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The carrier yield in a-Se under electron bombardment

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Abstract. We describe measurements of the field and dose dependence of the carrier yield produced in a-Se by electron beam excitation, using a flight-time technique. The results show that the yield is controlled by stronger initial recombination than that predicted by the Onsager mechanism which governs the photoelectric yield. This strong process is attributed to bimolecular recombination along the tracks of the primary electrons (columnar recombination). On this interpretation, the effective average pair-production energy is ~18 eV. We speculate on the origin of this high value.

1. Introduction

In the course of a study of the drift mobility and its field dependence in films of pure and doped amorphous (a-) Se, using the standard flight-time technique with electron beam excitation, some measurements of carrier generation efficiency were also made (Jahankhani 1987). These suggested that the carrier yield was far below saturation even at an extraction field F of 300 kV cm^{-1}, and could not be reconciled with the well documented quantum efficiency of photo-generation. Since there was some doubt about the quality of the samples, the measurements have since been repeated and extended. Their results, reported in this paper, reinforce the earlier conclusions.

Definitive measurements of the quantum efficiency η of photo-generation in a-Se were carried out by Pai and Enck (1975). The results show that, for $F \geqslant 10$ kV cm^{-1}, η is equal for electrons and holes, and is well described by the Onsager theory for geminate recombination in an ion plasma (Onsager 1938). In particular, for excitation at or above the 'photoexcitation edge' at ~2.8 eV (Hartke and Regensburger 1965), the field dependence is weak, η only dropping from near unity at $F \sim 300$ kV cm^{-1} to 0.3 at ~10 kV cm^{-1} in an S-like manner. However, the measurements show a knee below which η becomes proportional to F and unequal for electrons and holes. This effect is attributed to surface recombination and supported by direct experimental evidence (Enck 1973).

A brief resumé of the Onsager model applied to electron–hole dissociation is indicated. Assume that after ionisation and subsequent degradation of the excess kinetic energy by collisions with phonons (Davis 1970) the electron and hole are separated by

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a 'thermalisation distance' r_0. If r_0 exceeds a critical distance $r_c = e^2/4\pi\varepsilon_0\varepsilon_r kT$ (the 'Onsager radius'), which describes the sphere of influence of the mutual Coulomb attraction, the carriers are free to drift away. If $r_0 < r_c$, the probability of escape from recombination depends on the combined effects of the diffusion and extraction fields, and increases with r_0, F and temperature T. For sufficiently high fields, saturation is achieved. However, even at zero F, the diffusing pair may miss each other, and low-field saturation is approached as F falls below kT/er_c (Warter 1971).

Although in principle other recombination mechanisms (such as surface recombination, see above) may come into play at low F, the onset of the low-field saturation region usually manifests itself as a short plateau in the region of 10 kV cm^{-1}, giving rise to the characteristic S-shape of the η–F relationship at higher fields. This has been very clearly observed not only in a-Se (Tabak and Warter 1968, Pai and Enck 1975) but also in Se–Te alloys (Hagen and Derks 1984), amorphous polycarbonate doped with various organic molecules (Borsenberger *et al* 1978) and polycrystalline triphenylamine (Borsenberger 1978). It is at least hinted at in the amorphous polymers PVK (Pfister and Williams 1974, Borsenberger and Ateya 1978) and TNK:PVK (Melz 1972), in a-Si (Carasco and Spear 1983) and a-As_2Se_3 (Mort *et al* 1981) as well as in crystalline anthracene (Chance and Braun 1973) and As_2S_3 (Blossey and Zallen 1974) where trapping is a problem.

The only prior study of carrier yield produced by electron bombardment of a-Se is that by Spear (1956). Under very low current densities and: (i) a steady, non-penetrating beam; (ii) a slowly pulsed, fully penetrating beam, the current yield was found to saturate at ~200 kV cm^{-1}. From the saturation value, the average pair production energy was estimated to lie between 18 and 25 eV. These findings gave us two further incentives for carrying out new measurements: (i) we observed no saturation in our preliminary experiments; (ii) it is now generally accepted that, in crystalline and polycrystalline semiconductors, the pair production energy averages about three times the band gap (Lappe 1959, Shockley 1961, Klein 1968, Alig and Bloom 1975). In a-Se, this would amount to ~7.5 eV.

2. Experimental details

The samples were prepared at Imperial College† by evaporation onto substrates held at 55–75 °C. The evaporation source was a stainless steel boat maintained at ~275 °C. The substrates were mostly polished and oxidised Al plates but, for low-temperature experiments, gold-plated fused Al_2O_3 wafers were used. The rate of evaporation produced a layer growth of 1–2 μm per minute. The typical layer thickness was 50 μm. An electrode of Au, 35–40 nm thick, was deposited on top of the films.

The charge yield was measured by a flight-time technique. The sample was bombarded by an electron beam of measured dose through the top Au electrode. The beam penetration range was normally 2–3 μm, and the pulse width 60 ns. The beam pulse was triggered as soon as possible after establishing the drift field. For low fields the delay had to be varied between 1 and 50 ms depending on detector sensitivity, but for high fields it was ~0.5 s. Separate experiments at intermediate fields showed that lengthening the delay had little effect on the yield (or transit time), although it tended to 'square' the transit profiles.

† See Kasap and Juhasz (1985). The authors wish to thank J G Killian for his kind assistance.

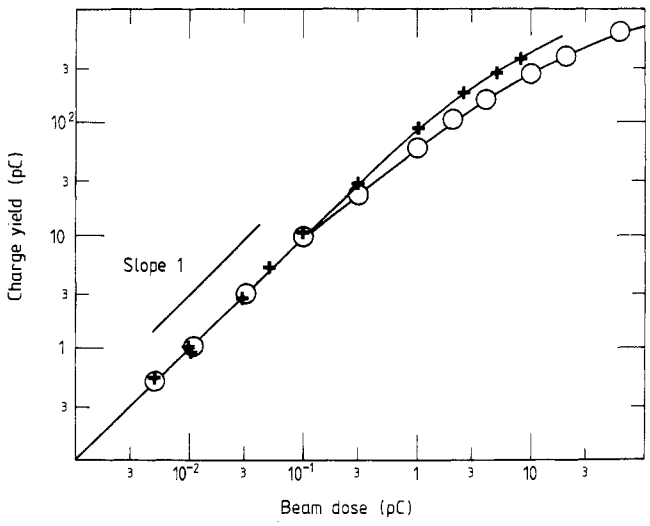

Figure 1. Charge yield as a function of beam dose. ○, electron yield; +, hole yield. 20 °C, beam voltage 12 kV, field 52 kV cm^{-1}.

The drift current was measured by an operational current amplifier with a rise time < 0.1 μs at the lowest sensitivity used to record hole transits, though longer for electron transits. The transit was captured by a fast digital storage oscilloscope. At room temperature (RT), the transits were esentially non-dispersive, with clearly defined transit times, though exhibiting 'tails'. The tail of a typical hole transit decayed away in a fraction of a transit time, as did that of an electron transit at high fields, but at low fields the tail of an electron transit could last about another transit time. The charge transiting the sample was deduced by integrating the area under the transit pulse including the tail.

At RT, the hole mobility was 0.15–0.16 $cm^2\ V^{-1}\ s^{-1}$ and the electron mobility 7–8×10^{-3} $cm^2\ V^{-1}\ s^{-1}$, both virtually independent of drift field. Both electron and hole lifetimes were $\geqslant 50$ μs. The mobilities varied by less than 10% between different samples irrespective of origin (Boliden Co., Hoboken Co., Nippon Co.). They compare well with the values quoted in the extensive literature, and demonstrate that the samples were of standard quality.

3. Experimental results

Figure 1 illustrates a representative measurement of the electron charge Q_e and hole charge Q_h collected following bombardment at RT with a beam dose Q_b, a constant beam voltage V_b and a fixed value of drift field F. Values of Q_e or Q_h measured at the same parameters Q_b, V_b and F varied by at most 10% from sample to sample, irrespective of the origin of the Se. This variation lies within the combined errors of thickness, parameter and charge measurements.

Since the transit times (0.5 μs for holes, 10 μs for electrons in this 40 μm thick film) are considerably shorter than the lifetimes before capture into deep levels ($\geqslant 50$ μs), the charge collected represents the charge generated. Figure 1 clearly demonstrates two important features observed in all samples: at low beam doses, Q_e and Q_h are (i)

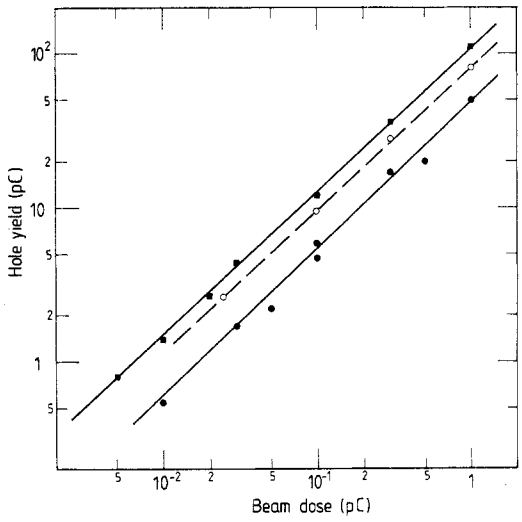

Figure 2. Hole yield as a function of beam dose. ■, 20 °C; ○, −33 °C; ●, −77 °C. Beam voltage 16 kV, field 55 kV cm^{-1}.

equal and (ii) strictly proportional to beam dose. Experiments other than those designed to investigate the relation between Q_e or Q_h and Q_b were carried out at sufficiently low Q_b for (i) and (ii) to apply (usually 0.1 pC). It then becomes possible to define a relative yield $Y = Q_{e,h}/Q_b$ which is independent of Q_b. In the literature, Y is often called charge 'gain' (e.g. Ehrenberg and Gibbons 1981).

Above Q_b ~0.1 pC in figure 1, Q_e becomes sub-linear and appears to approach saturation at ~1 nC. The proportionality of Q_h persists for another decade, so that Q_h exceeds Q_e, but eventually Q_h also shows signs of impending saturation. These features are readily understood.

(i) The induced charge must saturate at 2 CV (Lampert and Mark 1970), where C is the capacitance corresponding to the bombarded area of the sample and V is the applied voltage. In figure 1, CV ~500 pC, so that saturation of both Q_e and Q_h is expected at ~1 nC.

(ii) When Q_e is being measured, the hole charge generated is drawn out of the excited region and into the cathode long before the last electrons have entered the drift space. The net negative space charge in the sample then reduces the extraction field and therefore the yield which, at 52 kV cm^{-1}, is approximately proportional to field (see figure 3 below). The effect should become noticeable when Q_e exceeds a few percent of CV; at $Q_b = 0.1$ pC in figure 1, Q_e in fact corresponds to 2% of CV. At higher doses, the electron transits exhibit the features typical of space-charge perturbation (Lampert and Mark 1970).

(iii) In contrast, in measurements of Q_h, electrons remain in the excited region and compensate the positive charge while the holes vacate this region. The extraction field, therefore, remains unperturbed up to much greater values of Q_h.

Figure 2 represents similar experiments carried out at different temperatures. The equality of Q_e and Q_h at low doses was established also at −33 °C but, at −77 °C, the electron current was too low and the transit profiles were too dispersive for reliable measurement. Therefore only hole yields are recorded in figure 2. The yield was also measured at +35 °C but could not be resolved from that at RT. The strict proportionality

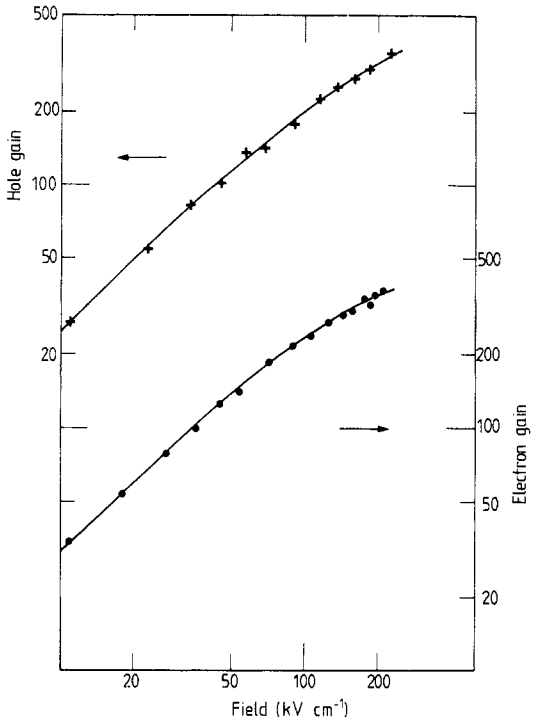

Figure 3. Charge gain (yield per unit beam dose) as a function of field. ●, electron gain at 20 °C, right-hand scale; +, hole gain at −33 °C, left-hand scale. Beam voltage 16 kV, dose 0.1 pC.

of Q_h to Q_b is seen to be preserved down to −77 °C. (Readings taken at −55 °C are not shown. They lie about midway between those at −33 °C and −77 °C and show the same trend.) We were unable to establish any clear functional temperature dependence of the yield; approximately, Q_h varies as $T^{1.5}$.

The most important result in this work, i.e. the field dependence of the charge gain Y, is presented in figure 3. In order to optimise accuracy, electron yields were measured at RT but hole yields at −33 °C; the findings of the experiments described above allows us to assume that $Q_e = Q_h$ at either temperature irrespective of field F. The scales of Y ($=Q_e/Q_b$ or Q_h/Q_b) are displaced for clarity. Below ~50 kV cm^{-1}, both graphs are nearly straight lines with slopes slightly less than unity. Above 50 kV cm^{-1}, the curvature observed suggests a very gradual approach to eventual saturation well above 300 kV cm^{-1}. There is no indication of the knee or S-region observed in the photoelectric quantum efficiency.

Two subsidiary experiments were carried out. In one (not illustrated), the beam voltage and dose (0.1 pC) as well as the field were kept constant, but the width of the bombarding pulse was varied between 60 ns and 10 μs. Within experimental errors, the gain Y remained invariant and equal for electrons and holes. We conclude that: (i) an equilibrium concentration of free carriers is established in $\ll$60 ns; (ii) the current gain behaves in the same way as the charge gain. However, it must be pointed out that the lowest dose for which transits could be reliably recorded was 5×10^{-15} C. The corresponding current density was ~0.2 μA cm^{-2}, which is still a factor of 10 greater than that at which Spear (1956) found voltage saturation (§ 1).

The result of the other experiment is shown in figure 4. Here the charge gains Y for electrons and holes are plotted as a function of beam voltage V_b at RT and a constant

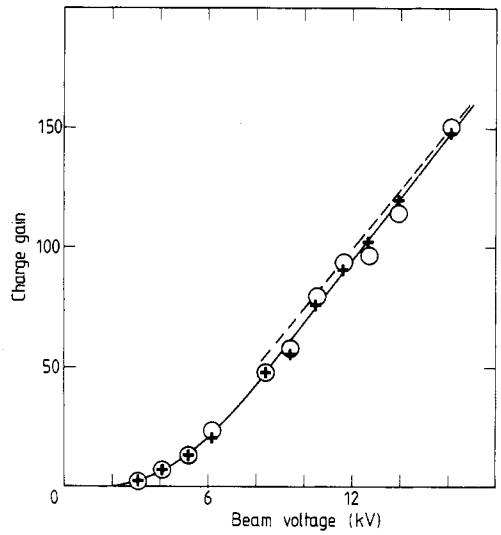

Figure 4. Charge gain as a function of beam voltage. ○, electron gain; +, hole gain. 20 °C, beam dose 0.1 pC, field 53 kV cm^{-1}. Broken line, gain corrected for energy loss in top Au electrode.

field. The cut-off at ~3 kV is accounted for by the absorption of the bombarding electrons in the top Au electrode of 35–40 nm thickness. According to Feldman (1960), full penetration of a 37.5 nm thick Au layer requires ~5.5 keV, so that at 3 kV we merely observe the effect of stragglers. In fact, the extrapolated straight-line portion of the gain curve intersects the V_b axis at ~5 kV. With increasing V_b, the energy lost in the Au layer decreases. At 16 kV the loss is only ~150 eV. The broken line in figure 4 represents the straight-line part of the gain curve corrected for the energy loss in a 37.5 nm thick Au electrode.

It is noted that, within experimental errors, electron and hole gains are again equal. This equality, over a penetration range from virtually zero to ~3 μm, leads to the conclusion that surface recombination plays a negligible part in our experiments, since at least the diffusion lengths if not the surface recombination velocities must differ for electrons and holes.

4. Discussion

Unlike excitation by monochromatic light, ionisation by electron bombardment should result in a broad distribution of the initial kinetic energies of the electron–hole pairs generated. However, we can estimate the *minimum* kinetic energy after impact ionisation by a slow secondary (δ) electron. We use a classical argument following Alig and Bloom (1975). Conservation of momentum requires that the newly created electron and hole, and the δ electron after impact, each possess kinetic energy $\frac{1}{3}E_0$, where E_0 is the energy of the δ electron before impact. If E_0 represents the minimum energy required for ionisation across the band gap ΔE_g, then $E_0 = \Delta E_g + \frac{1}{3}E_0$, or $E_0 = \frac{3}{2}\Delta E_g$. That is, the electron–hole pair is left with a combined excess kinetic energy of $\frac{1}{3}\Delta E_g$. Taking $\Delta E_g = 2.4$ eV (e.g. Stuke 1970), this excess is 0.8 eV, and corresponds to excitation by a photon of energy 3.2 eV or wavelength 390 nm. Comparison with the curve of Pai and Enck (1975) for 400 nm then shows that the gain Y should be within 70% of saturation at 200 kV cm^{-1} and only drop to ~30% at 10 kV cm^{-1}. It is quite obvious that our results (figure 3) do not conform to this pattern.

Under electron bombardment, therefore, another and stronger recombination mechanism predominates. This must still possess monomolecular character (figures 1 and 2), but recombination through traps (Rose 1963) must be excluded. Quite apart from the fact that the only known candidates for recombination centres are the mid-gap centres observed in transit measurements, with lifetimes $\geqslant 50\ \mu s$ whereas equilibrium is established in <60 ns (§ 3), such recombination cannot produce the equal *schubwege* (mobility-lifetime products) required for equal electron and hole yields. Nor can new recombination centres be produced by the beam itself, since this would lead to bimolecular kinetics. Surface recombination can, at most, play a subsidiary role. We are forced to the conclusion that we are observing an initial recombination mechanism, different from the Onsager mechanism characteristic of photoexcitation, but inherent in the generation of electron–hole pairs by electron bombardment.

In the Onsager model, the ionisation events must be separated by at least the Onsager radius r_c (§ 1) which, in Se, is ~ 10 nm. Ionisation by electron bombardment, however, is highly non-uniform and heavily concentrated in small volumes. We illustrate this by summarising the history of a 10 keV electron incident on water (Mozumder and Magee 1966). Initially, the electron suffers mainly glancing collisions and produces isolated δ spurs, each abstracting ~ 100 eV. Losses between 100 and 500 eV produce overlapping spurs or 'blobs'. When the primary electron has been slowed down to between 5 keV and 500 eV, head-on collisions become probable. In this section of the track, the δ trajectories are almost collinear with the axis, and energy transfer is high. Absorption in the end track accounts for $\sim 65\%$, in blobs for $\sim 10\%$, and in non-overlapping spurs for $\sim 25\%$ of the total primary energy. Branch tracks are unlikely at 10 keV.

Say a primary electron enters the final part of its track with an energy of 1 keV. Its range in a-Se is then ~ 10 nm, as calculated from the formulae of Feldman (1960) or Bethe (1930). If, on average, ionisation required 20 eV, the average axial distance between ionisation sites would be only one atomic spacing. The radial spread is difficult to estimate, but it is clear that the Onsager spheres strongly overlap. Under these circumstances the controlling mechanism is that of bimolecular recombination between non-sibling electrons and holes within each microvolume of high-energy transfer. Nevertheless, the overall kinetics are of monomolecular character. This is because, as the beam current density is increased, the number of tracks increases in proportion, but the distribution of energy loss along each track remains on average unchanged.

The problem of initial bimolecular recombination (of ions in gases or liquids) was first solved by Jaffé (1913). He termed it 'columnar' recombination, cylindrical geometry being assumed for the volumes of high electron energy loss. In Jaffé's solution, the ions are initially distributed in Gaussian manner with arbitrary mean radius (standard deviation) r_i. During subsequent diffusion, they either recombine or escape the column, the extraction field F being introduced as a perturbation. Kramers (1952) pointed out that, when $F \gg kT/er_i$, the effect of the field outweighs that of diffusion, so that diffusion may be neglected and Jaffé's treatment is invalid. For this case, Kramers predicts a high-field solution of the form

$$Y(F)/Y_{sat} \sim 1 - \text{constant}/F \tag{1}$$

where Y_{sat} is the high-field saturation value of Y. We could not fit our results (figure 3) to equation (1), and conclude that r_i is less than ~ 10 nm.

Ausman and McLean (1975) and Taylor and Al-Jassar (1981)† applied Jaffé's theory

† In a later paper, however, Taylor and Al-Jassar (1984) concluded that, on balance, Onsager recombination afforded a better explanation of the yield in SiO_2.

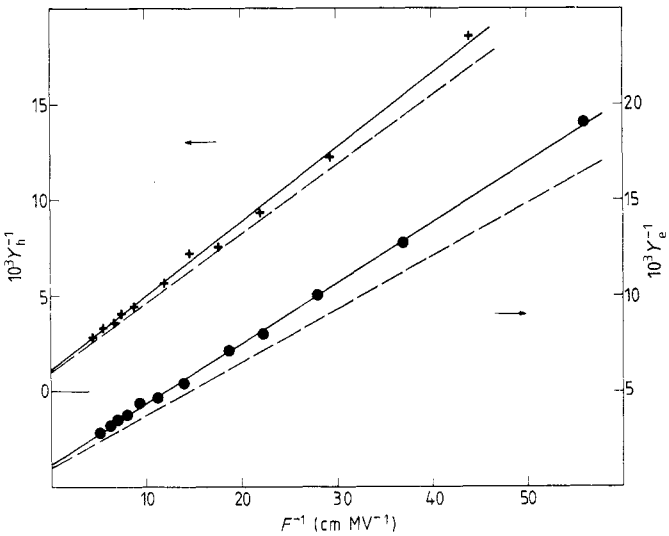

Figure 5. Reciprocal charge gain Y^{-1} as a function of reciprocal field F^{-1}. ●, electrons at 20 °C, right-hand scale; +, holes at −33 °C, left-hand scale; from figure 3. Intercepts give $(Y_{sat})^{-1}$. Broken lines, Y_{sat}/Y.

to the current yield in electron-bombarded SiO_2 layers. They utilised the fact that, at sufficiently high fields ($F > 2\,kT/er_i$), Jaffé's result reduces to the approximate form

$$Y(F)/Y_{sat} \sim (1 + F_0/F)^{-1} \tag{2}$$

where

$$F_0 \sim 0.3e/\varepsilon_0\varepsilon_r l r_i \tag{3}$$

and l is the initial (axial) separation of ion pairs in the columns. It will be noted that, at high enough fields, the functional form of equation (2) resembles that of equation (1).

In figure 5, the two gain curves shown in figure 3 have been replotted in the form Y^{-1} versus F^{-1} to test the applicability of equation (2). Excellent straight regression lines can be drawn through all the points above 20 kV cm^{-1}. The intercepts for $F^{-1} = 0$ correspond to saturation values $Y_{sat} \sim 870$ for electrons at RT and ~930 for holes at −33 °C. The difference may be the result of an experimental artifact: when measuring Y_h, the top Au electrode is made positive with respect to ground. For $F = 100$ kV cm^{-1}, the increase in potential and therefore in primary electron energy is ~500 eV. When measuring Y_e, the top electrode is negative and the incident electron energy is correspondingly reduced. We conclude that Y_{sat} ~900 for both curves and therefore independent on temperature, as one would expect.

However, the actual gain is undoubtedly temperature dependent (figure 2), and this seems to be due to a temperature dependence of F_0. This is brought out more clearly by plotting $Y_{sat}/Y(F)$ (broken lines in figure 5). The slopes of these lines correspond to $F_0 \sim 275$ kV cm^{-1} at RT and ~360 kV cm^{-1} at −33 °C, a difference significantly greater than attributable to experimental errors. Equation (3) reveals no physical basis for a temperature dependence of F_0. The only parameter which might be envisaged to vary with temperature is the arbitrarily chosen r_i; we would require r_i to increase with increasing temperature. However, r_i is related to the thermalisation distance r_0 and this

is, in first approximation, invariant with temperature (Davis 1970, Knights and Davis 1974, Pai and Enck 1975). We can only suggest that equations (2) and (3) represent a gross over-simplification of the situation in a-Se, where capture into shallow traps takes place during initial diffusion and at different rates for electron and holes (e.g. Grunwald and Blakney 1968), so that the diffusion becomes dispersive. Unfortunately, we must leave the problem of the temperature dependence of F_0 unresolved.

The average energy required for pair production is given by $\Delta E_{pp} = E_b/Y_{sat}$, where E_b is the energy of the primary electrons after allowing for the loss in the top electrode. At $V_b = 16$ kV this loss is negligible (figure 4), so that $\Delta E_{pp} = 16 \times 10^3/900 \sim 18$ eV. This result is in good agreement with the early estimate by Spear (1956: see § 1). We can understand Spear's observation of voltage saturation under full penetration of the 43 keV electron beam used on the basis of the columnar recombination model. Under these conditions, the average energy loss per unit track length would be significantly smaller than in our experiments. Consequently, in equation (3), l would be greater and F_0 lower. We cannot, however, reconcile the observation of saturation under weak bombardment by a steady 6 keV beam (Spear 1956) with our simple model.

Our value of ΔE_{pp} also agrees, probably fortuitously, with the estimates of this parameter in SiO_2 by Ausman and McLean (1975) and Taylor and Al-Jassar (1981). Ausman and McLean consider that the predominant ionisation mechanism in SiO_2 is via plasma oscillations excited by the electron beam. Assuming that all six valence electrons are involved in plasma oscillations in a-Se, we calculate a plasmon energy of 17.5 eV which is indeed close to ΔE_{pp}. Ausman and McLean deduce $r_i \sim 6.5$ nm in SiO_2, but we find it difficult to understand why plasma oscillations should remain so highly localised.

We offer a tentative alternative explanation of the high value of ΔE_{pp} on the conventional basis that pair production occurs mainly by δ electrons. In a wide-band semiconductor, an electron or hole with kinetic energy greater than $E_0 = \frac{3}{2}\Delta E_g$ (see above) can excite another pair, so that there is little waste. In a tight-binding molecular semiconductor, the conduction and valence bands are too narrow to allow this. Assuming equal band widths ΔE_{bd}, and again invoking the principle of momentum conservation, the maximum primary energy which can be productively absorbed is $E_0' = \Delta E_g + 3\Delta E_{bd}$. Assuming $\Delta E_{bd} = 0.8$ eV, say, and $\Delta E_g = 2.4$ eV as before, we estimate $E_0' = 4.8$ eV. If all δ energies are equally likely, and the density of band states is taken as constant for simplicity, the probability of a productive collision is only $(E_0' - E_0)/E_0' = \frac{1}{4}$. Since the average energy loss is $\frac{1}{2}(E_0' + E_0)$, the effective pair production energy becomes 16.8 eV.

Finally, we ask what are the conditions which lead to the observation of columnar recombination under electron bombardment or other energetic radiation. Evidently, they are the same as those which cause Onsager recombination to control photoelectric yield. In wide-band crystalline or polycrystalline semiconductors, electrons and holes are able to escape initial recombination of either kind by virtue of their long mean free paths. In molecular and especially amorphous semiconductors, on the other hand, the short mean free paths prevent such escape (Mort 1981). Initial recombination of one kind or the other, depending on the mode of excitation, then becomes unavoidable, with a-Si (Carasco and Spear 1983) apparently representing a borderline case. The example of crystalline anthracene lends support to our conclusion. Here the photoelectric yield is Onsager-like (Chance and Braun 1973), while Schott (1969) found it necessary to invoke columnar recombination to account for the low electron-bombardment yield reported by Delany and Hirsch (1968).

Acknowledgments

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