Transport and recombination properties of amorphous arsenic telluride

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Considerable information about transport and recombination properties of amorphous As_2Te_3 has been obtained from studies of steady-state and transient photoconductivity, dark conductivity, and thermopower in the temperature range from 200 to 350°K: (i) The conductivity is dominated by holes both in the dark and under illumination. The holes move in states 0.28 ± 0.02 eV below the Fermi level; the mobility is activated, with $\mu \simeq 10 \exp(-0.16/kT)$ cm^2/V sec. The room-temperature concentration of holes is about 10^{16} cm^{-3}. (ii) The dependence of the lifetime on light intensity and temperature strongly suggests a direct recombination mechanism between electrons and holes. The temperature independence of the recombination constant ($b = 10^{-10}$ cm^3 sec^{-1}) implies that the process is not diffusion limited. (iii) Lack of dependence of the photoconductivity on wavelength for larger than band gap excitation shows that the recombination takes place by bulk rather than by surface processes.

I. INTRODUCTION

Investigations of the steady-state photoconductivity of several amorphous chalcogenide films,[1–15] and of amorphous silicon prepared by decomposition of silane,[16] have revealed that the photoconductivity $\Delta\sigma$ has a maximum in its dependence on temperature and that it varies linearly with light intensity I at temperatures above the maximum and as $I^{1/2}$ below it. At yet lower temperatures, another regime has been reported,[9] in which the photoconductivity becomes linear with light intensity again, and is substantially temperature independent.

Several authors have suggested[1,9,12,16] that the behavior around the temperature maximum is due to a recombination scheme involving direct recombination between electrons and holes, though opinions vary as to whether the recombination takes place between both carriers in localized states, one carrier only in delocalized states, or both carriers in delocalized states. It is also not clear whether the photocurrent or, for that matter, the dark current is carried predominantly by carriers in localized or delocalized states and whether the dominant photocarriers are the same as the dark carriers.

The investigations described in this paper throw considerable light on these questions. Both steady-state and transient photoconductivity measurements were carried out as a function of temperature and light intensity. Particular attention was devoted to obtaining the initial decay curves of the photocurrent after illumination was turned off without being limited by the fall time of the light source and the response time of the measuring circuit. In this manner direct information was obtained about recombination constants. By combining the photoconductivity measurements with studies of the dark conductivity and the thermoelectric power on samples prepared simultaneously with the photoconductivity samples, we reach the conclusion that the recombination is indeed direct and that the same recombination mechanism prevails on both sides of the photoconductivity maximum. The analysis also suggests that the dominant photocarriers are holes as are the dark carriers. We find that the holes move with a thermally activated mobility, though it is not possible to conclude whether they move in localized or delocalized states.

II. EXPERIMENTAL METHODS

As_2Te_3 films were deposited by sublimation of bulk material on sapphire substrates. Thicknesses were typically between 0.5 to 1.5 μm and the films were stoichiometric to within about 10% as determined from microprobe analysis. Samples for four-probe dark conductivity and for thermopower measurements were deposited simultaneously with photoconductivity samples. The latter were of an open-cell geometry made by depositing the films on preevaporated gold electrodes spaced about 100 μm apart. The applied field was kept below 4 kV cm^{-1}, well within the Ohmic regime. All measurements were done in vacuum.

Radiation sources included a He-Ne laser ($\lambda = 6328$ Å), a GaAs laser at 77 °K ($\lambda = 8400$ Å), and a YAG: Nd laser ($\lambda = 1.06$ μm), all of which should produce carriers in extended states since the optical gap of the material is about 1.0 eV.[17] The He-Ne and YAG: Nd lasers were used for steady-state

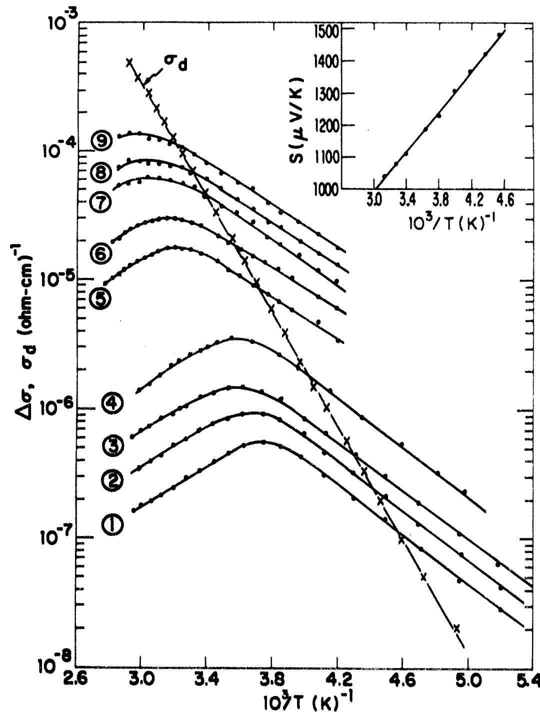

FIG. 1. Photoconductivity in a-As_2Te_3 plotted against $1/T$. The data were taken with three different excitation sources and photon flux (cm^{-2} sec^{-1}): I_1 (YAG) $=1.6\times10^{16}$, I_2 (YAG) $=3.2\times10^{16}$, I_3 (YAG) $=6\times10^{16}$, I_4 (He-Ne) $=1.4\times10^{17}$, I_5 (GaAs) $=1.2\times10^{18}$, I_6 (GaAs) $=2.3\times10^{18}$, I_7 (GaAs) $=7.4\times10^{18}$, I_8 (GaAs) $=1.2\times10^{19}$, I_9 (GaAs) $=2.3\times10^{19}$. In this figure we plot the photoconductivity $\Delta\sigma$ which was calculated by multiplying the experimentally measured quantity Σ_{ph} with the geometrical factor of the sample 2.2×10^3 cm^{-1}. σ_d denotes the dark conductivity. The insert is the plot of thermoelectric power S vs $1/T$.

measurements while the GaAs laser was modulated electrically to produce pulses of 2–3 μsec duration with rise and fall times of less than 40 nsec. The pulses were long enough for the photoconductivity to reach the steady-state value and the rise and fall times were much shorter than the measured lifetimes. The GaAs radiation was therefore used both for steady-state and transient measurements.

The photoconductivity signal was measured across a load resistor in series with the sample and a battery. For transient measurements, care was taken to insure that the response time of the measuring system, including amplifiers and boxcar integrator, was less than the 40 nsec rise time of the GaAs laser. The light intensity was measured with a calibrated Si detector positioned in the place of the sample and covered with an aperture having the dimensions of the sample. Calibrated neutral-density filters were used to alter the light intensity. Experimental details on both thermoelectric power and photoconductivity measurements have been published elsewhere.[18]

III. EXPERIMENTAL RESULTS

A. Dark transport measurements

The dark conductivity was found to be thermally activated between 200–350 °K with an activation energy of 0.44 eV and a preexponential factor of 2×10^3 Ω^{-1} cm^{-1} (Fig. 1). The thermoelectric power S (insert of Fig. 1) was found to be positive and a plot of S vs $1/T$ yields a slope of 0.30 eV with an intercept of 50 μV/K. Both results are similar to results reported elsewhere.[12,15,17,19]

B. Steady-state photoconductivity

Figure 1 also depicts the behavior of the steady-state photoconductivity $\Delta\sigma$ at different light intensities I as a function of temperature. At temperatures above the maximum $\Delta\sigma\propto I$ and is thermally activated with an average activation energy of -0.15 ± 0.02 eV. Below the maximum $\Delta\sigma\propto I^{1/2}$ with an average activation energy of 0.17 ± 0.02 eV. The low-temperature data agree with those reported by Main and Owen[12] and Bube *et al.*,[15] but these authors report activation energies of about 0.20 eV for the high-temperature data. The lack of significant dependence of $\Delta\sigma$ on the laser wavelength, which was corroborated by measurements with a nonlasing light source down to $h\nu=1.2$ eV, is in agreement with the results of Marshall *et al.*,[11] but disagree with those of Stourac *et al.*[20] Our results suggest the absence of preferential surface recombination.

C. Transient photoconductivity

The photoconductivity kinetics were examined by studying the decay of $\Delta\sigma$ after excitation by a GaAs-laser pulse whose properties were described in Sec. II. All lifetimes, as defined below, were substantially longer than the 40 nsec decay time of the laser pulse or the RC time of the measuring circuit. As seen in Fig. 2, $\Delta\sigma$ in the linear regime decays exponentially down to about $\frac{1}{3}$ of the steady-state value. In the square-root regime the decay curves could be fitted to the relation

$$\Delta\sigma/\Delta\sigma_{ss}=[\ln(1+At)]/At\ , \qquad (1)$$

where $\Delta\sigma_{ss}$ is the steady-state photoconductivity and A is an adjustable parameter. We define a lifetime τ_0 as $(d\ln\Delta\sigma/dt)^{-1}_{t=0}$, where $t=0$ denotes the end of the excitation. From Eq. (1) it is clear that the lifetime thus defined equals $2/A$ in the square-root regime. In Fig. 3 we plot the lifetime τ_0 as a function of temperature and light intensity, obtained from data such as those in Fig. 2. The striking behavior shown in Fig. 3 is that τ_0 in the linear regime (light intensities I_3, I_4 at all tem-

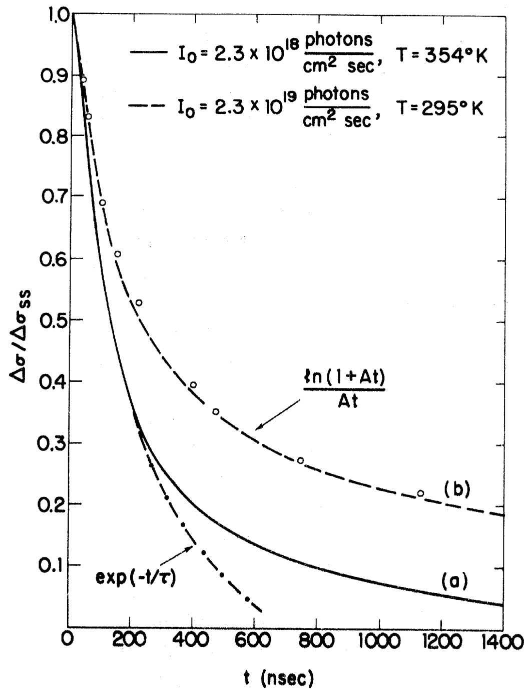

FIG. 2. Photoconductivity relaxation curves. Curve (a) is taken at $T = 354$ °K and $I = 2.3\times 10^{18}$ photons/cm² sec. It decays exponentially down to about $\frac{1}{3}$ of the steady-state value and corresponds to the linear regime. Curve (b) is taken at $T = 295$ °K and $I = 2.3\times 10^{19}$ photons/cm² sec. It decays according to Eq. (1). Circles are best fit to Eq. (1) with $A = 10^7$ sec^{-1}.

ly independent of detailed modeling. Values of the various parameters calculated in this section and Sec. IV B are for one particular sample and are representative of other samples.

In the linear regime, above the maximum in Fig. 1, the photocurrent was found to depend linearly on light intensity, and the initial decay rate is exponential with a decay constant independent of the light intensity. The rate equation for the dominant photocarrier Δp_d must therefore be of the form

$$\frac{d(\Delta p_d)}{dt} = -\frac{\Delta p_d}{\tau} + G , \qquad (2)$$

where τ is the recombination time of these carriers and G is the volume generation rate by the light.

In the presence of appreciable "fast" trapping, Δp_d of Eq. (2) is the sum of the concentration of carriers in mobile states and in traps. By mobile states we mean states in which the carriers can carry current, in contrast to traps in which they are immobilized. By "fast" traps we mean traps which are characterized by capture and release rates of carriers between them and mobile states, which are much greater than the recombination time of the photocarriers.[21,22] The steady-state photoconductance Σ_{ph} derived from Eq. (2) is then given by[23]

$$\Sigma_{ph} = (h/l) q (1-R) I \mu \tau (1 - e^{-\alpha d}) . \qquad (3)$$

In this expression d is the thickness and h the width

peratures and I_1, I_2 at high temperatures) increases exponentially with $1/T$ with an activation energy of -0.26 ± 0.02 eV and is independent of I, while τ_0 in the square-root regime (I_1 and I_2 at low temperatures) is independent of temperature, but is proportional to $I^{-1/2}$.

A similar curve for light intensities approximately 1000 times smaller and for generally much lower temperatures has been published by Main and Owen.[12] These authors report a much larger slope for the intensity-independent regime than that shown in Fig. 3, and instead of a temperature-independent behavior of the lifetime at high light intensities and low temperatures, they report a slight decrease of lifetime with decreasing temperature in this regime.

IV. DISCUSSION

A. Determination of mobility and recombination constant

We first examine the experimental results without any reference to the details of the recombination mechanism. The information and parameters which result from this analysis are therefore large-

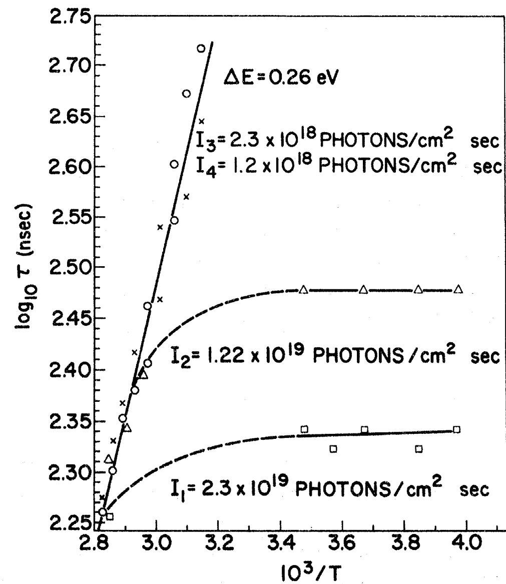

FIG. 3. Temperature and light-intensity dependence of recombination lifetime. The thermally activated part was derived from the linear data and the horizontal lines were derived from the square-root data, as explained in the text.

of the sample, l is the distance between electrodes, R is the reflectivity, I is the incident flux, α is the absorption constant, q is the electronic charge, and μ is the effective mobility of carriers with a common lifetime. Given the values for R and α it is therefore possible to calculate μ. The value of the effective mobility thus calculated will place a lower limit on the mobility of the mobile carriers because of the possibility of "fast" trapping phenomena. At room temperature, using $\alpha = 1\times10^5$ cm^{-1},[17] and $R = 0.40$,[24] one obtains $\mu = 2.3\times10^{-2}$ cm^2/V sec. The temperature dependence of the mobility is given by

$$\frac{d\ln\Sigma}{d(1/T)} - \frac{d\ln\tau}{d(1/T)} = -0.14 + 0.26 = 0.12 \text{ eV},$$

with an uncertainty of approximately ± 0.04 eV. Whether the mobility is truly activated or whether it merely appears to be activated because of trapping effects cannot be determined from these measurements alone.

In the square-root regime, at temperatures below the maximum of Fig. 1, the steady-state value of the photocurrent and the initial time constant were found to depend on the square root of the light intensity. This behavior is consistent with a rate equation of the form

$$\frac{d\Delta p_d}{dt} = -b\,\Delta p_d^2 + G\ , \qquad (4)$$

where b is the recombination parameter and Δp_d is again the dominant photocarrier concentration. As shown elsewhere[25] the above rate equation yields Eq. (1) for the time dependence of the photocurrent, with A given by

$$A = [\alpha b(1-R)I]^{1/2}\ . \qquad (5)$$

From the experimental value of τ_0, the recombination parameter b was found to equal 7.3×10^{-11} cm^3/sec, again using α and R from Refs. 17 and 24. Similar values for b have been obtained from measurements on other chalcogenides glasses, suggesting a similar recombination mechanism.[1,13]

The steady-state photoconductance in the square-root regime is of the form[1,23]

$$\Sigma_{ph} = \frac{h}{l}\,2q\left(\frac{(1-R)I}{\alpha b}\right)^{1/2}\mu(1-e^{-\alpha d/2})\ . \qquad (6)$$

Inserting the value of b from the photoconductivity decay measurements, a room temperature mobility of 1.8×10^{-2} cm^2/V sec is obtained, which is in excellent agreement with the value of 2.3×10^{-2} cm^2/V sec calculated from the linear regime. Since b was found to be temperature independent the temperature dependence of the mobility is that of the photoconductance, so that $\mu \simeq 10e^{-0.16\ \text{eV}/kT}$ cm^2/V sec.

From the measurements in the square-root regime it is again not possible to decide whether the activation energy is only apparent, due to fast trapping, or whether the dominant photocarriers really move with an activated mobility. One may argue that the temperature independence of the lifetime in the square-root regime proves the absence of significant trapping effect by the following reasoning: In the presence of fast traps the measured decay time will be longer than the decay time of the mobile carriers if the carriers recombine mainly via mobile states. In that case[21,22] the decay time of the mobile carriers will be enhanced by a factor $\Delta n_t/\Delta n$, the ratio of trapped to free carriers. This ratio has a temperature dependence given approximately by $e^{E_t/kT}$, where E_t is the trap depth relative to the lowest energy of the free carriers. Thus, the temperature independence of b would argue against fast-trapping effects. It is, however, possible that the recombination occurs mainly via the trapped carriers,[22] in which case the measured decay time would not be temperature dependent. The mobility determined from the steady-state and kinetic photoconductivity measurements would, however, still be an effective mobility given by $\mu = \mu_0 g\, e^{-E_t/kT}$, where μ_0 is the mobility of the mobile carriers and g is a factor which depends on the effective density of states of the free carriers and of the trapped carriers. Thus, the temperature independence of b alone cannot rule out the presence of fast traps. Additional strong evidence against such traps, however, comes from comparing the activation energy of the mobility obtained from the photoconductivity experiments with the activation energies obtained from the dark conductivity and the thermopower. The activation energies obtained from the temperature dependence of the dark conductivity and of the thermopower are 0.44 and 0.30 eV, respectively. The difference between them, 0.14 eV, is in striking agreement with the mean of the mobility activation energies obtained from the two regimes of the photoconductivity, namely, 0.12 and 0.16 eV. As already mentioned elsewhere,[26] the agreement strongly suggests that the dominant photocarriers are holes and that the mobility determined from the photoconductivity results is the true microscopic mobility.[27] It thus appears that the holes move in states about 0.3 eV below the Fermi level and that they recombine from these states. Without further modeling it is not possible to say anything about the states from which the electrons recombine. All one can say is that since the dominant photocarriers appear to be holes, the mobility of electrons in these states must be very small. Furthermore, since b is temperature independent the electrons apparently recombine from these states rather than having to be first reemitted into mobile states.

B. Direct recombination model

As already mentioned in Sec. I a number of authors[1,9,12,16] have speculated that the dominant recombination mechanism around the photoconductivity maximum is due to a direct recombination of electrons and holes rather than due to a Shockley-Read-type[28] recombination scheme. Simmons and Taylor[29] have used such a scheme recently to explain the maximum in the photoconductivity, but the results of their model contradict at least one important experimental observation: While their model predicts that the photoconductivity maximum is due to a maximum in the carrier concentration with temperature, it is clear from Fig. 3 that the carrier concentration becomes independent of temperature at temperatures below that of the maximum since the lifetime is constant.

In this section we show that in the absence of deep trapping the direct recombination model can account for the experimental observations. If, in addition, the assumption is made that the material is p type because the hole concentration is much greater than the electron concentration, one can conclude from the model that the same recombination mechanism prevails on both sides of the photoconductivity maximum. This model also stands the test of self-consistency in that it allows one to calculate the dark conductivity from photoconductivity measurements. An alternative version of the direct recombination model, namely, that the material is p type not because the hole concentration is greater, but because the hole mobility is greater than that of electrons will also be considered briefly.

The rate equation for direct recombination is given by

$$\frac{dn}{dt}=\frac{dp}{dt}=-b(n_0+\Delta n)(p_0+\Delta p)+G+G_{th}, \qquad (7)$$

where we do not distinguish between mobile carriers and carriers in fast states since they all have a common recombination time. In this equation n_0 and p_0 are the carriers in thermal equilibrium; Δn, Δp are photocarriers. As before, G is the generation rate of carriers due to the light; G_{th} is the generation rate in the absence of light, which cancels the term bn_0p_0. In this equation $\Delta n=\Delta p$ only in the absence of deep traps. If one or both of the photocarriers are trapped in states out of which they are released at a rate much lower than the rate at which the mobile carriers and those in fast traps recombine,[22] then $\Delta n \neq \Delta p$. We *assume* the absence of such deep traps and show that with this assumption Eqs. (2) and (4), which were seen to fit the experimental data, are the low- and high-intensity limits of Eq. (7).

At high-light intensities, with $\Delta n=\Delta p$, Eq. (4) is obtained since Δn, $\Delta p \gg n_0$, p_0. At low intensities the analysis is slightly more ambiguous since three possibilities exist.

Case (a). The material is p type because $p_0 \gg n_0$. In this case Eq. (7) leads to Eq. (2), with $\tau_0=1/bp_0$. If the same recombination mechanism occurs in both the high- and low-intensity regimes, the temperature dependence of the recombination time in the low-intensity regime should be that of p_0, since b was found to be temperature independent. If p_0 contains mainly trapped carriers, the experimental value of 0.26 eV for $d\ln\tau/d(1/T)$ should be lower than $-dS/d(1/T)$, since the temperature dependence of the thermopower measures the temperature dependence of the mobile carriers only.[30,31] Within experimental error, however, $dS/d(1/T)=0.30$ eV agrees with $-d\ln\tau/d(1/T)$, suggesting that p_0 comprises mainly free carriers, in agreement with the conclusion reached in Sec. IV A.

If the assumption that $p_0 \gg n_0$ is correct, it should be possible to check the self-consistency of the model by calculating the dark conductivity from photoconductivity data. With $\tau=1/bp_0$ in the low-intensity regime, p_0 can be obtained from decay measurements in the two regimes since the measurement in the high-intensity regime yields b. At room temperature, one thus obtains $p_0=2\times10^{16}$ cm^{-3}. Combining this value with the mobility obtained from the photoconductivity measurements, 2×10^{-2} cm^2/V sec, one estimates a room-temperature conductivity ($qp_0\mu$) of $5\times10^{-5}\,\Omega^{-1}\,cm^{-1}$. This value is in excellent agreement with measured values, and is therefore consistent with the notion that the dominant photocarriers are holes as are the dark carriers.

The above analysis, as already mentioned, does not shed much light on the states from which the electrons recombine. One must assume that they fall into states of negligible mobility, or else they would be the dominant photocarriers, and that they recombine from these states.

Case (b). The material is intrinsic, $n_0=p_0$, but is p type because $\mu_p > \mu_n$. In this case the low-intensity decay time is $1/2bp_0$, which differs only by the trivial factor 2 from the result for case (a). The rest of the analysis and the conclusions are the same as for case (a). Since, on the basis of the thermopower measurements, the levels where the hole conduction takes place are about 0.3 eV below the Fermi level, the corresponding electron levels are about the same energy above E_F, barring an unreasonably great difference in the effective density of states of the two sets of levels.

Case (c). The material is p type in spite of $n_0 \gg p_0$ because the hole mobility is so much larger than the electron mobility. In the high-intensity regime the results would be the same as for (a)

and (b). In the low-intensity regime, however, the lifetime would be given by $1/bn_0$. The temperature dependence of the lifetime would therefore reflect the difference between the electron energy levels and the Fermi level. Since the experimental value for this difference is 0.26 eV, this would lead to a rather symmetric distribution of electron and hole states around the Fermi level. Clearly, case (c), whatever its plausibility, cannot be subjected to the same scrutiny of self-consistency as case (a) and case (b) which must therefore be considered as having greater merit.

In summary the direct recombination model with the additional assumption that $p_0 = n_0$ or $p_0 > n_0$ leads to the conclusion that the recombination mechanism is the same on both sides of the photoconductivity maximum, that the dominant photocarriers are holes as are the dark carriers, and that trapping effects are negligible.

C. Additional comments on recombination mechanism, nature of hole states, and mobility

It is beyond the scope of the paper to construct a truly detailed model to account for the transport and recombination properties investigated here. Thus, for example, the phenomenological analysis of Secs. IV A and IV B yielded the conclusion that the recombination of electrons and holes is via a direct mechanism and that the hole mobility is thermally activated. Just how the recombination takes place, how the energy is dissipated, and why the recombination constant is temperature independent and therefore not diffusion limited, cannot be answered on the strength of the experimental observations. It is known from photoluminescence experiments that the energy of recombination is released nonradiatively in the temperature range of interest though the luminescence efficiency is quite high at temperatures below 77 °K.[32] The energy must therefore be liberated by phonon emission. Perhaps the nonradiative recombination takes place in regions of the material in which the energy can be more readily dissipated by phonon emission due to some special atomic configurations.[33] Such regions may become accessible only at higher temperatures due to increased mobility of the carriers.[34] When electrons and holes arrive at such special recombination regions the probability that recombination takes place must be much smaller than the probability that the carriers move away from each other so as not to make the recombination constant b diffusion limited.[35]

One important question raised here concerns the nature of the states located about 0.3 eV below the Fermi level at which the hole conduction takes place and from which the holes recombine. Since the hole mobility is thermally activated one may surmise that these states are localized states though these experiments shed no light on whether they are band-tail states or a relatively narrow band of states within the gap as has been postulated by Main and Owen.[12] Indeed, from the results of these experiments one may even consider the possibility that these states are not localized at all but are band states, particularly since the preexponential factor of the mobility is comparable to that expected for carriers near the mobility edge.[36] In that case the mobility activation energy presumably stems either from polaronic effects[37] or long-range potential fluctuations.[38] Much more theoretical and experimental work is clearly needed to elucidate these questions.

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