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Long-time photocurrent decay in *a*-Si:H films: A bulk property

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The photon-energy dependence of the long-time photocurrent decay in undoped *a*-Si:H has been studied for a coplanar electrode configuration. The agreement between a bulk decay model and the experimental results provides conclusive evidence that the long-time photocurrent decay is controlled by the bulk of the sample.

Transient photocurrent measurements have been widely used to study the states in the band gap as well as the properties of carrier transport and recombination in amorphous semiconductors.[1-3] A sandwich configuration is used in time-of-flight (TOF) experiments and a coplanar configuration in photoconductivity decay (PD) measurements. Usually a power-law decay behavior is observed in both TOF and PD experiments. However, one often finds that in PD experiments the photocurrent decay extends to long times, greater than 10^{-3} s for pulsed excitation, depending on the sample.[4-6] A power-law decay of the photoconductivity is also observed in hydrogenated amorphous silicon (*a*-Si:H) films near room temperature after steady-state excitation.[7,8] In the case of steady-state excitation, the photocurrent persists to times greater than 100 s. The underlying assumption of the PD measurements, which utilize coplanar electrode configurations, is that the time decay of the photocurrent is governed by bulk transport and recombination. The validity of this assumption, however, is inevitably questionable due to the nature of the electrode configuration. For instance, it has been shown that the decay of the photoconductivity in *a*-Si:H can be dominated by contact effects.[9] Jackson, Street, and Thompson[10] also suggest that the long-time photocurrent decay (LTPD) in coplanar structure *a*-Si:H samples is altered from the true bulk decay because of surface band bending. It is the purpose of this paper to show that, at least for steady-state excitation, the long-time photocurrent decay in coplanar structure *a*-Si:H samples is indeed controlled by the bulk of the sample.

In our study of the LTPD in *a*-Si:H we have found that the decay is a strong function of the steady-state excitation intensity when the light intensity is low. As the light intensity is increased, however, the decay gradually approaches a limiting power-law behavior $I_{\text{ph}} \sim t^{-\beta}$ and then becomes intensity independent as if "saturation" has been achieved for the decay process. Typical results on the intensity dependence of the LTPD are shown in Fig. 1. For convenience in the discussion that follows, we present the intensity dependence of the LTPD by showing its dependence on the steady-state photocurrent. The power-law decay exponent β is a function of temperature over the entire temperature range studied, from 300 down to 30 K. Detailed results on the LTPD will be published separately. However, from the intensity dependence of the LTPD an experiment can be devised which should provide a conclusive answer to the question whether the LTPD in *a*-Si:H is a bulk property.

One starts by finding out the steady-state photocurrent that is just high enough for the limiting power-law decay behavior to be observed for uniformly absorbed photons. Then the steady-state photocurrent is kept the same and the LTPD is measured for illuminations with different photon energies. Obviously, if the LTPD is controlled by the surface, one will observe the same power-law decay behavior irrespective of the photon energy, as the surface of the sample is always saturated. The same argument applies if the LTPD is controlled by the film-substrate interface, only with the illumination from the substrate side. On the other hand, if the LTPD is controlled by the bulk of the sample, then one will normally observe different decay curves for different photon energies. This is because the absorption depth depends strongly on photon energy. For photon energies for which the absorption depths are smaller than the thickness of the sample, only a fraction of the sample is saturated. In addition,

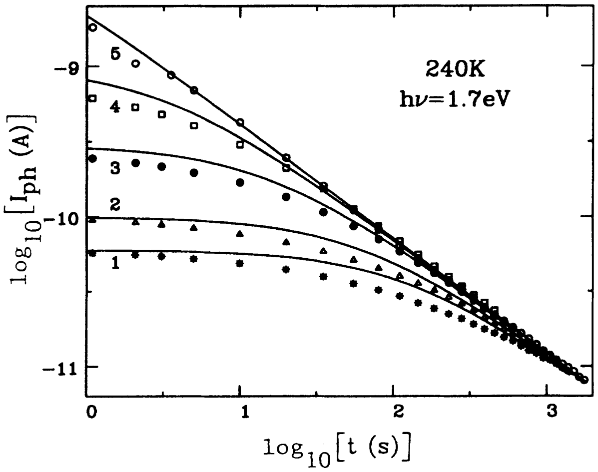

FIG. 1. Excitation-intensity dependence of the LTPD in *a*-Si:H, presented in terms of the steady-state photocurrent. The steady-state photocurrent for each decay curve is as follows: 1, 6×10^{-11} A; 2, 1×10^{-10} A; 3, 3×10^{-10} A; 4, 1×10^{-9} A; 5, 1×10^{-8} A. Symbols, experimental data. Lines, model calculations.

the "saturation depth," which we define as the depth of the region exhibiting power-law decay behavior, decreases rapidly with increasing photon energy. The above argument is illustrated in Fig. 2 which shows schematically the distribution of carrier density through the thickness of the sample under steady-state illumination for two photon energies: $h\nu_1$ is uniformly absorbed and $h\nu_2$ is highly absorbed. n_s and n_d are, respectively, the lowest steady-state carrier density necessary for saturating the sample and the carrier density in the dark. L_s is the saturation depth and D, the sample thickness. Within the saturated region the decay has, by definition, a power-law behavior, but in the rest of the sample the decay cannot be represented by a single power law. The decay of the total photocurrent is an integration of the elementary decays over the thickness of the sample and will, as a consequence, deviate from power-law behavior. Further, the magnitude of the photocurrent will decrease with increasing photon energy once saturation occurs in part of the sample. We will show that this bulk decay model agrees, not only qualitatively but also quantitatively, with the experimental results.

The samples studied in this work were undoped rf glow-discharge *a*-Si:H films deposited on Corning 7059 glass, silica, or sapphire substrates. The substrate temperature was 300 °C and film thicknesses were ~ 2 μm. Evaporated coplanar Al electrodes were deposited on the top of the films, with the electrode spacing being 0.2, 0.38, or 0.56 mm. Ohmic contacts were used. The surfaces of some of the samples were freshly etched with 10% HF solution before they were put into the measurement chamber. All measurements were carried out with the sample in a vacuum less than 4×10^{-5} Torr maintained by a diffusion pump with a liquid-N_2 cold trap. No dependence was found on the electrode spacing or on the surface condition (etched or unetched) in the present study. A monochromator was used to produce the monochromatic light. The photocurrent is defined as the difference between the total current and the thermal-equilibrium dark current.

Before discussing the main results we would like to point out the following. The possibility of contact effects in the samples was checked by measuring decays for different electric fields. The same decay behavior was observed with the magnitude of the photocurrent linearly dependent on the electric field over the entire time range in the experiment. The results presented in this paper are for one sample, but are typical of all the samples studied. The same decay measurements have been made with illumination from the film side and from the substrate side, respectively. Very similar results were obtained. The results presented here are in fact for illumination from the substrate side.

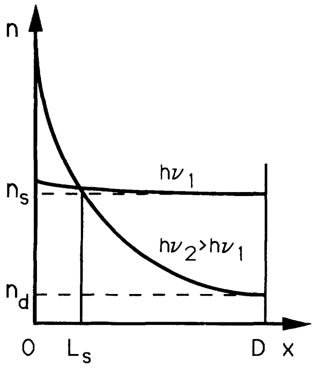

FIG. 2. Schematic diagram showing how the "saturation depth" varies with photon energy.

Figure 1 shows the time decay curves at 240 K for a number of steady-state photocurrents. The photon energy is 1.7 eV. One can see clearly the saturation phenomenon. The photon-energy dependence of the decay is shown in Fig. 3 for a constant steady-state photocurrent of 1×10^{-8} A. As predicted by the bulk decay model, the decay depends strongly on photon energy. Keeping the steady-state photocurrent the same for all photon energies enables us to avoid dealing with the light intensities directly. This simplifies both the experimental procedure and the quantitative analysis considerably. It should be pointed out that, in order to obtain the true decay for each photon energy, the decay measurements should be performed in the order starting with the most highly absorbed light. Otherwise, the measured decay may contain contributions from the residual decay generated by the previous more penetrating light, depending on the time allowed between the decay measurements. It should also be mentioned that, for the sample studied here, the decays for photon energies lower than 2 eV are almost identical under the present experimental conditions. Only the decay curve for 1.8 eV is included in the figure. In principle, in order for the results on photon-energy dependence to be most revealing one should use as low a steady-state photocurrent as is necessary to generate a power-law decay for uniformly absorbed photons. In actual practice, however, it is necessary to use a slightly higher steady-state photocurrent to ensure that the sample is definitely saturated. We used 1×10^{-8} A in the measurement of the photon-energy dependence of the de-

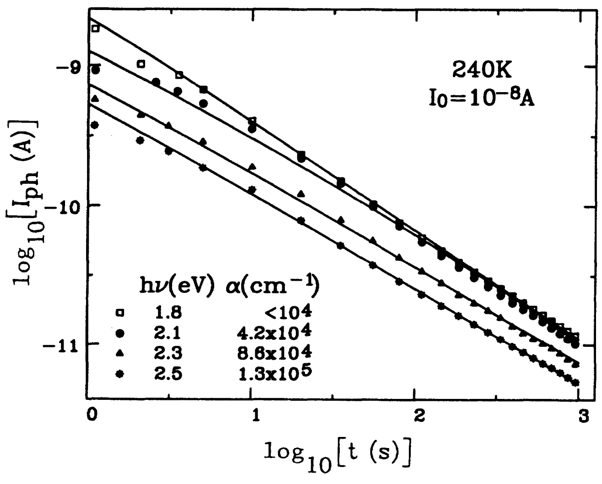

FIG. 3. Photon-energy dependence of the LTPD in *a*-Si:H. Symbols, experimental data. Lines, model calculations.

cay, compared with $\sim 5\times 10^{-9}$ A which is the lowest steady-state photocurrent necessary for generating a power-law decay for the lowest photon energy. This is why we observed the same power-law decay behavior for photon energies up to 2 eV which, almost certainly, cannot be considered as being uniformly absorbed in the sample.

In the following we will try to fit the experimental results with the bulk decay model. For the sake of argument, we define a current density $J(x,t)$ so that

$$I_{\mathrm{ph}}(t)=\int_0^D J(x,t)dx \ , \tag{1}$$

where $I_{\mathrm{ph}}(t)$ is the total photocurrent at time t after the illumination is switched off. $I_{\mathrm{ph}}(0)$ is therefore the steady-state photocurrent. The question now is how to relate $J(x,t)$ to $J(x,0)$ and, thus, to $I_{\mathrm{ph}}(0)$.

In general, $J(x,t)$ is a function of x. However, for uniformly absorbed light $J(x,t)$ is constant throughout the sample thickness. As a result, $J(x,t)=J(t)=I_{\mathrm{ph}}(t)/D$. This means that to obtain $J(t)$ for a given $J(0)$ what one needs is just the decay for the steady-state photocurrent $I_{\mathrm{ph}}(0)=J(0)D$. In other words, if the decay for any $I_{\mathrm{ph}}(0)$ is known, then the $J(t)$ for that $J(0)$ is known. In principle, the relation between $J(t)$ and $J(0)$ has to be found numerically from the decay curves in Fig. 1 because the processes responsible for the LTPD are not known in detail. However, in order to make a quantitative analysis possible, an empirical analytic relationship must be found. The fact that the saturated decay can be described by a power-law behavior tempts us to fit all the decays with the equation

$$dI_{\mathrm{ph}}(t)/dt=-A\,[I_{\mathrm{ph}}(t)]^{1+1/\beta} \ , \tag{2a}$$

with the initial condition $I_{\mathrm{ph}}(0)=I_0$, or equivalently

$$I_{\mathrm{ph}}(t)=I_0(1+Bt)^{-\beta} \ , \tag{2b}$$

where A is the decay constant and $B=AI_0^{1/\beta}/\beta$. It can be seen from Fig. 1 that although the fitting is not very good in detail, Eq. (2) does represent the decays reasonably well. Fitting of the limiting power-law decay was done by using least-square fitting (curve 5). The first two experimental data points, which have large uncertainty, were ignored in the fitting. Fitting of the limiting power-law decay determines all the constants in Eq. (2). The other fits in Fig. 1 were obtained by substituting the corresponding steady-state photocurrent into Eq. (2). The result of the fitting indicates that the steady-state photocurrent plays a more complicated role in the LTPD than implied by Eq. (2). Detailed analysis shows that the decay constant A must be considered to be time dependent if Eq. (2a) is to be used to describe the LTPD. That the decay constant is time dependent is one of the most important features of the LTPD. For the purpose of this paper, however, Eq. (2b) is a good approximation. This is justified by the fact that in the bulk decay model the total photocurrent at a given time is contributed mainly by the region which exhibits the limiting power-law decay behavior at that time.

Rewriting Eq. (2b) in terms of the current density we obtain

$$J(t)=J_0(1+B't)^{-\beta} \ , \tag{3a}$$

where $J_0=I_0/D$ and $B'=B=AJ_0^{1/\beta}D^{1/\beta}/\beta$. It is important to note that Eq. (3a) is valid for any J_0. Therefore it can be generalized for any distribution of current density as

$$J(x,t)=J(x,0)[1+B'(x)t]^{-\beta} \ , \tag{3b}$$

where $B'(x)=A\,[J(x,0)]^{1/\beta}D^{1/\beta}/\beta$. To calculate $J(x,0)$, we need to know its dependence on the carrier generation rate, G. If, for uniformly absorbed light, $I_{\mathrm{ph}}\sim G^{\gamma}$, then we can assume that $J(x,0)\sim G^{\gamma}$, unless the photons are highly absorbed and x is small compared with D so that $J(x,0)$ is considerably greater than I_s/D, where I_s is the lowest steady-state photocurrent necessary for generating the limiting power-law decay for uniformly absorbed light. However, for $J(x,0)\geq I_s/D$ the decay has a single power-law behavior and is independent of $J(x,0)$ and, therefore, a relation between $J(x,0)$ and G is no longer needed. Simple mathematical deduction allows us to obtain

$$J(x,0)=I_0\gamma\propto[1-\exp(-\gamma\alpha D)]^{-1}\exp(-\gamma\alpha x) \ , \tag{4}$$

where α is the absorption coefficient. A very-well-defined value for γ is obtained for the photocurrent values covered in this study and $\gamma=0.84$.

We are now in the position to fit the decay curves in Fig. 3 by utilizing Eqs. (3b) and (4). The integration of Eq. (3b) was performed numerically. The fitting is very sensitive to α when $\alpha>2\times 10^4\ \mathrm{cm}^{-1}$. The fitting for each photon energy is done by carefully adjusting α until the best fit is obtained. Again, the first two data points were ignored in the fittings. It can be seen from Fig. 3 that the bulk decay model predicts the overall behavior exhibited by the experimental results. The absorption coefficients determined from the fitting are listed in the figure. They agree extremely well with those appearing in the literature.[11]

In summary, we have measured the excitation-intensity and the photon-energy dependence of the long-time photocurrent decay in coplanar structure *a*-Si:H samples. The excellent agreement between the bulk decay model and the experimental results leads us to the conclusion that the LTPD is a bulk property.

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