ELECTRONIC PROPERTIES OF AMORPHOUS SEMICONDUCTORS

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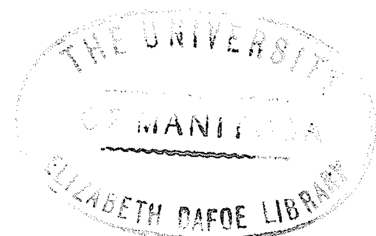

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ABSTRACT

The electrical switching and memory properties of the amorphous composition $Te_{48} As_{30} Si_{12} Ge_{10}$ have been investigated, and emphasis has been placed on the initial switching behaviour of new devices, the durability of devices when subjected to continuous operation, and the energy required to obtain the memory state. It has been found that the degree of stability in the switching threshold voltage, both initially and after many thousands of switching cycles, depends on the operating current levels, switching involving higher current being less stable than that involving a smaller current. It has also been found that the durability of the devices which were investigated depends on operating current levels; operating currents of 1 mA resulted in stable switching for more than $5X10^4$ switching cycles, whereas operating currents of 18 mA resulted in rapid deterioration after only several switching cycles. The transition from threshold switching without memory, to threshold switching with memory, depends mainly on the energy input to the device, and for the devices used in the present investigation this energy is about 0.03 Joules. All of the results can be explained on the basis that the transition which occurs in conductivity for both ordinary switching and memory switching is due to the formation of conductive filaments, and that their formation is preceded by a diffusion of certain constituent atoms away from the filament region and other atoms towards the region, so that the final composition of atoms along the filamentary path is more favourable to structural changes towards a more crystalline structure.

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LIST OF MOST USED SYMBOLS

A	activation energy for electrical conduction (expressed in temperature units)
a	interatomic separation
C	heat capacity per unit volume
D	carrier diffusion constant
d	thickness of active material (electrode separation)
E_c	energy at the conduction band edge
E_v	energy at the valence band edge
E_F	energy at the Fermi level
E_g	band gap energy
E_h	average activation energy required for hopping
E_p	energy of polarization near a localized state of order
E_{K_i}	energy of state K_i
ΔE	activation energy for electrical conduction
F_{cr}	critical electric field required for switching
f	jump frequency between two states
$f(E)$	Fermi-Dirac function
I_H	holding current required to maintain the high conductivity state
K_B	Boltzmann constant
K_c	degree of compensation
ℓ	current distubution factor (ratio of filament current to total current)
m_n	effective electron mass

N_m	concentration of majority carriers
N_τ	total trap density
N_τ'	fraction of charged traps
$N(E)$	density of states at energy level E
P_h	hopping probability per unit time
$p(R)$	tunnel factor
q	electronic charge
R	mean separation between trapping centers
R_T	thermal resistance
R_{pp}	average rate of electron-hole production
r	hydrogenic radius of a state
T_e	electron temperature
T_{cr}	critical temperature reached during switching
ΔT	incremental temperature changes
t_D	switching delay time
t_R	switching recovery time
V_c	conduction voltage
V_o	average random potential for state localization
V_{TH}	switching threshold voltage
v_d	carrier drift velocity
v_τ	thermal velocity of carriers
ε	dielectric constant
ε_o	dielectric constant of free space
λ	mean free path of carriers
σ	electrical conductivity

σ_o	pre-expoential conductivity factor
σ_T	thermal conductivity
ν_σ	frequency factor for potential fluctuation
ν_{ph}	phonon frequency
ν_{el}	electron frequency
τ_{pp}	time constant for electron-hole pair production
θ	temperature of the switching filament
ρ_T	thermal resistance of switching filament
μ	carrier mobility

CHAPTER 1

INTRODUCTION

The properties of crystalline semiconductors have been extensively investigated in the past two decades. Quantum theory, supported by experiments, has resulted in the understanding of conduction processes and many other important phenomena in crystals. The development of the transistor, for example, is the result of the research in the field of crystalline semiconductors. Until recently, solid state physics and electronics have been primarily concerned with the properties of materials having crystalline structure (i.e. with long range order and periodicity). Non-crystalline materials, or more specifically amorphous semiconductors however, have no long range order (i.e. no regular periodicity) although local short range order does exist. This means that each atom sees its neighbouring atoms in much the same relative positions as in a crystal.

Several aspects of amorphous semiconductors may be considered as characteristic of the disordered phase. For example, anomalous Hall effects, temperature activated drift mobilities, frequency dependence of conductivities, and electric field dependence of quantum efficiencies, to mention but a few properties, have all been widely reported in the literature (Male 1967, Mott 1967, Cohen 1968). Also, several investigators have proposed a number of models and theories in an attempt to explain the observed facts (Anderson 1958, Mott 1967, Böer 1970, Cohen 1970). It has been postulated that conduction in amorphous semiconductors is due to the coexistence of two basic mechanisms-band conduction and conduction due to hopping,

and that in the amorphous phase a material may have a continuous range of energy states in which all of the wave functions are localized.

One of the important factors which has stimulated the present interest in amorphous materials is the apparent inability of the present theory of solid state physics to explain the optical, electrical, and thermal properties of these materials. The concept of energy bands and Fermi surface, which have been formulated and developed based on the periodic nature of the crystal lattice, cannot be applied directly to amorphous materials. Another factor which has resulted in a great deal of research in this field is the potentiality of amorphous semiconductors for switching and memory devices. The switching phenomenon in these materials has been reported by many investigators over the past decade. Pearson (1962) has reported switching amd memory effects in the As-Te-I system. Kolomiets and Lebedev (1963) has observed the switching phenomenon in $TlAs(Se, Te)_2$, and Simmons and Verberder (1967) have reported that amorphous silicon monoxide could be used for memory switches. Switching in amorphous films of various semiconducting materials has also been observed by Chopra (1965). Some details of the Ovonic switch based on evaporated films of a mixture of tellurium, arsenic, germanium, and silicon have been reported by several investigations (Ovshinsky 1968, Mott and Davis 1972).

The most important elements in amorphous semiconductors and those which have been extensively investigated are Si and Ge in Group IV; P, As, Sb, and Bi in Group V; and S, Se, and Te in Group VI. The methods

on preparation include: evaporation and condensation on a cold substrate, cathode sputtering, exposing crystalline materials to very high doses of high energy radiation, rapid quenching techniques, anodic oxidation, and, in some cases, cooling the material from the melt. The two most commonly used procedures are: (1) the formation of films - by evaporation and condensation onto a cold substrate, by cathode sputtering or other techniques, and (2) by rapid cooling of the melt. The variety of materials and methods of preparation has made it possible to obtain a wide range of configurational structures ranging from the quasi-crystalline state to the highly disordered state. The structural properties of these materials depends greatly on the specific elements used, and on the method of preparation.

In the investigation being reported in this thesis the covalent alloy Te_{48} As_{30} Si_{12} Ge_{10} was used. All of the devices used had an active material thickness of 100 μm between two vacuum evaporated electrodes. The specific properties to be studied are: (1) the initial switching characteristics of new devices under various operating currents and switching rates, (2) the characteristics and lifetime of devices when subjected to continuous switching over a long period of time, (3) the effect of operating current on the durability, and reliability of switching devices, (4) the memory behaviour and the conditions required to obtain and to erase memory, and (5) the variations of switching threshold voltage, switching time, and holding current with temperature. The primary purpose of the investigation is to gain a better understanding of the actual mechanisms responsible for the switching and memory phenomena in amorphous materials, and to determine some guidelines with respect to

the practical applications of devices made of amorphous semiconductors.

Chapter 2 of this thesis gives a brief systematic review of previous theoretical and experimental work. Chapter 3 presents several mathematical models for elucidating the properties of amorphous semiconductors with emphasis on models for switching and memory behaviour. The experimental procedures and circuits are described in detail in Chapter 4, and the results and discussion are given in Chapter 5. Tentative conclusions arising from this investigation are given in Chapter 6.

CHAPTER 2

REVIEW OF PREVIOUS WORK ON AMORPHOUS SEMICONDUCTORS

The first part of this chapter will deal with several of the more fundamental and widely accepted explanations and models for the basic phenomena observed in amorphous semiconductors including the energy band model for these materials and a review of several mechanisms responsible for conduction, switching, and memory behaviour. The second part of this chapter will be devoted to a review of experimental work with particular emphasis on the Te-As-Si-Ge system. Throughout the present chapter a strong emphasis will be given on the correlation between the theoretical and experimental results. It should be noted, however, that although some models have been highly successful in elucidating certain isolated observations, there is no unified theoretical approach capable of explaining all experimental results.

2.1 ELECTRONIC STATES AND ENERGY BANDS

The simplest possible model for an amorphous semiconductor is shown in Fig. 2.1 (a). There is a valence band with a tail of localized states above E_V and a conduction band with a tail of localized states below E_C . Thus only a pseudogap exists between the valence and conduction bands. The simple model presented in Fig. 2.1 (a) is probably only adaquate for elemental and compound amorphous semiconductors. Though these materials are transitionally disordered, the short range order is in general so well defined that only structural defects can occur. Examples of such defects are broken bonds in a covalently bonded network, nonbridging atoms, and chain ends. Being structurally well defined,

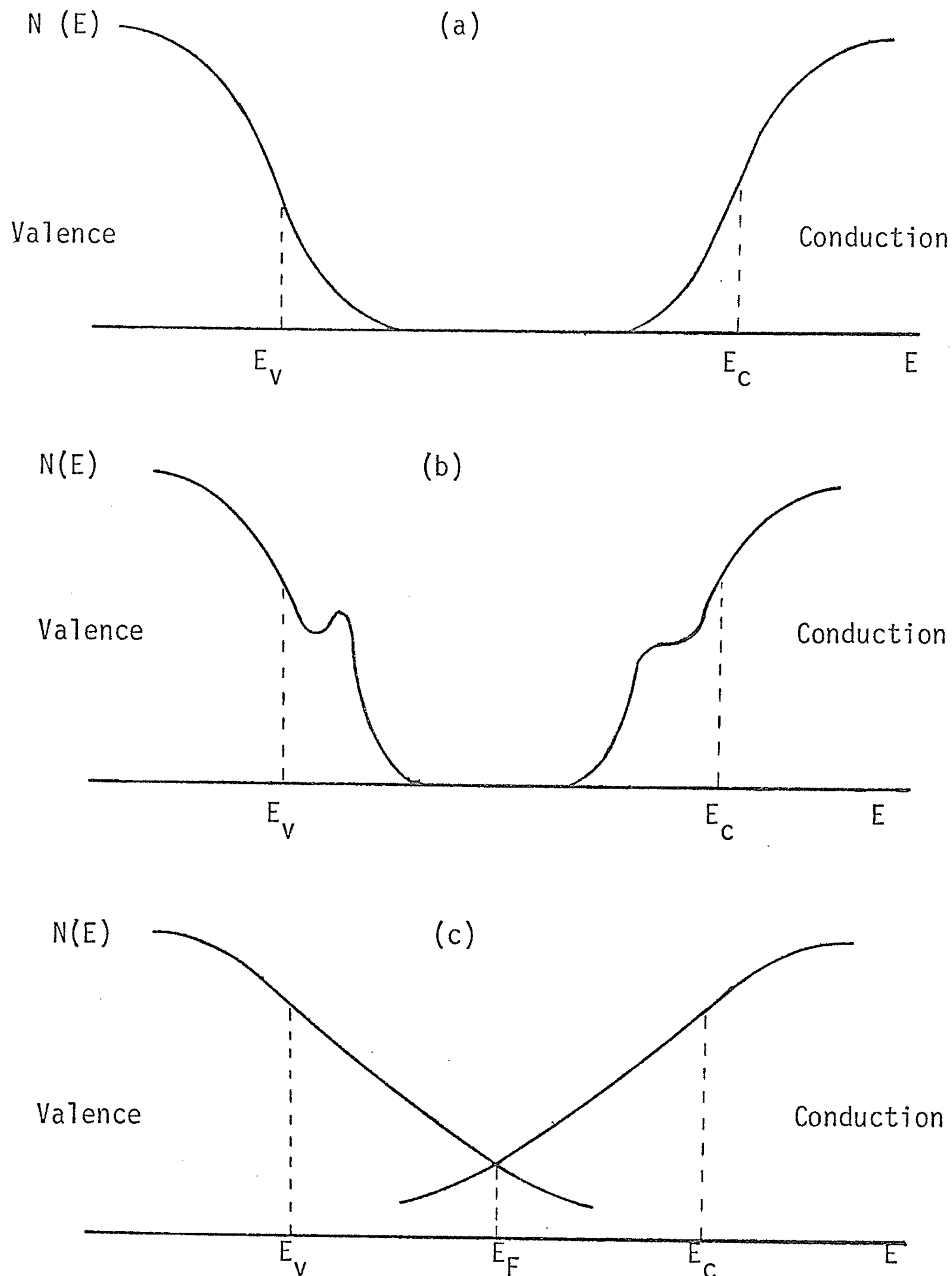

Fig. 2.1 Band models for amorphous semiconductors.
(a) Valence and conduction bands with extended states for $E>E_c$ and tails of localized states on each band.
(b) Local order leads to the presence of well defined structural defects and nonmonotonoicity in N(E) in or near the tails.
(c) The tails of the conduction and valence bands overlap, leading to a finite density of states at the Fermi energy and a finite concentration of localized charged states near E_F (after Cohen 1970).

these defects can be expected to have well defined energy levels associated with them. These would not be sharp as in a crystal but would vary somewhat with local environment giving rise to peaks in the density of states as illustrated in Fig. 2.1 (b).

Neither of Fig. 2.1 (a) and (b) is adaquate for concentrated amorphous covalent alloys such as those based on the chalcogenides Se and Te. Such alloys contain atoms of varying valences in large concentrations. The original work by Anderson (1958), and Mott (1967), has led Cohen et al. (1970) to formulate the CFO (Cohen-Fritzsche-Ovshinsky) model which has been quite successful for the covalent amorphous semiconducting alloys. The CFO model is based on four signicicant postulations: 1) amorphous semiconducting alloys with widely varying valences behave as intrinsic materials because the individual atoms have their valence requirements satisfied locally, i.e., all valence bonds are saturated, 2) these alloys are characterized by valence and conduction bands of extended states as shown in Fig. 2.1 (c), 3) both bands of extended states have tails of localized states, and 4) there exists a well defined energy, E_V or E_C , in each band at which a transition from extended to localized states occurs. The CFO model incorporated with these four points formulates four additional features. 5) In amorphous semiconducting alloys the disorder is sufficiently great that the tails of the conduction and valence bands overlap. Because bonding conditions and hence valence requirements are satisfied locally, the connectivity of the atomic structure varies randomly throughout the material. There are not merely the typical translational disorder of an amorphous material and the typical compositional disorder of an alloy, but also an enhanced

translational disorder because of the additional randomness of the structural network itself, forced by variations in its connectivity from site to site. 6) The localized states which come off the valence band and those which come off the conduction band are always identifiable as either valence band states or conduction band states even when their energy levels lie in the region of overlap. 7) There exists a continuous distribution of charged traps in the middle of the band gap. That is, valence band states are locally neutral when occupied. Therefore, an empty valence tail state above the Fermi level contributes a localized positive charge. Conversely, conduction band states are locally neutral when empty, and an occupied conduction tail state contributes a localized negative charge. 8) In the transition from extended states to localized states the transport processes change their character. It is speculated that above E_c in the conduction band, charge transport is achieved primarily through band conduction mechanisms, whereas below E_c transport occurs by phonon-assisted hopping between the localized states. A similar process also occurs in the valence band.

The eight features of the CFO model have been used to explain several experimental results with varying degrees of success. For example, this model has been used to explain the existence of a well defined activation energy in the conduction process i.e. , the apparent intrinsic character of the conductivity (Cohen et. al 1969). This model may also give some insight into the relationship between photoconductivity and frequency of incident radiation, and quantum efficiencies, observed by Fagen and Fritzsche (1970). Feature (8) of this model may explain the difference in the values of mobility caused by the various conduction

mechanisms and may therefore help to clarify the anomalous behaviour of the Hall effect reported in the literature (Pearson 1963, Kolomiets and Nazarowa 1960, Male 1967, Peck and Dewald 1964).

2.2 MECHANISMS OF ELECTRIC CONDUCTION

The electrical conductivities of many amorphous semiconductors, particularly bulk chalcogenide glasses produced by cooling a melt, follow the relation

$$\sigma = \sigma_0 \exp(-\Delta E/K_B T) \quad , \qquad (2.1)$$

where σ_0 is the pre-exponential conductivity factor and is of the order $10^3 - 10^4\ \Omega^{-1}\ cm^{-1}$, and ΔE is the activation energy of the electrical conduction which is usually 0.2 eV larger than half the band gap of the material. A typical plot of the temperature dependence of conductivity for several amorphous semiconductors is given in Fig. 2.2. The insensitivity of these materials to doping suggests that the conduction is intrinsic so that the slope of these curves is proportional to one half of an energy gap. It seems as though the presence of short range order is sufficient to ensure that the electronic density of states in the amorphous phase is not significantly different from that of the crystalline phase, and that the concept of a forbidden gap, with an energy equal to that for breaking covalent bonds between atoms, is acceptable. It is reasonable to assume, on the basis of other properties of amorphous semiconductors, that the electronic states at the band edges are perturbed on going from the crystalline to the amorphous phase and that the density

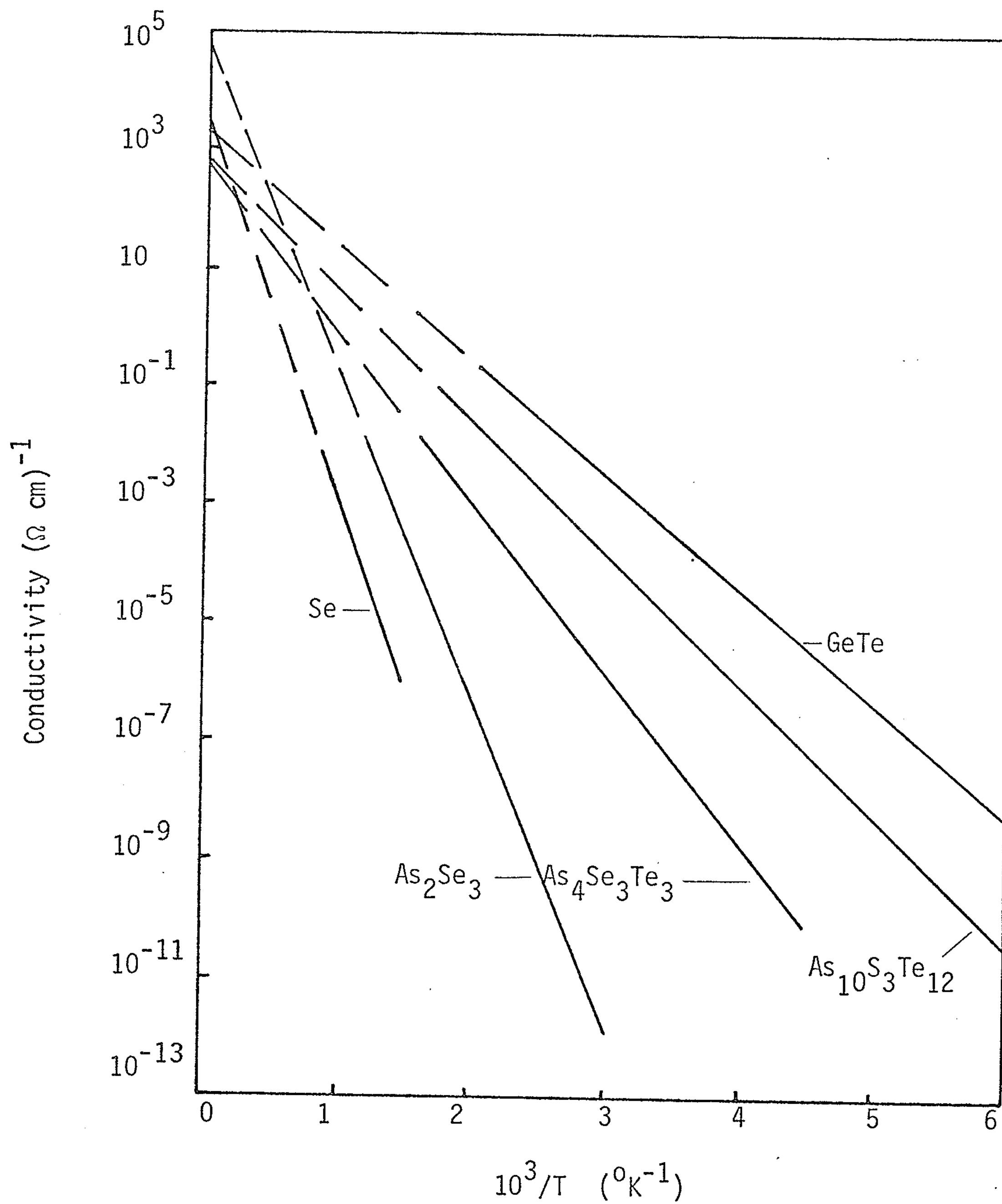

Fig. 2.2. Temperature variation of conductivity of a few amorphous semiconductors. (after Davis and Shaw 1970).

of states is finite in the energy gap (Banyai 1964, Mott 1967, Fagan and Fritzsche 1970). Fig. 2.3 shows schematically the tailing of the states into the gap to a degree in which valence states and conduction states overlap near the center of the gap. As mentioned previously, the states in these tails arise from structural defects, and large variations in density and composition throughout the material. The levels are distributed over a sufficiently wide energy range to satisfy the Anderson criterion for state localization (Anderson 1958, Anderson 1961). The Anderson criterion is

$$V_o > 10 z I^o \exp(-\alpha R) , \quad (2.2)$$

where V_o is the average random potential, z is the coordination number, I^o represents an overlap integral, R is the mean separation between centers, and α gives a measure of the extent of the wave function on isolated centers. In addition to these tails of localized states, the absence of long range order will, if the wave functions on the atoms are not s - like, lead to the localization of states at the band extremities up to well defined energies A and B where the density of states is perhaps equal to about 20% of the effective density of states associated with the band.

Because the states between A and B are localized, conduction can occur in these energy states only by a thermally activated tunneling process similar to that occuring between centers in a heavily doped semiconductor and commonly referred to as impurity conduction. The mobility associated with this process is generally quite small and may be expressed in the form

$$\mu = \frac{q \nu_{Ph} R^2 e^{-2\alpha R} e^{-E_h/K_B T}}{K_B T} , \quad (2.3)$$

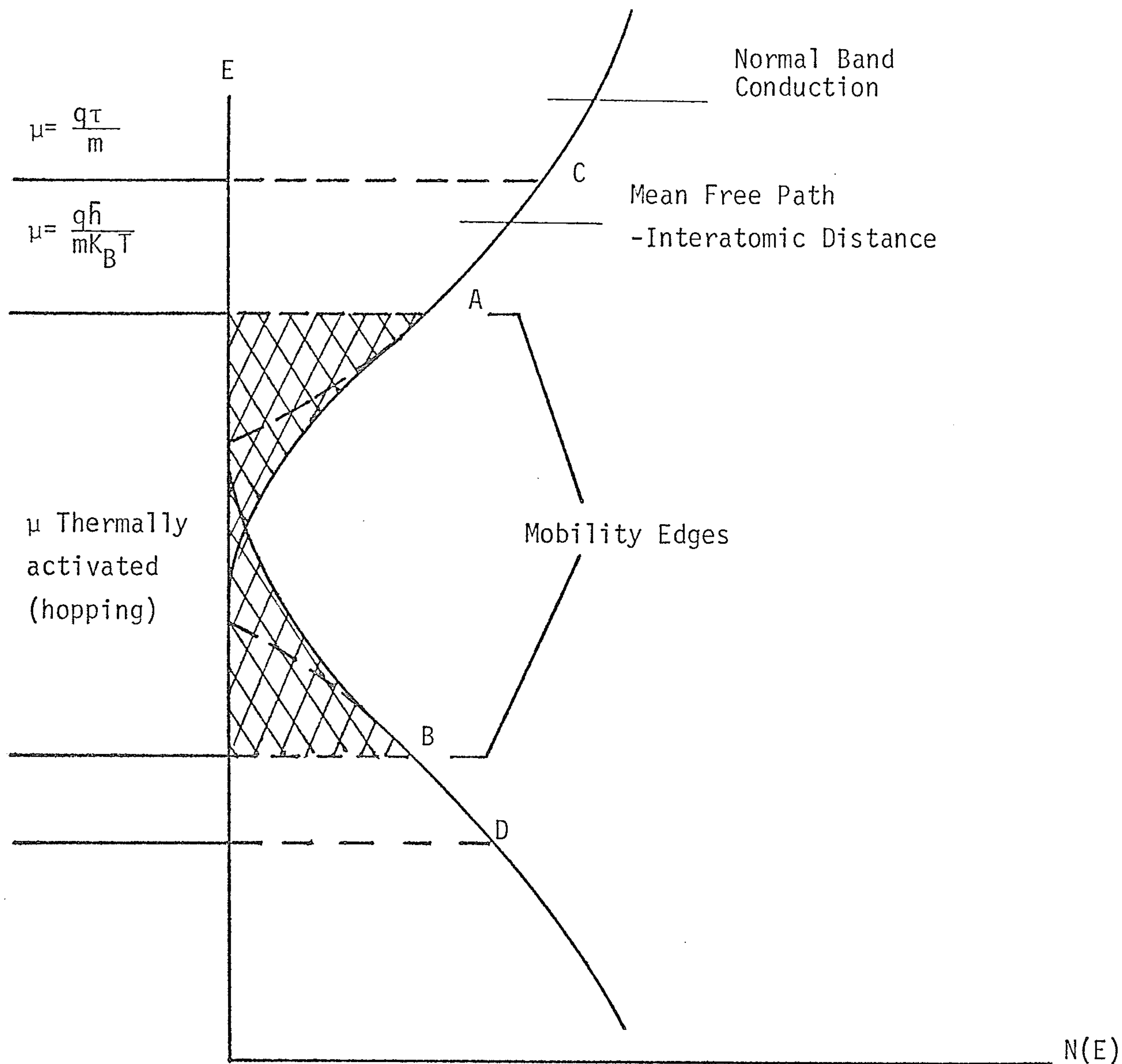

Fig. 2.3. Schematic representation of the density of states in an amorphous semiconductor, showing the regions (shaded) in which the electronic states are localized and the regions in which the carriers move with short mean free path. (after Davis and Shaw 1970).

where V_{Ph} is a phonon frequency, and E_h is the average activation energy required for hopping. For amorphous materials a quantitative calculation of μ is quite difficult. Near A and B, however, E_h is expected to be small and the tunneling factor exp $(-2\alpha R)$ approaches unity. Under these conditions the hopping mobility has its maximum value, typically $\sim 0.1\ cm^2/V$ sec.

Immediately beyond A and B where the states are delocalized the carriers can move without thermal activation. In the region of delocalized states however, the mean free path is expected to be of the order of the interatomic separation and the appropriate expression for the mobility becomes

$$\mu = \frac{qa^2}{K_B T} \nu_{el} \quad . \tag{2.4}$$

ν_{el} is the electronic frequency which is equal to $\hbar / ma^2 \sim 10^{15}\ sec^{-1}$, and a is the interatomic separation. The mobility in this region is expected to be approximatly 40 cm^2/V sec. Deep into the bands, beyond C and D, band conduction with long mean free paths will occur and the normal formula for the mobility

$$\mu = \frac{q\ \tau}{m} \tag{2.5}$$

can be used.

The energy gap in a crystalline semiconductor is usually a gap between extremities in the density of states, whereas in amorphous solids it is really a gap between mobility edges A and B (Cohen et al. 1969). The conductivity curves of Fig. 2.2 therefore, represent intrinsic activation across a mobility gap AB. Extrapolation of these curves to 1/T=0 yields a value of σ_o which ranges between 10^3 and $10^4\ \Omega^{-1}\ cm^{-1}$ in most

amorphous materials, corresponding to values of mobility between 10 and 100 cm^2/V sec for a reasonable density of states. This substantiates the idea of a mean free path of the order of the inter - atomic separation.

One of the most important conduction mechanisms in amorphous materials involves the transport of carriers by a process of hopping from one localized state to another. In the evaluation of the conductivity due to hopping there are two basic points to consider. First of all it is necessary to evaluate the probability per unit time that the electron jumps from one localized state to another. Mott (1967) has formulated an expression for this probability and it is

$$P_h = \nu_{Ph}\, p\,(R) \exp\,[-(\Delta E_h + \tfrac{1}{2} E_p) / K_B T] \qquad (2.6)$$

In this expression p (R) is a tunnel factor which must be introduced if the distance R between states is large, ΔE_h is the difference in energy of the two levels, and E_p is the energy of polarization round a localized state of order. The energy of polarization is given by (Miller and Abrahams 1961)

$$E_p = \tfrac{1}{2} \frac{q^2}{r_o} \left(\frac{1}{\varepsilon_o} - \frac{1}{\varepsilon} \right) , \qquad (2.7)$$

where r_o is the radius of the center belonging to a state, and ε_o and ε are the dielectric constants of free space and the amorphous material respectively. The second point is that the tunnel factor p (R) , and the energy difference between states ΔE_h, vary greatly from one jump to another and as a result it is necessary to average the hopping probability P_h over all possible jumps in order to obtain the a c or d c conductivity. By means of this averaging process it has been shown that the a c conductivity is higher than

the d c conductivity and increases with frequency (Pollak and Geballe 1961, Pollak 1962), and that the activation energy for conduction decreases with decreasing temperature.

Very closely associated with the hopping mechanism is the phenomenon known as impurity - band conduction. In the present context impurity - band conduction will refer to the motion of an electron from one impurity center to another under the condition that the overlap between the orbitals of neighbouring centers is great enough to allow tunneling but not great enough for a transition to the metallic state. This process can only occur if compensation is present and it usually but not always involves a hopping process. The phenomenon identified as impurity - band conduction was first observed by Hung (1950) and by Hung and Gleissman (1950, 1954). Latter Conwell (1956) and Mott (1956) emphasized that this process could only occur when compensation was present. The work of Fritzsche (1958, 1959, 1960) demonstrated experimentally the role of compensation. Mott (1956) was the first to emphasize that the process involved an activation energy.

At low concentrations, the states will certainly be localized and motion will be due to the hopping process. Under this condition two cases are of major interest: 1) Very small compensation - we then have a few vacancies in n-type centers (or electrons in p-type centers) which are bound to the nearest charged minority carrier with the binding energy ΔE equal to $q^2/\varepsilon R$. The number of free carriers is proportional to exp $(-\frac{1}{2} \Delta E/K_B T)$ and ΔE is thus the activation energy of the hopping process. This case has been studied extensively by Mott (1956). 2) Moderate compensation- this is the case considered by Miller and Abrahams (1960) in which they used many different materials for their investigation.

In this case the energy difference ΔE between neighbouring centers depends on the random electric fields due to charged centers of both types. If K_c is the degree of compensation, then for small K_c Miller and Abrahams have found that the linking energy is given by the relation

$$\Delta E = (q^2/\varepsilon_0 R)\ (1-1.35\ K_c^{1/3}) \quad , \tag{2.8}$$

where R is equal to $(4\pi N_m/3)^{-1/3}$, and N_m is the concentration of majority carriers. For larger values of K_c Eqn. (2.8) becomes

$$\Delta E = 0.285\ \frac{q^2}{\varepsilon_0 R}\ , \tag{2.9}$$

when $K_c \sim \frac{1}{2}$.

The jump frequency between two states with energy difference ΔE is given by (Miller and Abrahams 1960)

$$f = \frac{1}{\tau} = 2\text{x}10^{12}\ (R/r)^{3/2}\ \exp\ (-2R/r)\ \ \tanh.\ (\Delta E/K_B T), \tag{3.10}$$

where r is the hydrogenic radius of each state. Using the averaging process Miller and Abrahams have calculated the d c conductivity and pointed out that: 1) the logarithm of the resistivity is proportional not to R, but to $R^{3/2}$; 2) the decrease in the apparent activation energy ΔE with T, which should occur in a random hopping process, does not appear in their investigation.

The Hall coefficient of impurity conduction in silicon and germanium

has been investigated in the hopping region by Amitay and Pollak (1966), but no Hall voltage was observed, and this has made it necessary to make some revisions of the averaging precedures of Holstein's (1961) theory.

Based on the averaging procedures the a c conductivity should be greater than the d c conductivity and should increase with frequency. This phenomenon has been investigated and confirmed both experimentally and theoretically by Pollak et al. (Pollak and Geballe 1961, Sewell 1963, Pollak 1964, 1965).

2.3 SWITCHING AND MEMORY BEHAVIOUR

It is well known that the majority of materials cannot withstand electric fields in excess of about 10^6 V/cm. In common insulators, fields of this magnitude invariably results in a destructive breakdown. This is, in general, a non-regenerative change of the material along the breakdown path. The amorphous semiconductors differ considerally from insulating materials because the abrupt change in electric conductivity in amorphous semiconductors can be regenerative and non-destructive (Ovshinsky 1965, Pearson 1962, Ovshinsky 1968). The physical processes which determine and sustain the conduction after the abrupt change in conductivity has taken place, may be quite different from those which initiate and lead to the transition. Before proceeding to the mechanisms leading to switching, as well as the processes taking place once the transition in conductivity has occurred, it may be desirable to classify some important types of switching characteristics.

There are basically four main current-voltage characteristics commonly

encountered in amorphous semiconductors and these are illustrated in Fig. 2.4. Fig. 2.4 (a) illustrates the negative differential resistance phenomenon. With a proper choice of the load resistor this negative differential resistance device can be kept at any point of the I-V curve. Fig. 2.4 (b) shows a typical characteristic of the amorphous semiconductor switch. Two states are possible, one of low conductivity and the other of high conductivity. The device can be switched from the low conductivity to the high conductivity state when the applied voltage reaches a characteristic value, termed the threshold voltage V_{TH}. The high conductivity state remains as long as the current does not fall below a typical limit known as the holding current I_H. This does not imply, however, that V_{TH} and I_H are the only parameters determining the switching behaviour. Fig. 2.4 (c) illustrates the negative differential resistance with memory characteristic of a typical device. The negative differential resistance device with memory has two stable states. The first state resembles that of Fig. 2.4 (a). The second state is conductive, which is established at higher currents and remains without decay. The first state can be re-established by increasing the current above a certain value and by switching it off rapidly. The final I-V characteristics observed is shown in Fig. 2.4 (d). This characteristic represents switching with memory. This device is normally in its low conductivity or blocking state for voltages less than the threshold value. The current increase monotonically with applied voltage and this I-V relation depends on the material used and on the mode of operation (a c or d c). When the voltage exceeds the threshold value a transition occurs from the low conductivity state to the high conductivity state. The high conductivity state may be maintained for a period of months even though the applied