish between localized and free-band conduction. In the case of localized conduction, the conductivity, $\sigma_{ac}$, increases monotonically with radial frequency, $\omega$, whilst for the free-band conduction the conductivity decreases with frequency. A number of investigations has been carried out on the frequency dependence of electrical conductivity in many glassy and amorphous semiconductors [1–9]. It has been pointed out by Elliott [1] that a variety of conduction mechanisms can yield the $\omega^s$ behaviour for the a.c. conductivity, but in general, it is difficult to establish which of the above effects determines a given observed conduction process. A careful study of the temperature dependence of $s$ (s is an index) can provide us with more information in order to make a choice between the different theories for explaining the law $\sigma_{ac}(\omega) \propto \omega^s$.

The experimental results can be interpreted in terms of a model initially proposed by Pollak and Geballe [10]. This model involves a thermally assisted hopping conduction mechanism between localized states. Pike [4] has analysed classical hopping over the barrier separating the localized states in such materials.

In this study, the a.c. electrical properties of mixed MoO$_3$/SiO amorphous films are reported. There is very little information available about MoO$_3$ because of the considerable difficulties in preparing specimens suitable for the investigation of physical properties. Nadkarni and Simmons [11, 12] reported that the existence of Schottky barriers is believed to be due to a strong donor band in the insulator established during the vacuum evaporation. SiO dielectric films are of considerable interest because of their use in the electronics industry as capacitor materials. Simmons [13] reported that both donor and trapping centres are created in SiO films due to the dissociation of SiO into SiO$_2$ and free silicon. Furthermore, the SiO structure contains active defects in both structure and composition. These defects are associated with oxygen deficiencies in the film giving rise to vacancy states and unstable structural defects, i.e. interstitial oxygen. Jourdain et al. [9] reported that their a.c. results on SiO cannot be analysed in terms of a quantum hopping conduction mechanism proposed by Pollak and Geballe [10], but the results seem to be in agreement with the theory derived by Pike [4]. The height of the barrier is lowered by the overlapping of the Coulomb potentials at the two sites. They further reported that the values of activation energy associated with the localized states and calculated from the Pike model agree with the d.c. activation energy as measured.

According to Kondo et al. [14], the a.c. conduction in the radio frequency range depends on frequency and temperature in amorphous chalcogenide semiconductors ($\sigma(\omega, T) = A\omega^s$ where A and s are temperature-dependent parameters). They interpreted their results in terms of a combined mechanism of correlated barrier hopping (CBH) of bipolarons (two electrons hopping between charged defects $D^+_1$, $D^-_1$) and single polarons (one electron hopping between a neutral defect $D_0$ and a charged defect $D^+_1$ and one hole hopping between $D_0$ and $D^-_1$). They further reported that intrinsic defects similar to those in amorphous SiO$_2$ are believed to exist and contribute to a.c. conduction in amorphous SiO films. No earlier work on mixed thin amorphous films of MoO$_3$/SiO is available in the literature, apart from recent work performed in this laboratory. In this work we have investigated the a.c. electrical properties of mixed MoO$_3$/SiO films as a function of composition. The effects of temperature on the conductance and capacitance are also reported.
2. Experimental procedure
Thin amorphous films of the mixed oxide system MoO$_3$/SiO$_2$ were fabricated in a Balzers BA510 coating unit by deposition on to clean Corning 7059 glass substrates in the form of metal–oxide–metal sandwich electrode structures at a pressure of about 6 × 10$^{-6}$ torr using a co-evaporation technique. All other techniques used to measure the film thickness, cleaning of the substrates and d.c. measurements are similar to those described earlier [15].

3. Results
A large number of films of different compositions were measured as functions of frequency and temperature. The measurements were carried out in the frequency range 10$^2$ to 10$^6$ Hz and in the temperature range 193 to 393 K. The experimental results are usually interpreted in terms of a model initially proposed by Pollak and Geballe [10], modified by Pike [4] and subsequently modified by Elliott [1]. This model involves a hopping conduction by electrons (thermally assisted) between localized states. Pike pointed out that classical hopping over the barrier separating the localized states can sometimes occur.

It is well known that the total measured a.c. conductivity, $\sigma_{a.c.}$, may be represented in a wide variety of amorphous semiconductors and insulators by the experimental relation

$$\sigma_{a.c.} = \sigma_{total} - \sigma_{d.c.}$$

where $\sigma_{d.c.}$ is the d.c. conductivity and $\sigma_{a.c.}$ is the true a.c. conductivity which is frequency, $f$, dependent. Fig. 1 shows the frequency ($f$) dependence of a.c. conductivity for 300 nm thick MoO$_3$/SiO$_2$ samples of various compositions and Fig. 2 illustrates the frequency dependence of a.c. conductivity for a typical specimen (70 mol % MoO$_3$/30 mol % SiO$_2$) at various temperatures. At high frequencies the curves approximate to a square law dependence on frequency and show relatively little dependence on temperature. At low frequencies the log $\sigma$–log $f$ graphs are good straight lines and are temperature sensitive. One can find a frequency range (depending on temperature), where the a.c. conductivity, $\sigma_{a.c.}$ obeys the law

$$\sigma_{a.c.} \propto \omega^s$$

where $s$ is an index which is temperature dependent and which tends to unity [1] as the temperature is lowered. For the sample shown in Fig. 2, $s$ decreases from 0.83 to 0.39 (as shown in Fig. 3) when the temperature increases from 193 to 393 K. Furthermore, the variation of a.c. conductivity with temperature can be expressed by the relation,

$$\sigma_{a.c.} = A(T)\omega^s(T)$$

where $A$ is a complex parameter weakly temperature dependent.