Picosecond Optical Time-of-Flight Studies of Carrier Transport in a-Si:H–a-SiN$_x$:H Multilayers

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We report time-of-flight experiments in the time range from 0.2 psec to 1.8 nsec in a-Si:H–a-SiN$_x$:H multilayer structures using a purely optical technique. The transport mechanism of photoexcited carriers is shown to be dispersive and its characteristic parameters are determined in the temperature range 70–300 K.

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The time-of-flight (TOF) technique is a powerful tool for the study of transport in amorphous semiconductors and led to the discovery of dispersive transport in disordered materials.[1] Dispersive transport exhibits photocurrents with power-law decays and the important feature of universality[1]; the current decays depend only through a time constant, the transit time, on the applied electric field and the thickness of the sample, while the power-law exponent is independent of these parameters. TOF experiments have been used extensively to measure the mobility of electrons and holes in amorphous semiconductors.[2,3] However, the time resolution of TOF experiments is limited by the RC response time of the sample and the circuit that is used to measure the current, with a lower limit of about 1 nsec.[3] Better time resolution has been obtained in photoconductivity experiments.[4] It would very valuable for the understanding of the carrier-transport mechanism in amorphous semiconductors to measure the transport of carriers close to the mobility edge, i.e., in the picosecond time range; only a purely optical method can have sufficient resolution.[5]

In this Letter, we report for the first time on TOF experiments in the picosecond time range. We use a-Si:H–a-SiN$_x$:H multilayer structures which are known to have internal electric fields as large as 10^5 V/cm.[6] After photoexcitation, we measured the drift of carriers to the interfaces between the a-Si:H and a-SiN$_x$:H layers and proved that the transport in a-Si:H in the picosecond range is dispersive. We were able to measure the current at temperatures as low as 20 K, compared with 150 K for conventional TOF experiments. From the temperature dependence we determined the transport mechanism; it is dominantly multiple trapping at high temperatures and hopping down at low temperatures.

We measured photoinduced changes in transmission (ΔT) between 0.2 psec and 1.8 nsec using the pump and probe technique. The pump beam excites carriers across the mobility gap, and the delayed probe monitors the change in transmission due to the excited carriers as a function of time. The pump and probe beams were derived from a passively mode-locked ring dye laser[7] which produces 0.1-psec pulses at 620 nm. The energy per pulse was 0.1 nJ, and the repetition rate was 108 MHz. The pump and probe beams were focused onto a 10-μm-diam spot on the sample. We recorded ΔT as a function of time delay between pump and probe on five different time scales to cover the whole time range with sufficient resolution. In a-Si:H, ΔT at 2 eV is proportional to the change in the absorption coefficient ($\Delta\alpha$) when the transmission is less than 10%,[8] and we use $\Delta\alpha$ in this Letter to refer to the experimental results.

The a-Si:H–aSiN$_x$:H multilayers were prepared by plasma-assisted chemical vapor deposition.[9] We measured $\Delta\alpha$ in four samples with silicon layer thicknesses of 39, 78, 126, and 204 Å, respectively. The ratio of the silicon layer thickness d_S to the nitride layer thickness was held constant at 0.85, and the total thickness of the multilayers was about 1 μm. The pump and probe beams were only absorbed in the a-Si:H layer since a-SiN$_x$:H has a band gap of about 3 eV, while the band gap in a-Si:H is 1.8 eV. The density of photoexcited carriers in the a-Si:H layer was estimated to be less than 10^{17} cm^{-3}/pulse.

The picosecond decay of $\Delta\alpha$ in a good quality film of a-Si:H is very slow with a time constant of the order of 1 nsec.[10] We observed much faster decays of $\Delta\alpha$ in a-Si:H–a-SiN$_x$:H multilayers, with time constants that depend strongly on the silicon layer thickness. The recorded responses in these multilayers were first averaged to remove the noise. Then we took the derivative of $\Delta\alpha$ with respect to time; we will show later that this quantity is proportional to the current of carriers to the interfaces. Finally, we spliced the decays together on a logarithmic time scale. A typical result is shown in Fig. 1 for a multilayer with $d_S = 126$ Å at 300 K, between 1 psec and 1.6 nsec. The time range from 0.2 to 1 psec has been omitted, since it takes carriers a few hundred femtoseconds to become trapped in the exponential band-tail states.[11,12]

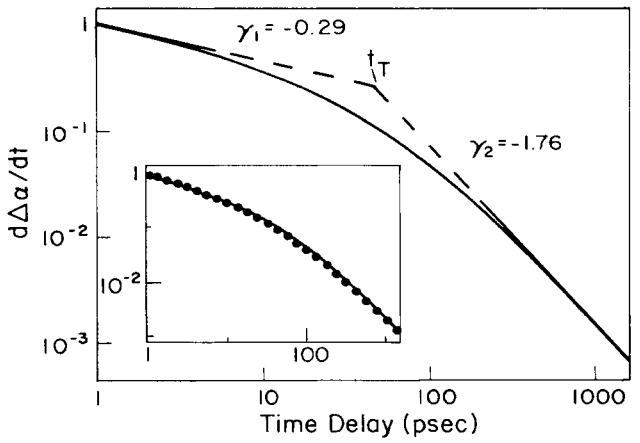

FIG. 1. Time derivative of $\Delta\alpha(t)$ vs time delay for the multilayer with $d_S = 126$ Å at 300 K including the asymptotic power-law branches with exponents γ_1 and γ_2 and the transit time t_T. The inset shows the response for $d_S = 126$ Å at 240 K (dots) and the fit (solid line) to the model described in the text.

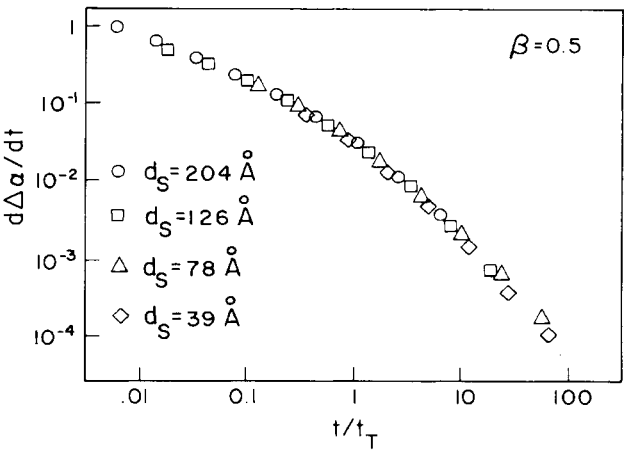

FIG. 2. Master plot of $d\Delta\alpha/dt$ for the a-Si:H–a-SiN$_x$:H multilayers. The transit times are given in Table I.

We clearly observe two power-law decay branches. At short times the exponent is γ_1, which is larger than -1, and at long times it is γ_2, which is smaller than -1. Between these two asymptotic power-law branches, there is a broad transition region from one exponent to the other. γ_1 and γ_2 are temperature dependent above 150 K, but it is important to note that within the experimental accuracy $\gamma_1 + \gamma_2 = -2$ independently of temperature. Because of this relation, γ_1 and γ_2 can be reduced to one parameter β with $\gamma_1 = -1 + \beta_1$, $\gamma_2 = -1 - \beta_2$, and $\beta_1 = \beta_2 = \beta$. The resemblance between the response observed in conventional TOF experiments[3] and Fig. 1 is remarkable. The difference lies in the width of the transition region between the asymptotic power-law branches. In conventional TOF experiments, the transit time t_T is defined as the intersection of the two power-law branches. We used a slightly different definition to determine t_T since for small d_S only the power-law branch with exponent γ_2 is well defined. Since $\gamma_1 > 1$ and $\gamma_2 < -1$, we obtained an average transit time from the measured responses using the equation

$$\frac{d\log[d\Delta\alpha(t)/dt]}{d\log(t)\,|_{t=t_T}} = -1. \qquad (1)$$

Table I contains the transit times for four samples with $\beta = 0.5$. These transit times were used to prove universality by plotting $\log[d\Delta(\alpha)/dt]$ vs $\log(t/t_T)$. Figure 2 shows this master plot for $d_S = 39$, 78, 126, and 204 Å. We clearly observe the universal behavior of dispersive transport.[1,2]

TABLE I. Transit times for different a-Si:H–a-SiN$_x$:H multilayers.

d_S (Å)	t_T (psec)
39	4
78	14
126	72
204	170

Figure 3 shows the temperature dependence of the dispersion parameter β for $d_S = 126$ Å between 70 and 300 K. Above 150 K, the relation $\beta_1 = \beta_2$ is very well obeyed. Below 150 K, it was only possible to determine β_1. The temperature dependence above 150 K can be well described by $\beta = (T - T_1)/T_0$, with $T_0 = 300$ K and $T_1 = 80$ K. Below 150 K, the dispersion parameter reaches a constant value of 0.2 at about 80 K. The samples with $d_S = 39$, 78, and 204 Å show a similar temperature dependence.

The transport of photoexcited carriers is interpreted in

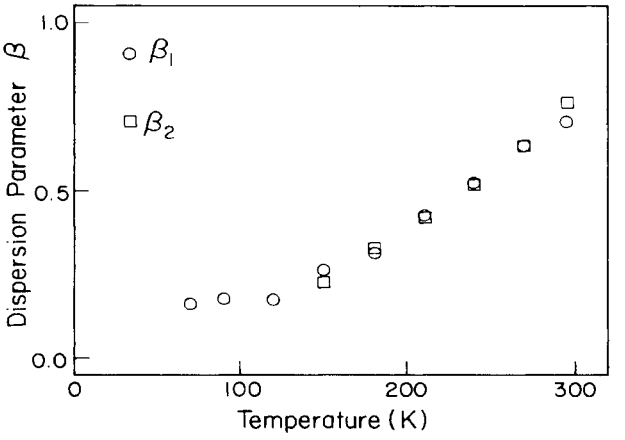

FIG. 3. Dispersion parameter β vs temperature for the multilayer with $d_S = 126$ Å. The relations between β_1 and β_2 and the power-law exponents γ_1 and γ_2 (Fig. 1) are given in the text.

terms of drift to the interfaces due to the internal electric fields. At $t=0$, carriers are excited across the mobility gap leading to an instantaneous change in the absorption coefficient, since the absorption cross sections for the ground and excited states are different.[11,12] A small fraction will drift to the interfaces while most of them are trapped in the band-tail states in the a-Si:H layer within the first picosecond. For times larger than 1 psec, carriers will drift by multiple trapping or hopping to the interfaces, where they are trapped in interfacial-defect states. Since band-tail states and interfacial-defects states have a different absorption cross section at 2 eV,[11] the induced absorption changes every time a carrier arrives at the interface. Consequently, the time-dependent part of $\Delta\alpha$ is proportional to the collected charge at the interfaces, i.e., the derivative of $\Delta\alpha$ with respect to time is proportional to the current of carriers moving to the interfaces $[d\Delta\alpha(t)/dt \propto I(t)]$, which is the measured quantity in regular TOF experiments. One difference between regular TOF and our experiments lies in the mode of excitation. In conventional TOF experiments the light is absorbed in a thin surface layer resulting in a sharply defined transit time. In our experiments we excite carriers homogeneously over the whole layer leading to a wide distribution of transit times. This accounts for the broad transition region between the two power-law branches in our data. Another difference between regular TOF and our experiment is that in conventional TOF only one type of carrier can drift across the film, while in our case both types of carriers, electrons and holes, can move and contribute to the current decay. However, since the internal electric fields in the a-Si:H–a-Si:N_x:H multilayers are produced by charge-transfer doping of electrons into the a-Si:H sublayers,[6] the interfacial defects are positively charged and, consequently, only electrons can be trapped at the interfaces and contribute to $\Delta\alpha$.

As a model for the carrier transport, we used the multiple-trapping model[13] to fit the experimental results. The current for homogeneous excitation for $\beta=0.5$ is given by

$$I(t)=(ev_0\pi^{1/2}g_0/2d_S)\int_0^d dz\int_0^z dz'\,dz''\,t'^{-3/2}\exp(-\pi^2z''^2/4t')\Theta(t'), \tag{2}$$

where $z''=z'/L$, $L=\mu_0E/v_0$, $t'=v_0t-z'$, μ_0 is the mobility at the mobility edge, E the average electric field, v_0 the attempt-to-escape frequency, d_S the thickness of the a-Si:H layer, g_0 the density of excited carriers, and Θ the unit step function. This equation differs from the result for surface-layer excitation[3] by an additional integration over the spatial variable z. The inset of Fig. 1 shows the data and fit by Eq. (2) for $d_S=126$ Å at 240 K ($\beta=0.5$). We also fitted the samples with $d_S=39$, 78, and 204 Å. The agreement of the data with the fits is very good. We used two adjustable parameters for each data set in these fits, the attempt-to-escape frequency v_0, and the product μ_0E of the mobility at the mobility edge with the electric field. We found that neither of these parameters depended on d_S. The attempt-to-escape frequency had a value of 32 THz in good agreement with the maximum hopping rate observed in a-Si:H.[14] The parameter μ_0E was about 100 Å/psec or 10^6 cm/sec.[15]

An important result of the theory of dispersive transport using the multiple-trapping model is the scaling law for the transit time[16]

$$t_T=v_0^{-1}\left[\frac{\pi\beta\Gamma(\beta)^2}{\sin(\pi\beta)}\right]^{1/2\beta}\left(\frac{v_0d_S}{\mu_0E}\right)^{1/\beta}, \tag{3}$$

where $\beta=T/T_0<1$, and T_0 refers to the width of the exponential band tail. Figure 4 shows the transit time versus d_S/μ_0E for $\beta=0.4$ with use of the values obtained from the fits for the parameter μ_0E. Also shown is a least-squares fit of the data points by Eq. (3) with a slope corresponding to a value for β of 0.41. This is in excellent agreement with the determination of β from the power-law branches. The scaling law [Eq. (3)] has also been confirmed for $\beta=0.5$.

In the multiple-trapping model, the dispersion parameter β is equal to T/T_0. As mentioned above, β varies linearly with temperature above 150 K (as shown in Fig. 3), but $\beta(T=0)$ is not equal to zero as expected from the multiple-trapping model. This deviation from the multiple-trapping model has also been observed for electrons in undoped[17] and arsenic-doped a-Si:H,[18] and holes in boron-doped a-Si:H[17] and a-As_2Se_3,[19] in conventional TOF and photoconductivity experiments. Recently, dispersive transport at low temperature has been attributed to hopping in exponential band tails,[20] which can account for the deviation from a T/T_0 dependence at

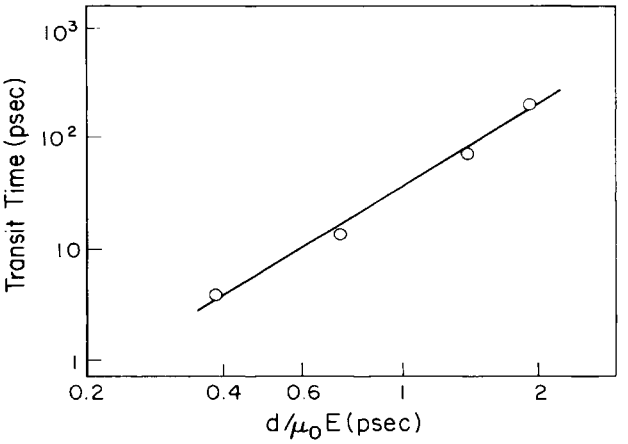

FIG. 4. Transit time vs d/μ_0E for $\beta=0.4$. The solid line is a least-squares fit with a slope corresponding to a dispersion parameter of 0.41.

lower temperatures. At high temperatures ($T_0/2 < T < T_0$) multiple trapping dominates, while for $T < T_0/2$, hopping down prevails at short times with a temperature-independent dispersion parameter. This is exactly what we observe in the a-Si:H–a-SiN_x:H multilayers. Above $T_0/2 \simeq 150$ K, the dispersion parameter exhibits the linear temperature dependence, while below 100 K, β is constant.

In summary, we have presented a new technique for the measurement of time of flight on the picosecond time scale. Using this technique we found that the transport in a-Si:H in this time range is dispersive. The temperature dependence of the dispersion parameter between 70 and 300 K indicates that at high temperatures carrier transport is by multiple trapping while at low temperatures hopping down dominates.

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