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Hole Drift Mobility Measurements on a-Si:H using Surface and Uniformly Absorbed Illumination

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ABSTRACT: The standard, time-of-flight method for measuring drift mobilities in semiconductors uses strongly absorbed illumination to create a sheet of photocarriers near an electrode interface. This method is problematic for solar cells deposited onto opaque substrates, and in particular cannot be used for hole photocarriers in hydrogenated amorphous silicon (a-Si:H) solar cells using stainless steel substrates. In this paper we report on the extension of the time-of-flight method that uses weakly absorbed illumination. We measured hole drift-mobilities on seven a-Si:H *nip* solar cells using strongly and weakly absorbed illumination incident through the *n*-layer. For thinner devices from two laboratories, the drift-mobilities agreed with each other to within a random error of about 15%. For thicker devices from United Solar, the drift-mobilities were about twice as large when measured using strongly absorbed illumination. We propose that this effect is due to a mobility profile in the intrinsic absorber layer in which the mobility decreases for increasing distance from the substrate.

INTRODUCTION

Hole drift mobilities are crucial to understanding amorphous silicon (a-Si:H) solar cells [1]. Experimentally, electron and hole drift mobilities are generally measured using the time-of-flight technique in which a pulse of illumination is absorbed near an electrode interface. Depending on the direction of the electric field, electrons or holes are swept across the structure, and an average drift-mobility μ_d is calculated from their transit time t_T. To measure holes in a-Si:H solar cells, the standard method requires fairly strong illumination through the *n*-layer to create a sheet of carriers near the *n/i* interface. One typically uses a 500 nm wavelength and reverse electrical bias on the cell. Such illumination is not possible for a-Si:H *nip* solar cells deposited onto opaque substrates such as stainless steel, and an alternative would be very desirable [2].
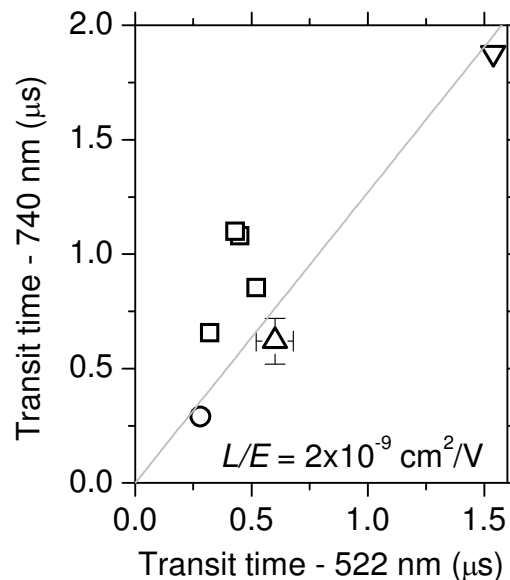

Figure1: Correlation of the transit times obtained with 740 nm illumination with transit times obtained with 522nm for seven a-Si:H cells. The symbol shapes indicate the sample substrate: □ US, ○ PSU, ($\triangledown \triangle$) BP1, BP2. The line represents the ratio predicted by Arkhipov, *et al.* (see text). The error bar was determined from multiple measurements on one device.

One alternative is to use uniformly absorbed illumination, which leads to two complications *vis a vis* conventional time-of-flight. The first is that both electron and hole motions contribute to the photocurrent. Because electrons are far more mobile than holes in a-Si:H, their half of the photocharge is swept out quickly, and the long-time photocurrents are dominated by holes. The second difficulty is that the initial positions of the holes are uniformly distributed; as we show later, a hole transit-time t_T can be measured even for uniform photoexcitation; the standard expression for the drift-mobility is:

$$\mu_D = L/(Et_T) \tag{1}$$

where L is the average displacement of the carriers at the transit-time and E is the electric field. We discuss the relation of L to the intrinsic layer thickness d later in this paper.

While this method should be suitable to measurements on samples with opaque substrates, it has never been carefully tested; we do this testing in the present paper. The method must accommodate the fact that holes in a-Si:H exhibit "anomalous dispersion" in their motions. When anomalous dispersion obtains, drift mobilities for different materials or different techniques must be compared for a specific value of the ratio L/E [3]. With this proviso, the work of Arkhipov, *et al.* [4,5] shows that one expects a non-unity ratio of the transit times for weakly absorbed light t_T^u to the conventional estimate based on strongly absorbed light t_T^s:

$$\frac{t_T^u}{t_T^s} = (4/3)^{1/(2\alpha)} \tag{2}$$

where α is the "dispersion parameter". For holes near room-temperature, $\alpha \approx 0.6$, so the predicted ratio is 1.3. The derivation assumes that hole transport properties are uniform throughout the material.

To our knowledge, there have been no experimental tests of this prediction. In Figure 1 we show a summary of room-temperature measurements of the transit time for seven a-Si:H devices at approximately the displacement to field (L/E) ratio of 2×10^{-9} cm^2/V. Several of the devices are consistent with the Arkhipov, *et al.* ratio of 1.27 (see the error bar for one point). However, some of the devices yielded transit times with ratios that are systematically larger than expected. We believe that this behavior is probably evidence for a hole mobility that declines for positions that are increasingly distant from the substrate. The possibility of inferring a mobility profile is an unexpected outcome of the present work. In addition, our results suggest that drift mobilities measured in special, thick samples may not be representative of thinner samples.

SPECIMENS

We studied seven a-Si:H *pin* devices on four substrates. Four of the devices were on one TCO-coated glass substrate prepared at United Solar Ovonic LLC (*nip* deposition sequence, VHF deposition, 2.0 µm a-Si:H intrinsic layer, no evidence for microcrystallinity). A second sample was prepared at Pennsylvania State University (*pin* deposition sequence, specular TCO as substrate, 0.59 µm a-Si:H intrinsic layer, semitransparent top Cr contact). Two additional samples were made in 2002 at BP Solar, Inc. (*pin* deposition sequence, DC plasma). BP1 was made with a hydrogen/silane dilution ratio of 10 (thickness 0.89 µm); BP2 was made with a hydrogen dilution of 20 (intrinsic layer thickness 1.13 µm). A semitransparent ZnO top electrode was deposited instead of the usual metal back reflector.

HOLE DRIFT MOBILITY MEASUREMENTS

The transient photocurrents measurements were done using a 4 ns illumination pulse from nitrogen laser-pumped dye laser. The devices were illuminated through their n-layers. The laser wavelengths used were 522 nm and 740 nm, corresponding to absorption depths of about 0.08 µm and 30 µm, respectively [6].

Fig. 2 (a) & (c) illustrates the transient photocurrents at 293 K at the two wavelengths for devices from US and BP1, respectively. The photocurrents are normalized as $i(t)d^2/Q_0(V+V_{bi})$ where d is the intrinsic-layer thickness, V is the applied external bias, V_{bi} is a correction for the built-in potential, and Q_0 is the total photocharge generated in the intrinsic layer. We made a correction for the built-in potential V_{bi} of the *pin* structures [7].
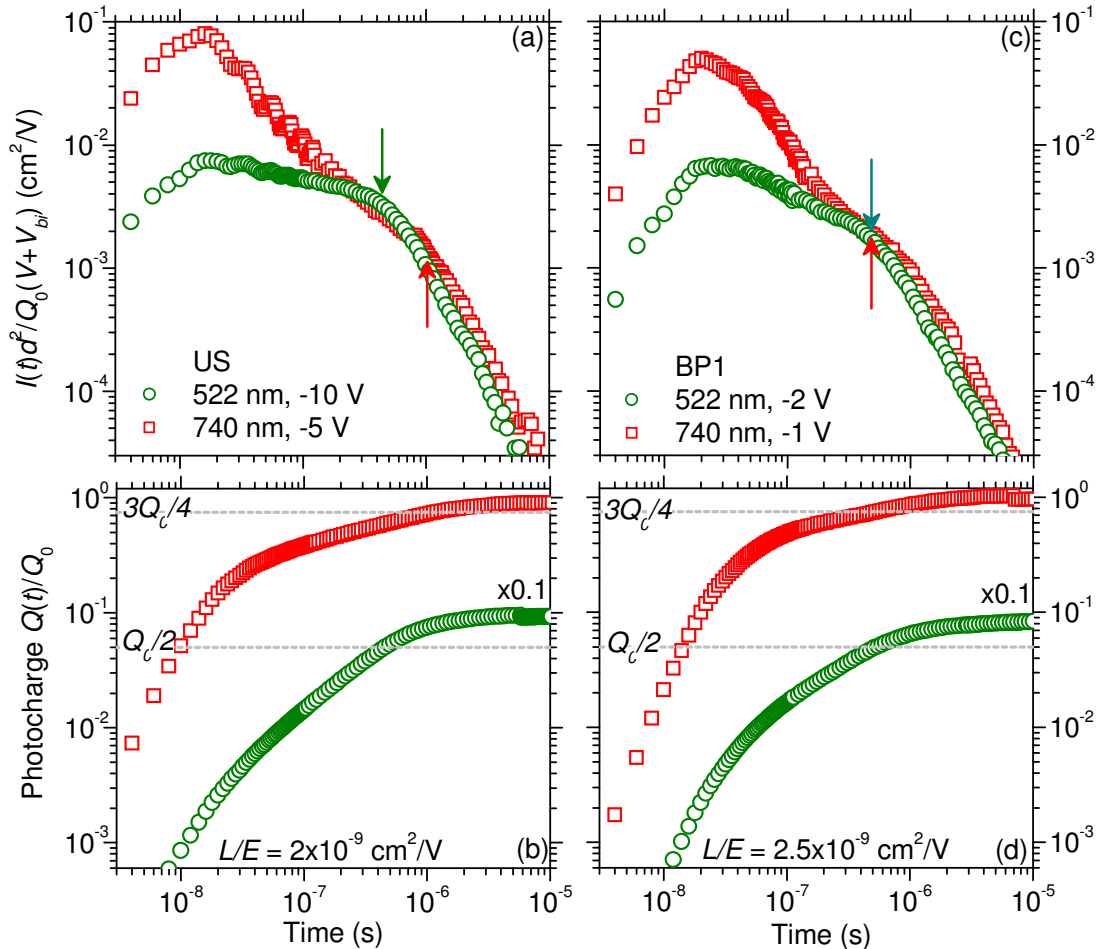

Figure 2: (a) & (c) Normalized transient photocurrents $i(t)d^2/Q_0(V+V_{bi})$ measured using two illumination wavelengths in a-Si:H *p-i-n* devices at 293 K; see text for the normalization procedure. The total photocharge Q_0 is defined as the total photocharge collected at longer times and larger bias voltages. (b) & (d) Normalized photocharge transients $Q(t)/Q_0$ obtained by time-integration of the transient photocurrent. The intersection of the transients with the horizontal lines at $Q_0/2$ (522 nm), and $3Q_0/4$ (740 nm) were used to determine the hole transit times t_T; the arrows on the upper panels indicate the resulting transit times.

For 522 nm wavelength, which is strongly absorbed near the *n/i* interface, the photocurrents are dominated by hole drift. There is a noticeable "kink" marking a transition from a shallow, power-law decay to a steeper decay that is typically identified as the hole transit time

t_T. As illustrated by the photocharge transients in the lower panels, we actually identify the transit time as the time at which half of the ultimate photocharge Q_0 has been reached. We have indicated the transit times in the upper panels with arrows, where they agree fairly well with the "kink" location. The "kink" and "half-charge" procedures are usually equivalent; we prefer the latter because we find it more reproducible [3].

Figure 3: Hole drift mobilities μ_d as a function of reciprocal temperature $1000/T$ for several experiments. Symbols are estimates for a United Solar device measured at two illumination wavelengths; the lines through these are multiple trapping fit using the parameters $\mu_p = 0.8$ cm^2/Vs, $\Delta E_V = 43$ meV, and $\nu = 0.8 \times 10^{12}$ s^{-1} (522 nm) and 1.7×10^{12} s^{-1} (740 nm). Palaiseau03-ref. 10, Stuttgart91-ref. 11.

For the 740 nm illumination the photocarriers are created uniformly throughout the a-Si:H film. Half of the total photocharge Q_0 is due to the electrons; since electrons in a-Si:H have a drift mobility at least 10^2 larger than holes [8], this fraction of the photocharge is collected in less than 100 ns; the photocharge in excess of $Q_0/2$ is due to holes, and the transit time for hole collection is thus reached at $(3/4)Q_0$. As can be seen in Fig. 2, this agrees reasonable well with the "kink" in the long-time transients.

For 740 nm, we have shown the transient at half the voltage we used for 522 nm; this halving compensates for the fact that the initial, mean position of the holes for uniform absorption is already halfway across the sample, so the transit times for the 2 wavelengths should be about the same. This procedure was assumed by eq. (2) above.

In Fig. 2, for the sample BP1 there is little difference in the transit-time estimates using 540 and 722 nm wavelengths, but there is about a factor two difference for the US sample. In Fig. 3, we illustrate the temperature-dependence of the drift-mobility for this cell for both wavelengths. For the uniformly absorbed, 740 nm data we have incorporated the results of Arkhipov, *et al*. (cf. eq. (2)) by using the definition for the drift-mobility:

$$\mu_D^u = (4/3)^{1/2\alpha} \left(\frac{L}{E t_T^u} \right) \tag{3}$$

Fits to these data using bandtail multiple-trapping line are also illustrated (see ref. [9] for procedures); these are based on the entire set of measurements, not just those illustrated in Fig. 3. For reference, we also present two lines based on previously published, conventional time-of-flight measurements on a "polymorphous" silicon sample [10] (denoted Palaiseau03) and some measurements on a sample from Universität Stuttgart [11].

DISCUSSION

In order to address the dependence of these drift-mobility estimates on the illumination wavelength, we again consider the transit time estimates illustrated in Figure 1. The transit times
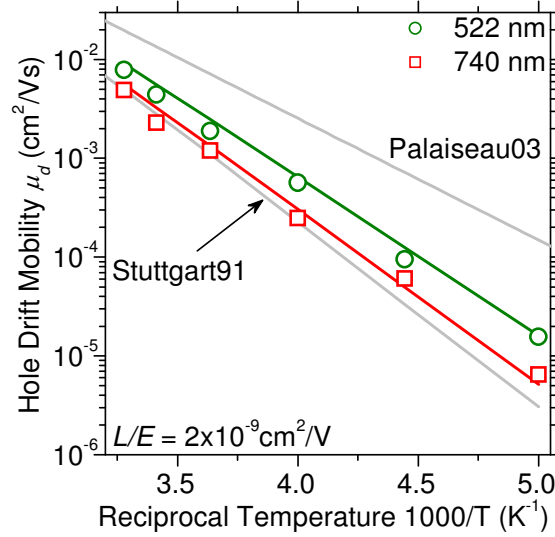
measured in the BP1, BP2, and PSU samples are reasonably consistent with the Arkhipov ratio as shown; for these devices, the dispersion parameters ranged from 0.55 to 0.66, corresponding to Arkhipov ratios of 1.30 to 1.27. For these three devices the Arkhipov, *et al.* theory appears to be a better description than the naïve ratio of unity. However, the United Solar devices have distinctly larger ratios than predicted by Arkhipov, *et al.* This difference cannot be accounted for by differences in dispersion; the United Solar samples had a similar range of dispersion parameters to the other samples.

We thus believe that the measurements for the PSU and BP devices are reasonably consistent with the conventional theory of dispersive transport, which assumes that hole transport properties are constant throughout the thickness of a material. For the US samples, which were considerably thicker than the PSU and BP samples, we believe the hole drift-mobility declines for larger distances from the substrate and the *n/i* interface. We fitted both sets of measurements in one US sample to the bandtail multiple-trapping model; a satisfactory fit was obtained if we increased the attempt-frequency ν about threefold to fit the measurements with uniformly absorbed illumination. Changes in ν are expected due to changes in the fundamental disorder through the bandedge density-of-states N_V [12], although an equally plausible argument could be made that increased disorder should have broaden the bandtail. In either case, the present data are reversed from expectations from a changeover in structure from amorphous to microcrystalline for thicker materials [13] as the film grew thicker.

ACKNOWLEDGMENTS

APPENDIX

In this appendix we explain how we used the calculations of Arkhipov, *et al.* to obtain eq. (2). These authors assumed that the photocurrent transients are governed by multiple-trapping in an exponential bandtail, although we believe the result of eq. (2) to be a general property of anomalously dispersive transport. For strongly absorbed illumination that generates a sheet of carriers near one electrode, they obtained the following equation (eq. (34) of ref. [4]) for the transit-time with strongly absorbed illumination:

$$t_T^s = \left(K/v\right)\left(\frac{vd}{\sqrt{2}\mu E}\right)^{1/\alpha} \tag{4}$$

where K is a dimensionless function of order unity, μ is the band mobility of the carrier, ν is the rate of bandtail trapping for a mobile carrier, and d the sample thickness. As before, α is the dispersion parameter. These authors defined the transit time as the "kink" in the transient photocurrent; the half-charge method used above is expected to be equivalent [3]. Arkhipov, *et al.* subsequently published a comparable expression for the transit time for uniformly absorbed illumination (eq. (26) of ref. [5]):

$$t_T^u = \left(K/\nu\right)\left(\frac{\nu d}{\sqrt{6}\mu E}\right)^{1/\alpha} \tag{5}$$

These two expressions do not correspond to the same mean displacements of the carriers. For uniformly distributed carriers, the displacement at the transit time is half of that for strongly absorbed illumination, since the carriers initially have a mean position that is halfway across the sample. As noted earlier, to compare corresponding drift-mobilities one must use the same displacements L, or more precisely of L/E [3]. Our definition of the transit-time corresponds to $d = 2L$ for strongly absorbed illumination [3], and thus to $d = 4L$ for uniformly absorbed illumination. Substituting into (4) and (5), we obtain:

$$t_T^s = \left(K/\nu\right)\left(\frac{\sqrt{2}\nu L}{\mu E}\right)^{1/\alpha} \text{ and} \tag{6}$$

$$t_T^u = \left(K/\nu\right)\left(\frac{2\sqrt{2}\nu L}{\sqrt{3}\mu E}\right)^{1/\alpha} \tag{7}$$

The ratio of the transit-time for uniform illumination to that with strong illumination – at constant displacement L – is thus $\left(4/3\right)^{1/2\alpha}$, as used in eq. (2).

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