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Research on Defects and Transport in Amorphous-Silicon-Based Semiconductors

Annual Subcontract Report 20 February 1992 – 19 February 1993

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PREFACE

This annual report covers the work performed at Syracuse University for the period February 20, 1992 to February 19, 1993 under National Renewable Energy Laboratory Subcontract XG-1-10063-7. Dr. Bolko von Roedern was the technical monitor of this project. The principal contributors to this report and their present institutions are:

Mr. Qing Gu, Syracuse University.

Dr. Eric A. Schiff (principal investigator), Syracuse University.

Mr. Qi Wang, Syracuse University.

We have benefitted from collaborations with Dr. Homer Antoniadis (University of Rochester), Dr. Subhendu Guha (United Solar Systems Corp.), Dr. Daxing Han (University of North Carolina), Dr. Jung-Keun Lee (Iowa State University), Dr. Yuan-Min Li (Solarex Corp.), Mr. Douglas Melcher (Syracuse University), and Dr. Marvin Silver (University of North Carolina).

SUMMARY OF PRINCIPAL RESULTS

1. Conduction Bandtails and Electron Drift in a-$Si_{1-x}Ge_x$:H and a-$Si_{1-x}C_x$:H Solar Cells.

Multijunction solar cells incorporating a-$Si_{1-x}Ge_x$:H for a narrow bandgap cell and a-$Si_{1-x}C_x$:H in a wide bandgap cell are generally viewed as the most promising avenue for achieving an amorphous silicon based solar cell with 15% stabilized conversion efficiency. Alloying a-Si:H with C or Ge typically leads to broadening of the conduction bandtail and degradation of the electron mobility. For Ge alloying, the effects on holes and the valence bandtail are much smaller; not much is known about hole transport in the C alloys. Our goal in this project is to measure these effects and determine whether they significantly affect solar cells.

Work on the a-$Si_{1-x}Ge_x$:H system has been in collaboration with Dr. Subhendu Guha (United Solar Systems Corp.); specimens were deposited at Energy Conversion Devices, Inc.. Time-of-flight measurements, optical characterization, as well as photovoltaic studies were performed. We found a very good correspondence between the bandgap of a-$Si_{1-x}Ge_x$:H alloys and the conduction bandtail width, suggesting that present deposition technology has achieved the minimum conduction bandtail width required by alloying itself. For p-i-n cells with a 0.35 micron i-layer we found a satisfactory correlation of time-of-flight parameters with "blue" fill factors, but the causal relationship is not established. Open-circuit voltages were very well accounted for by the alloy bandgap, and are thus probably *not* significantly affected by the conduction bandtail width.

Work on the a-$Si_{1-x}C_x$:H system has been in collaboration with Dr. Yuan-Min Li of Solarex, Inc. who also deposited the specimens. A short study has established that carbon does diminish the electron drift mobility. Further work on the electron and hole drift mobility is in progress.

2. Defect Relaxation and Shockley-Read Kinetics in a-Si:H.

Quantitative modeling of solar cells is usually based on "Shockley-Read kinetics." An important assumption of this approach is that the rate of emission of a photocarrier trapped on a defect is independent of quasi-Fermi level location.

We have made measurements of electron drift in the presence of optical bias which appear inconsistent with this assumption. This research is in collaboration with Dr. Daxing Han and Dr. Marvin Silver at University of North Carolina, Chapel Hill. We have proposed that relaxation of a deep level between differing atomic configurations may explain the effect. As a consequence the "trap depth" describing emission depends upon the defect's history of charge state transitions. Such a breakdown in Shockley Read kinetics goes a long way towards resolving apparent discrepancies in the interpretation of electronic characterization studies in a-Si:H performed under different conditions.

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1. INTRODUCTION

During Phase II of this research project we have made significant progress in research in two areas: electron drift and conduction bandtails in a-$Si_{1-x}Ge_x$:H and a-$Si_{1-x}C_x$:H alloys, and optical bias effects on electron trapping and emission in a-Si:H.

We have completed a comprehensive study of electron drift in a-$Si_{1-x}Ge_x$:H alloys, including correlations of the drift parameters with "blue" fill factor measurements and open-circuit voltages in solar cells. Our results suggest that the degradation of the electron mobility with Ge alloying may be intrinsic to bandgap narrowing. It this interpretation is correct, the effects are not likely to be greatly affected by improvements in deposition technology. On the other hand we found little if any indication that V_{OC} is adversely affected by the broadened conduction bandtail in solar cells.

We commenced a study of electron and hole drift in a-$Si_{1-x}C_x$:H. At present we have confirmed only that C alloying does degrade the electron drift mobility in a-$Si_{1-x}C_x$:H.

We also completed a study of optical bias effects on transient photocharge measurements in a-Si:H. These studies clarify that optical bias effects found in earlier photocurrent transient measurements occur because deep-trapping is suppressed by optical bias. We do not believe that these results are consistent with the standard, "Shockley-Read" analysis of photoconductivity, and we have proposed instead that they reflect metastable changes in the atomic configuration of defects in a-Si:H following photocarrier capture and emission events. Most a-Si:H solar cell models are based on Shockley-Read kinetics, so this proposal may lead to some refocusing of modeling efforts.

1.1 Annotated Bibliography of 1992 Publications

In addition to these new projects a number of papers were published in 1992 summarizing research done primarily in previous years. These include:

1. "Electron drift mobility measurements on annealed and light-soaked hydrogenated amorphous silicon," Qi Wang, Homer Antoniadis, and E. A. Schiff, *Appl. Phys. Lett.* **60**, 2791-2793 (1992). We found little effect of light soaking on electron mobility in a-Si:H.
2. "Modulated Electron-Spin-Resonance Measurements and Defect Correlation Energies in Amorphous Silicon," J.-K. Lee and E. A. Schiff, *Phys. Rev. Lett.* **68**, 2972-2975. We believe that a correlation energy of 0.3 eV is satisfactory for the *D*-center in a-Si:H.
3. "Transient photocharge measurements and electron emission from deep levels in undoped a-Si:H," Homer Antoniadis and E. A. Schiff, *Phys. Rev. B* **46**, 9482-9492 (1992). We found a new technique for measuring deep trap emission times in a-Si:H, and reported an apparent "Meyer-Neldel" rule connecting the trap depth and escape frequency.
4. "Effects of hydrogen plasma treatment on hydrogenated amorphous silicon," J.-K. Lee and E. A. Schiff, in *Amorphous Silicon Technology - 1992*, edited by M. J. Thompson, *et al* (Materials Research Society, Pittsburgh, 1992), pp. 185-190. We conducted a rough experiment attempting high temperature plasma hydrogenation of as-deposited a-Si:H. No improvements in electronic properties were noted, and in fact we speculated that the plasma may have dehydrogenated specimens more rapidly than thermal treatment alone would indicate.

5. "Delayed field transient photocurrent measurements in a-Si:H," Homer Antoniadis and E. A. Schiff, in *Amorphous Silicon Technology - 1992*, *loc. cit.*, pp. 783 - 788. We believe this technique may be promising for probing deep level effects in a-Si:H, but no remarkable effects were found in this preliminary study.
6. "Role of Hydrogen Microstructure in Amorphous Silicon," Sufi Zafar and E. A. Schiff, in *Amorphous Silicon Technology - 1992*, *loc. cit*, pp. 199-209. A review of previous work connecting the clustered and dilute phases of bonded hydrogen in a-Si:H to its electronic defects.

2. CONDUCTION BANDTAILS AND ELECTRON DRIFT MOBILITY

2.1 Measurements in a-$Si_{1-x}Ge_x$:H Solar Cells.

Our research on electron transport in a-$Si_{1-x}Ge_x$:H solar cells addresses two important issues:

- Are the electron mobility and conduction bandtail width in a-$Si_{1-x}Ge_x$:H significantly limiting V_{OC} or FF in a-$Si_{1-x}Ge_x$:H solar cells?
- Are the electron mobility and conduction bandtail width in current a-$Si_{1-x}Ge_x$:H materials intrinsic, or can improvements be anticipated from further deposition research?

Some of the work reported here has been published [1]. The specimens were deposited at Energy Conversion Devices, Inc. using plasma deposition techniques. Five types of material were studied spanning a range of about 0.3 eV in optical gap. The specific gas mixtures, substrate temperatures, and plasma parameters were varied in an effort to obtain optimized solar cells; there was no intentional doping of the i-layers of the films.

Table I: Summary of the properties of a-$Si_{1-x}Ge_x$:H specimens. d is the thickness of the i-layer, E_T is the Tauc Gap, V_{oc} is the open circuit voltage measured in conditions comparable to AM 1.5 (100 mW/cm^2), and FF (Blue) is the fill factor measured using a $\lambda < 450$ nm filter. Several parameters were estimated using transient photocurrent techniques: $\mathcal{E}_o$ is the conduction bandtail width, $\mu\tau_{e,t}$ is the electron deep trapping mobility lifetime product, and $\mu\tau_{h,t}$ is the hole deep trapping mobility lifetime product.

Specimen (a-$Si_{1-x}Ge_x$:H)	A (x=0)	B (x=10)	C (x=25)	D (x=50)	E
d (μm)	0.32	0.32	0.32	0.32	0.32
E_T (eV)	1.72	1.60	1.55	1.47	1.42
V_{oc} (V)	0.94	0.82	0.74	0.68	0.58
FF (Blue)	0.75	0.70	0.62	0.60	0.5
d (μm)	2.65	0.95	1.25	1.00	
$\mathcal{E}_o$ (meV)	20	25	32	41	
$\mu\tau_{e,t}$(cm^2/V)	6.2x10^{-8}	1.6x10^{-8}	9.2x10^{-9}	5.1x10^{-9}	
$\mu\tau_{h,t}$(cm^2/V)	6.1x10^{-8}	-	1.4x10^{-9}	5.5x10^{-10}	

For each type of material (denoted A-E in Table I) three or more specimens of varying thickness were deposited. The optical transmittance was measured for films about 1 μm

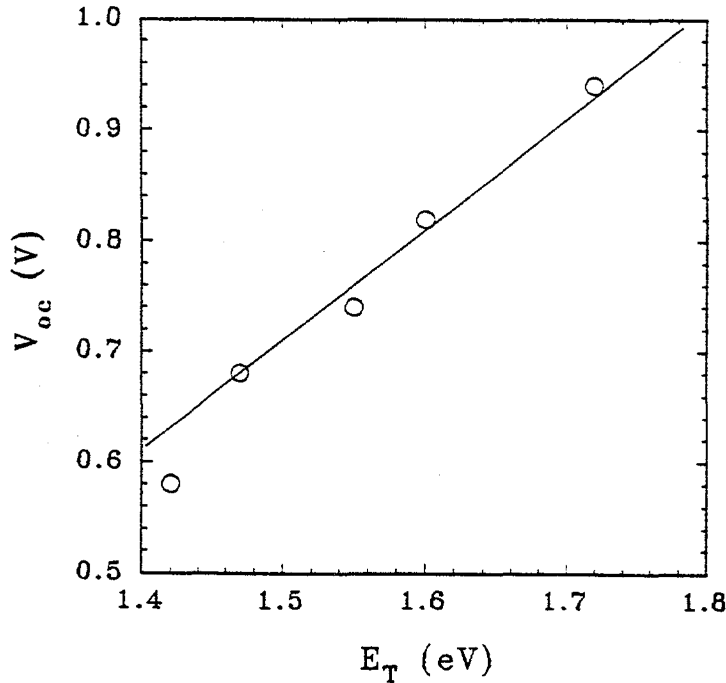

Fig. 2-1: Correlation of the open circuit voltage V_{OC} with the bandgap E_T for five a-$Si_{1-x}Ge_x$:H p-i-n solar cells. The solid line represents the model that V_{OC} and E_T change by equal increments as the alloy parameter x increases.

thick deposited onto glass. We estimated a bandgap E_T using the "Tauc" procedure; these are given in Table I. We also estimated the Urbach parameter $\mathcal{E}_U$ from these measurements; we found little variation between specimens. The results may be summarized by $\mathcal{E}_U = 47 \pm 3$ meV.

Solar cell parameters were measured on specimens with fairly thin i-layers as indicated in Table I. The open circuit voltage V_{OC} was measured using an illuminator corresponding approximately to "air mass" AM 1.5 (100 mW/cm^2). The diode's current-voltage relation was also measured with this illuminator and a "blue" optical filter ($\lambda < 450$ nm). The fill factor measured under these conditions is presented in Table I. Our experience with solar cells fabricated with differing p-i and n-i interfaces suggests that this blue fill factor is primarily determined by electron transport properties in the i-layer, and is not interface limited.

Transport properties were measured on p-i-n diode structures; the p and n layer thicknesses were 200 Å. We performed transient photocurrent measurements on thicker structures as noted in Table I. The "deep-trapping" mobility-lifetime products for electrons and holes ($\mu\tau_{e,t}$ and $\mu\tau_{h,t}$, respectively) were estimated using standard "charge-collection" procedures [2]. We also measured the temperature-dependent electron drift mobility in these structures using time-of-flight techniques. These latter measurements were broadly consistent with the standard "multiple-trapping in exponential bandtail" model, which uses the following parameters: an effective band mobility μ_0, an escape frquency ν, and a bandtail width $\mathcal{E}_0$. We found adequate agreement with experiment by choosing *specimen independent* values for μ_0 (1 cm^2/Vs) and ν (5×10^{11} s^{-1}). The best fit values for $\mathcal{E}_0$ are

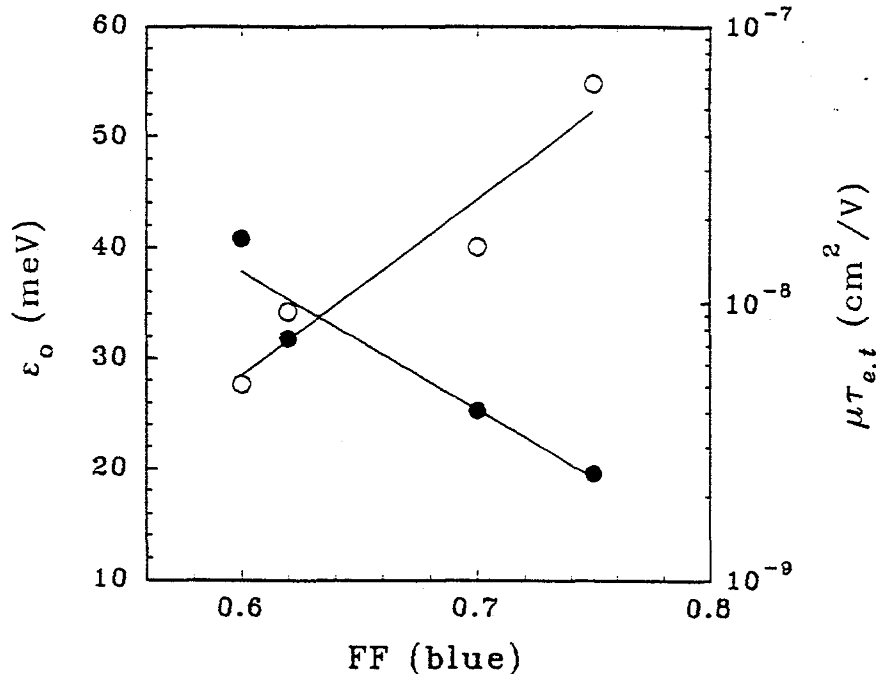

Fig. 2-2: (*solid circles*) Correlation of the conduction bandtail width $\mathcal{E}_0$ estimated from temperature-dependent electron drift mobility measurements with the fill factor measured using blue light. (*open circles*) Correlation of the electron deep trapping mobility lifetime product $\mu\tau_{e,t}$ with the blue fill factor; note the logarithmic scale for $\mu\tau_{e,t}$. Results from solar cells fabricated from four a-$Si_{1-x}Ge_x$:H materials (A-D in Table I) are shown.

given for each type of material in Table I.

In Fig. 2-1 we have graphed the correlation of the AM 1.5 open circuit voltage V_{OC} with the optical bandgap for the five materials. We have also illustrated the fitting

$$V_{OC} = E_T/e - 0.78 \quad , \tag{3}$$

representing the model that V_{OC} is simply shifted from the optical gap by a constant of about 0.8 V for AM 1.5.

This "rigid" shift of V_{OC} with E_T seems remarkable. It suggests that for these specimens the open circuit voltage is determined primarily by the *i*-layer of the films, and was not strongly dependent upon the built-in potential V_{BI} established by the n and p materials used. Since both the conduction bandtail width and the deep level density of a-$Si_{1-x}Ge_x$:H increase rapidly as the optical gap decreased, we conclude that neither of these materials parameters strongly affected V_{OC}.

We now turn to the correlations of the blue fill factor with $\mu\tau_{e,t}$ and with $\mathcal{E}_0$ in Fig. 2-2. It is evident that the fill-factor changes approximately logarithmically with the electron deep-trapping mobility-lifetime product, and approximately linearly with the parameter $\mathcal{E}_0$. It is not obvious which, if either, of these two parameters should be correlated with the fill factor. It seems most reasonable to ascribe the effect to an increase in deep level density, which causes the built-in potential to drop faster and concomitantly degrades $\mu\tau_{e,t}$.

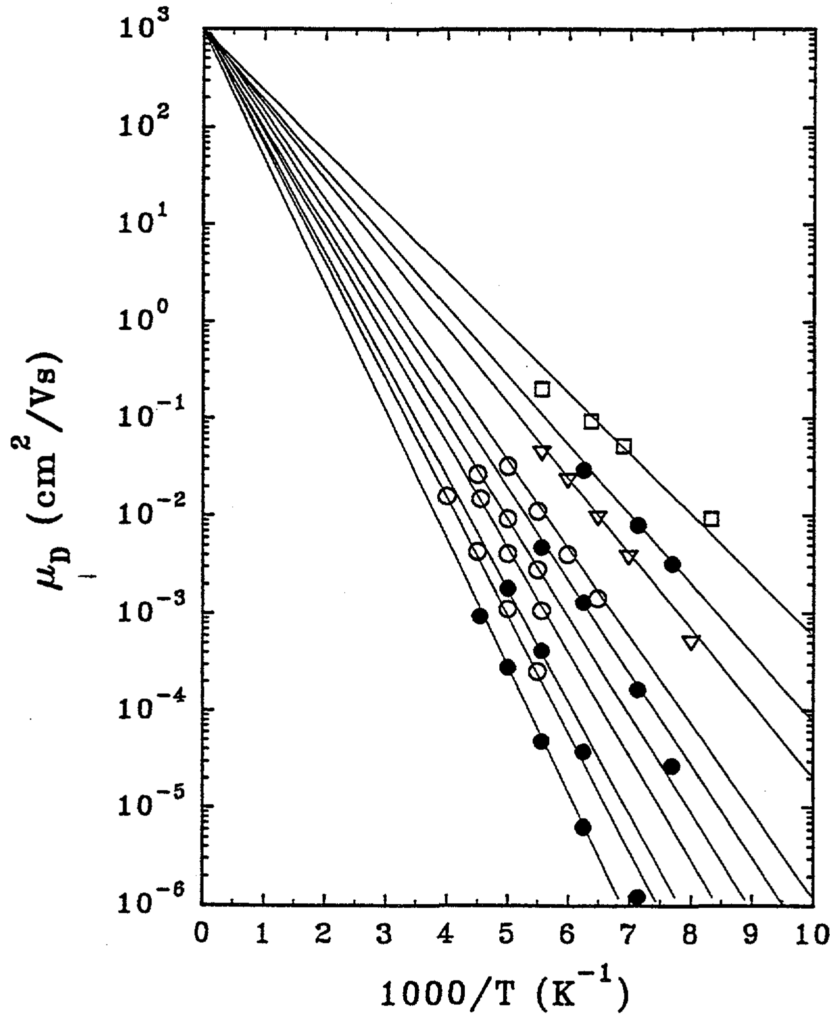

Fig. 2-3: Average electron drift mobility μ_D for ten specimens of a-$Si_{1-x}Ge_x$:H as a function of reciprocal temperature. All points correspond to a ratio $L/E = 2\times10^{-9}$ cm^2/V between the electron displacement and the external electric field. • - this work. ○ - ref. [3]. ▽ - ref. [4]. ⊓ - ref. [5]. The straight lines drawn through the data are fits to the standard exponential bandtail multiple-trapping model. Only one parameter, the conduction bandtail width $\mathcal{E}_{0C}$, was adjusted for each specimen.

Finally, we turn to the question of whether the electron mobility and conduction bandtail in a-$Si_{1-x}Ge_x$:H as measured in the present materials are intrinsic properties of the a-$Si_{1-x}Ge_x$:H network, or might be improved after further deposition research. Our approach to this was to compare drift mobilities in many materials and from several laboratories for a specific, nominal ratio of distance and electric field.

Despite the simplicity of this idea, this program has not been previously executed. The results are presented as Fig. 2-3. Four of the specimens were measured in the present work, and the remaining six specimens are from previously published measurements by other groups. The three upper curves are unalloyed a-Si:H; the successively steeper curves correspond to a-$Si_{1-x}Ge_x$:H specimens of increasing Ge content x and decreasing bandgap.

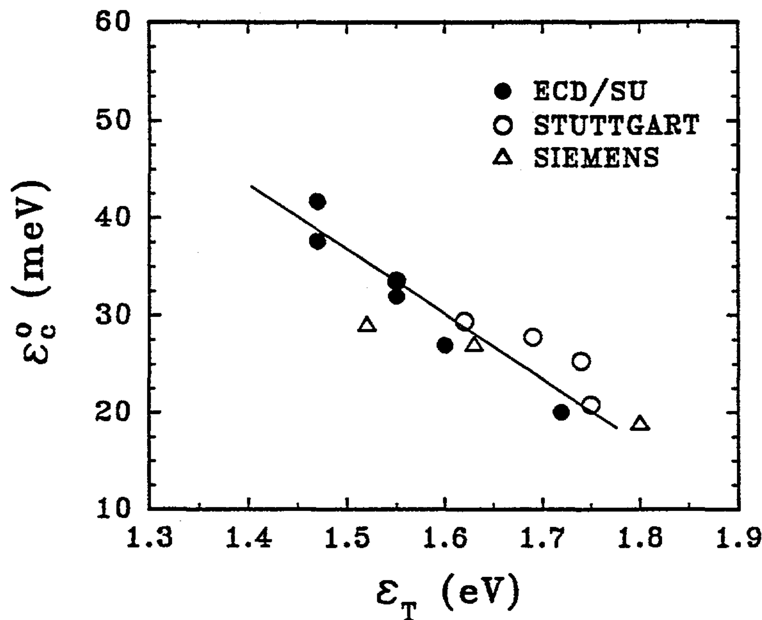

Fig. 2-4: Summary plot of the correlation between conduction bandtail width $\mathcal{E}_{0C}$ and Tauc gap E_T in a-$Si_{1-x}Ge_x$:H. Note the roughly linear relationship as guided by the line. Sources for the data are: ECD/SU , this work; Stuttgart, ref. [3]; Siemens - ref. [6].

The simple pattern which we discern in these data is indicated by the straight lines which focus at $\mu_D = 10^3$ cm^2/Vs for $1/T = 0$. This simple behavior is an extended prediction of the exponential bandtail, multiple-trapping model. The choice we made in Fig. 2-3 to make all the fitting lines converge at $1/T = 0$ means that we kept the attempt-to-escape frequency ν of bandtail traps constant; we were fairly successful in dealing with all available data by keeping the microscopic mobility (μ_0) constant as well, and allowing variation *only* in $\mathcal{E}_{0C}$.

In Fig. 2-4 we have plotted the correlation of $\mathcal{E}_{0C}$ based on this procedure with the bandgap E_T based on Tauc plots. Although there is considerable scatter, the measurements suggest that there is a *linear* relationship between E_T and $\mathcal{E}_{0C}$ in the range shown. More important, perhaps, than the precise form for this relationship is the fact that these data suggest that the conduction bandtail is associated with the optical bandgap. If this interpretation is correct, we suggest that current deposition procedures have essentially reached an intrinsic limit for the narrowness of the conduction bandtail which cannot be further improved. A more complete account of this research is given in ref. [7].

2.2 Measurements in a-$Si_{1-x}C_x$:H Solar Cells

To our knowledge there have been no previous time-of-flight measurements of drift mobilities in a-$Si_{1-x}C_x$:H alloys. The Urbach parameter estimated from optical transmittance studies indicates that C alloying broadens the valence bandtail. The conduction bandtail effects appear completely unknown.

We have measured the temperature-dependent electron drift mobility in a series of hydrogenated amorphous silicon-carbon alloys using time-of-flight. The specimens were p-i-n diodes prepared at Solarex by PECVD. The i-layer was deposited using hydrogen-diluted silane/disilylmethane. The p^+ layer was B doped a-$Si_{1-x}C_x$:H; the $n+$ layer was P doped a-Si:H.

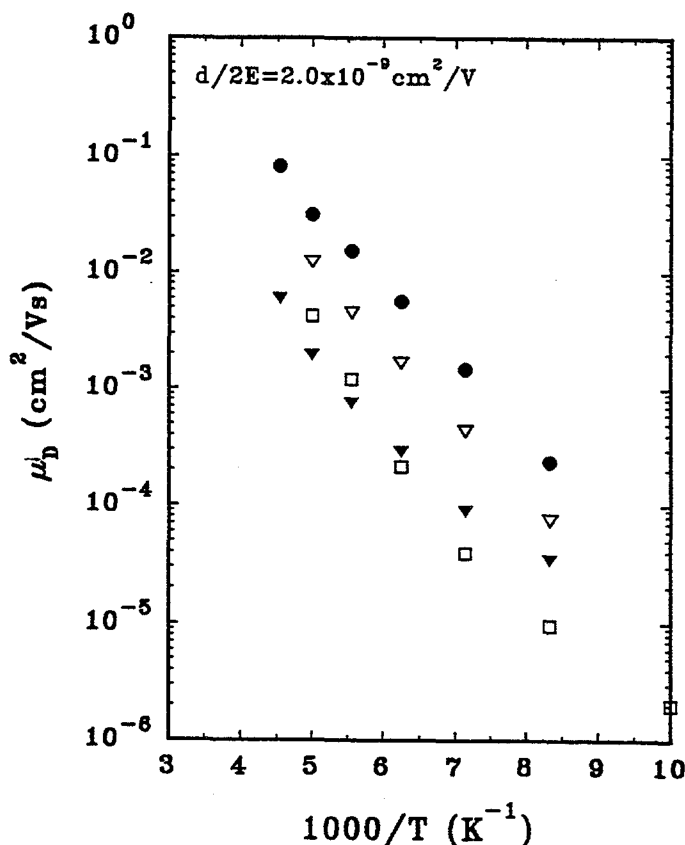

Fig. 2-5: Temperature-dependence of the electron drift mobility in four specimens of a-$Si_{1-x}C_x$:H estimated using time-of-flight techniques ($d/2E = 2 \times 10^{-9}$ cm^2/V). The bandgaps for the four specimens were: • - 1.75 eV (a-Si:H). ▽ - 1.81 eV. ⊓ - 1.88 eV. ▽ - 1.90 eV.

Electron time-of-flight measurements as a function of temperature are presented in Fig. 2-5 for the same ratio $d/2E = 2 \times 10^{-9}$ cm^2/V of thickness and field we used to compare a-$Si_{1-x}Ge_x$:H materials. As the bandgap increased due to carbon alloying the electron drift mobility decreased by as much as a factor 30 at some temperatures. The 1.75 eV bandgap specimen (a-Si:H) and the 1.81 eV specimens have thermally activated drift mobilities over the temperature range 100 - 200 K, corresponding to simple multiple-trapping behavior. We have not attempted to force-fit these data to obtain conduction

bandtail width parameters. Specimens with bandgaps near 1.90 eV did not have simply activated drift mobilities; we have not accounted for this behavior, but it suggests that the bandtail broadening description used to account for the effects of germanium alloying on the electron drift mobility may not be simply applicable to carbon alloying.

Electron and hole time-of-flight measurements on additional a-$Si_{1-x}C_x$:H specimens are planned for 1993.

3. OPTICAL BIAS EFFECTS AND SHOCKLEY-READ KINETICS IN a-Si:H

In this section we describe some measurements of the transient photocharge $Q(t)$ measured using coplanar electrodes on a-Si:H films. The transient photocharge technique clearly reveals the time evolution of an electron photocarrier - from generation to recombination. It thus permits the assumptions of various models for steady-state photocarrier transport to be tested.

Our measurements reveal that an important, "deep-trapping" step in photocarrier evolution found by time-of-flight measurements under near-dark conditions is suppressed when a-Si:H is optically biased. We could not account for this effect using the standard, "Shockley-Read" analysis and the defect density-of-states estimated from time-of-flight techniques. We believe that this discrepancy reveals an important role for metastable defect configurations in determining photocarrier kinetics. Related ideas have been discussed recently for the properties of defects in doped a-Si:H [8].

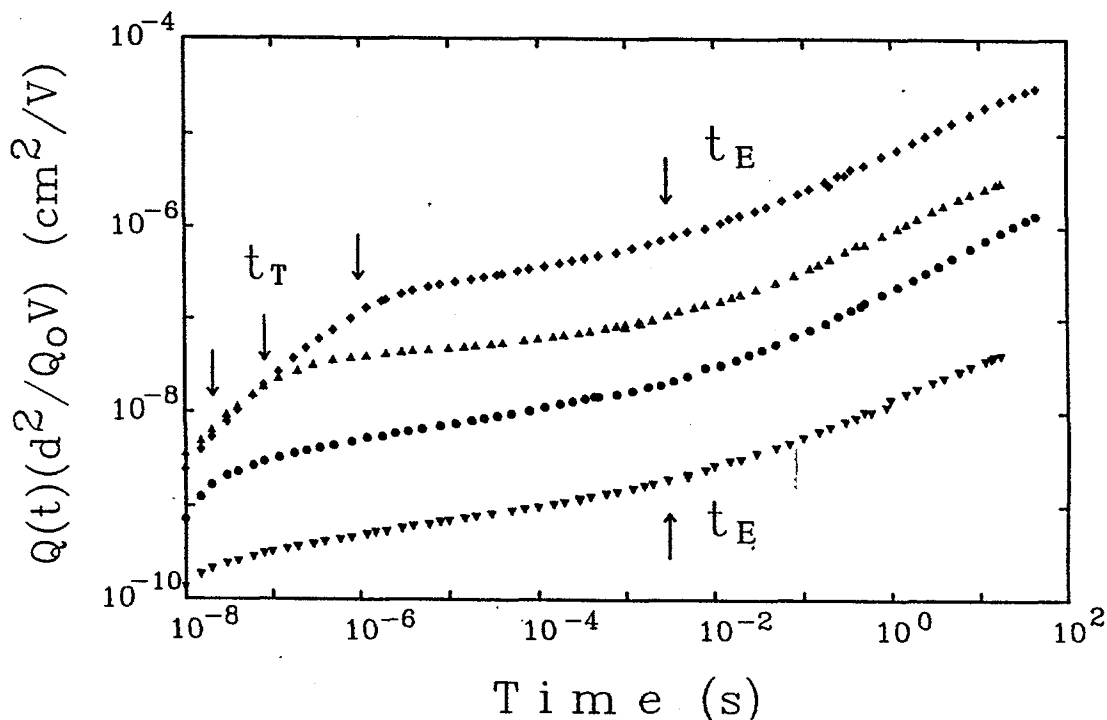

Fig. 3-1: Normalized transient photocharge $Q(t)(d^2/Q_0V)$ at room temperature for four different a-Si:H specimens having defect densities varying between 3×10^{15} and 3×10^{17} cm^{-3}. We indicate trapping times t_T and emission times t_E with arrows.

We comment briefly on the significance of this research for solar cells. Essentially all solar cell models use a Hall-Shockley-Read analysis of photocarrier trapping and recombination. The models seek to find a single, *fixed* density of states which accounts for photoconductivity, optical properties, and other materials characterizations. When defect relaxation effects are large, any fixed density-of-states is probably useful only in a small-signal sense: the density-of-states actually reflects the excitation and history of the material. Thus we would not expect a density-of-states obtained under near dark conditions to

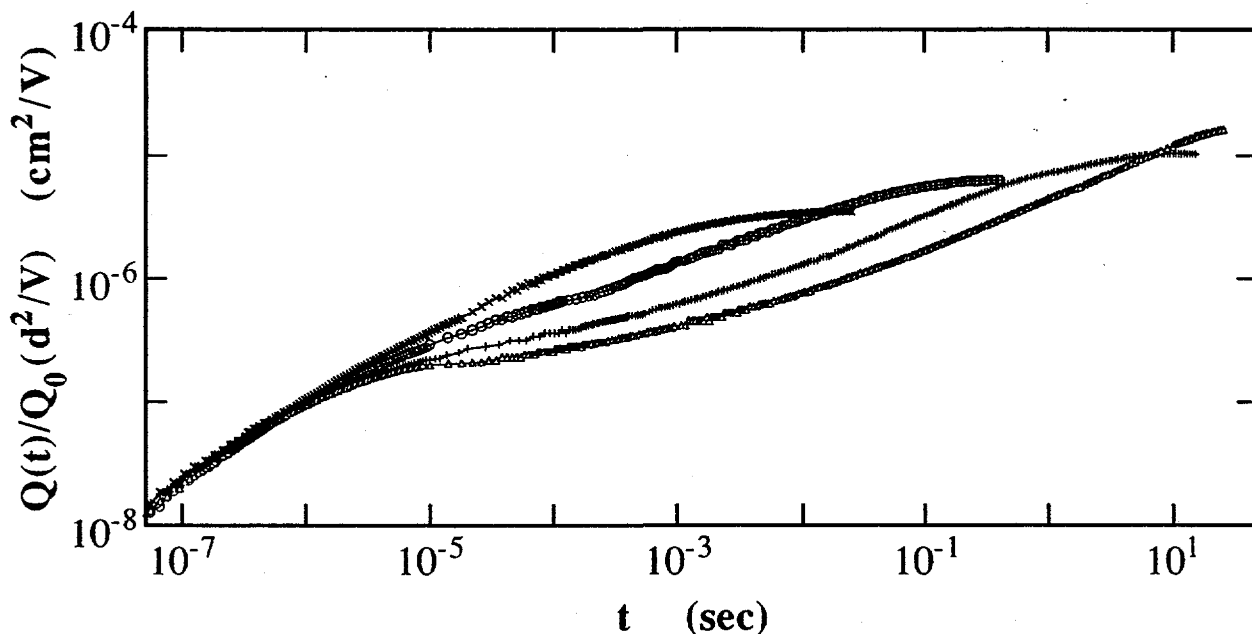

Fig. 3-2: The transient photocharge $Q(t)(d^2/Q_0V)$ recorded at three optical bias levels and in the dark in an a-Si:H specimen in its annealed state. The photogeneration rates were (from upper to lower trace at 10^{-4} s) 7.6×10^{19}, 3.7×10^{18}, and 2.3×10^{17} $cm^{-3}s^{-1}$; the lowest curve was measured without optical bias. Note that optical bias impedes deep trapping, thus enhancing electron drift.

apply under optical bias, or in a solar cell we would not expect the density-of-states under short circuit conditions to be necessarily the same as that under open circuit conditions.

As Fig. 3-1 we present some transient photocharge measurements for four specimens of undoped a-Si:H of widely varying defect density. The measurements show the total photocharge $Q(t)$ which has flowed through the voltage bias circuit of the specimen following a laser pulse. For these data the specimens were not optically biased. The specimens were at room-temperature and had coplanar electrodes; dark currents in the specimen were subtracted from photocurrents before the photocharge was calculated. The photocharge data have been normalized using the bias Voltage V between the electrodes, the interelectrode gap d, and the magnitude Q_0 of the charge of electrons photogenerated by the laser. Fig. 3-1 is taken from a recent paper by Antoniadis and Schiff [8], which should be consulted for further details.

There are three distinct regions to these transient measurements. In the first regime the photocharge is nearly proportional to the delay time t, as expected for ordinary, "non-dispersive" transport; this regime is only clear for the upper two curves. A mobility of order 1 cm^2/Vs can be calculated from these measurements. The second regime is a broad plateau, lasting until about 1 ms for all four specimens. This plateau reflects the aftermath of "deep trapping" of electrons by defects in a-Si:H, and has been extensively studied before using time-of-flight techniques. The upturn in the transient photocharge following 1 ms is due to re-emission of electrons from their deep traps. A final plateau in the photocharge

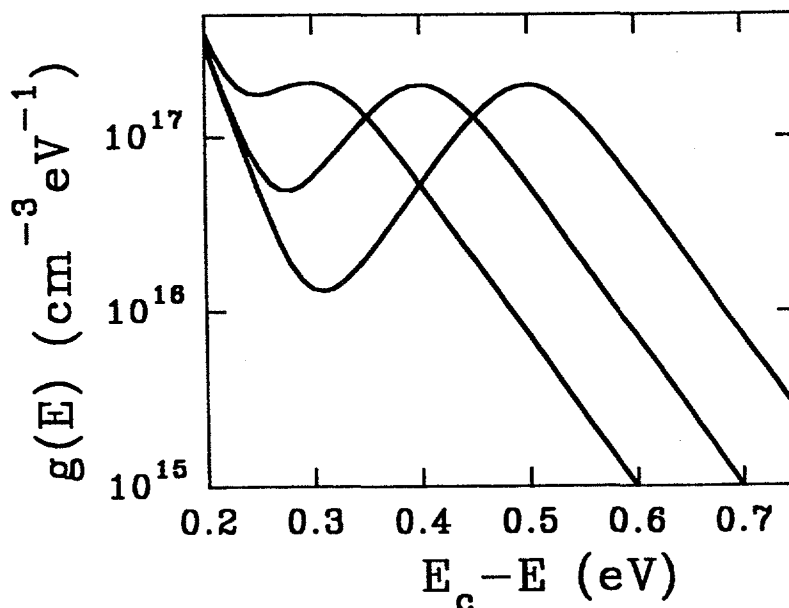

Fig. 3-3: A possible model for the density of states probed by transient measurements under different levels of optical bias. The peak of the defect distribution becomes shallower, reflecting the unrelaxed states of the defect when it traps electrons.

due to recombination is not evident in this figure, but was observed at higher specimen temperatures.

In Fig. 3-2 we illustrate how the transient photocharge is affected by optical bias. The transient photocharge is recorded using techniques similar to those used to obtain Fig. 1. However, the steady-state photocurrent through the specimen due to the optical bias is sunk into a constant current source. The lowest curve was taken without optical bias; the three successively higher curves show the effects of increasing optical bias. The curve without optical bias shows the "deep-trapping" plateau, but successively higher levels of bias essentially "fill in" the plateau. Such an effect requires either that all of the deep-traps are filled under optical bias, or that the apparent density-of-states associated with the deep traps shifts to such shallow energies that it loses its role as a deep trap.

We could not account satisfactorily for this effect using "trap filling" and quasi-Fermi level motion. Instead we favor the second possibility noted above: that the effective trap density-of-states itself changes under optical bias. This model is illustrated schematically in Fig. 3-3, where we show the peak of the deep level moving towards the conduction band for higher levels of optical bias. We speculate that the deep levels are rearranged atomically following charge capture or emission, and that these rearrangements relax sufficiently slowly that the defect system's average atomic configuration changes under optical bias. A manuscript reporting these measurements and the related discussion at greater length is currently submitted for publication.

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This report describes Phase 2 progress in two areas: (1) electron drift and conduction band tails in a-$Si_{1-x}Ge_x$:H and a-$Si_{1-x}C_x$:H alloys, and (2) optical bias effects on electron trapping and emission in a-Si:H. We completed a comprehensive study of electron drift in a-$Si_{1-x}Ge_x$:H alloys, including correlations of the drift parameters with "blue" fill factor measurements and open-circuit voltages in solar cells. We began a study of electron and hole drift in a-$Si_{1-x}C_x$:H, but, at present, we have only confirmed that C alloying degrades the electron drift mobility in a-$Si_{1-x}C_x$:H. We completed a study of optical bias effects on transient photocharge measurements in a-Si:H, which clarified that optical bias effects found in earlier photocurrent transient measurements occur because deep trapping is suppressed by optical bias.

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