TIME RESOLVED SPECTROSCOPY OF THE PHOTOINDUCED ABSORPTION IN GLOW-DISCHARGE a-Si:H

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Abstract.- The time and temperature dependence of the photoinduced absorption spectra was measured in glow-discharge a-Si:H in the energy range 0.7 to 1.4 eV, temperature interval 10 to 306K and time domain 2 μs to 2 ms. The results are consistent with a model based on thermalization of excess carriers in trap states.

Introduction.- Illumination of a-Si:H in the fundamental absorption band produces a midgap absorption band (photoinduced absorption PA).[1-3] It was shown[4] that the CW spectrum of the PA band is consistent with photoionization of holes trapped in states close to the valence band into the valence band. The shape of the spectrum can be explained if the initial states of trapped holes are confined to a narrow energy interval (approximated by a delta function) and the density of final states in the valence band has a square root dependence. The origin of the delta-like distribution of initial states was ascribed to the peaking of the product (trap density) x (hole distribution function) at the quasi-Fermi level E_{pf} above the valence band edge E_v assuming that the trap density decreases sharply into the gap. We have recently studied the time-dependence of the total strength of the PA band in a-Si:H.[5,6] In this paper we report data on the temperature and time-dependence of the PA spectra and show that they can be explained by the model discussed below.

Model of PA spectrum.- The time-dependence is introduced into the CW model by following the ideas of Tiedje and Rose[7] and Orenstein and Kastner.[8] They considered the kinetics of the relaxation of trapped carriers preceeding the thermal equilibrium state. They introduced a demarcation energy E_d which plays the same role as the quasi-Fermi level in equilibrium, namely separates the occupied and unoccupied traps. They also assume that the trap distribution decreases sharply into the gap so that the hole concentration strongly peaks in an energy interval close to E_d (Fig. 1). This "hole packet" moves with temperature and time according to the relation
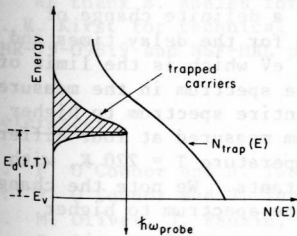

Fig. 1 - Model for photo-induced absorption.

$$E_d(t,T) = kT \ln(\nu t) \tag{1}$$

where ν is a vibrational frequency. The time and temperature dependence of the photoinduced absorption spectra should follow the equation

$$\Delta\alpha(\omega) \sim (\hbar\omega - E_d)^{1/2}/\hbar\omega \tag{2}$$

E_d from Eq. (1) figures here as the threshold energy; we assumed constant matrix element and $\hbar\omega \gg kT$. Our spectral studies provide for the first time a direct check of this model compared to other transport measurements such as transient photocurrent and time of flight. In our case recombination takes place in parallel with thermalization. However, if the trapped carrier distribution is strongly peaked at $E_d(t,T)$ we expect that recombination does not change the motion of the peak (although it changes the total concentration of holes) since the release times of trapped carriers on which the motion of E_d is based (Eq. (1)) does not depend on recombination.

Experiment.- The PA band was excited by a dye laser pumped with a nitrogen laser with the following parameters: photon energy 2.1 eV, pulse duration 10 ns, repetition rate 20 Hz and pulse energy $60 \mu J$. The sample was amorphous silicon prepared by glow discharge deposition with 16 at. % H_2 and thickness $d \simeq 5 \mu m$. The PA was probed with light from a tungsten lamp in the energy range of 0.7 to 1.4 eV. The detection system consisted of a Ge photodiode (Judson J-16 LD), a broad band amplifier (PAR 115) followed by a boxcar integrator (PARC 162) and an X-Y recorder. The time resolution of the experiment was $0.5 \mu s$. The average energy resolution of the monochromator placed between the sample and the detector was $\simeq 0.03$ eV.
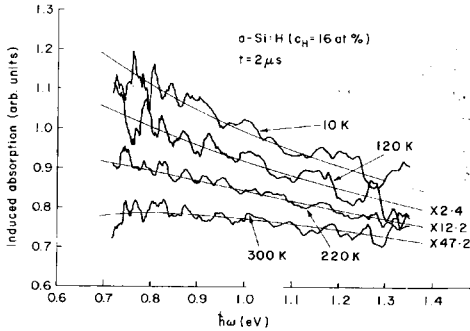

Fig. 2 - Temperature dependence of PA spectra at delay time t = 2 μs. Smooth curves are calculated from Eqs. (1) and (2) with $\nu = 10^{13} sec^{-1}$.
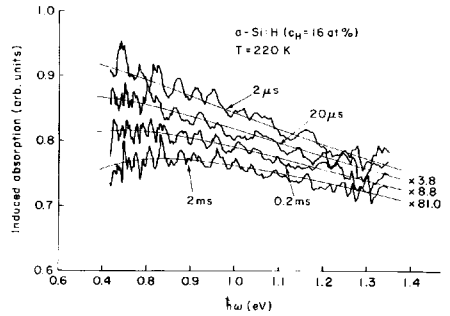
Fig. 3 - Time dependence of PA spectra at 220 K.

Results and Discussions.- Figure 2 shows the PA spectrum measured at four different temperatures T = 10, 120, 220 and 306 K, for a fixed time t = 2 μs. The spectra at higher temperatures have been multiplied by constants indicated in the figure for better comparison of their shapes. The structures which appear in the curves are the remnants of interference fringes which could not be completely averaged out. In spite of being relatively featureless the PA specrum shows a definite change of shape with increasing temperature. The entire PA spectrum for the delay times and temperatures used in the experiment extends much below 0.7 eV which is the limit of detection for our measurements. The change in shape of the spectrum in the measured energy range can be interpreted as due to a shift of the entire spectrum to higher energies with increasing T. Figure 3 shows the PA spectrum measured at four different times t = 2 μs, 20 μs, 0.2 ms and 2 ms for a fixed temperature T = 220 K, with the spectra at longer times multiplied by appropriate constants. We note the change in shape of the spectrum showing roughly equal shifts of the spectrum to higher energies when t changes by a decade.
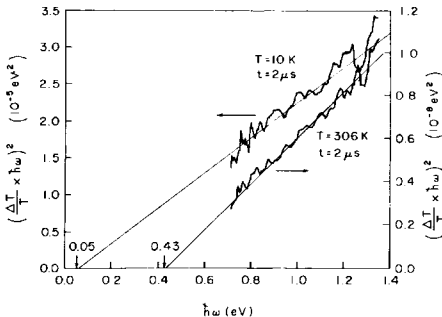

The shifts of the spectrum can be seen more quantitatively by extracting the absorption threshold E_d from the measured spectrum using Eq. (2). Figure 4 shows the normalized spectra of PA plotted as $[\hbar\omega\Delta T/T]^2$ vs. $\hbar\omega$ for the spectrum at T = 10 K, t = 2μ s and at T = 306 K, t = 2μ s ($\Delta T/T = d\Delta\alpha$). The fit to Eq. (2) is good over the energy range of the measurement; this permits the determination of E_d. Similar plots for the remaining spectra shown in Fig. 2 and 3 were also in good agreement with Eq. (2) and yielded threshold energies in the range from 0.05 eV to 0.44 eV (Fig. 5).
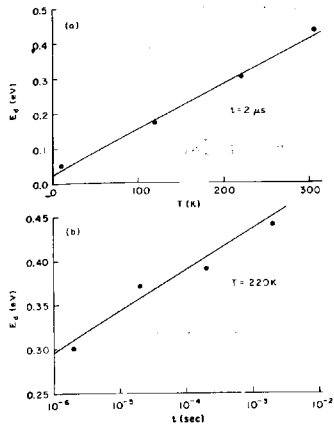
Fig. 4 - Normalized PA spectra (arrows show E_d).

Fig. 5 – Threshold energy E_d as a function of temperature (curve a) and time (curve b).

Figure 5(a) shows a plot of E_d, obtained from the measured PA spectra, as a function of T for a fixed t ($t = 2 \mu s$). We see that E_d depends linearly on T in agreement with Eq. (1). Using the slope of the E_d vs. T curve we obtain $\nu \simeq 10^{13} sec^{-1}$ which is a reasonable value. In Fig. 5(b) we plotted E_d as a function of log t for a fixed T (T = 220 K). The straight line behavior is also in agreement with Eq. (1). The shift of E_d at 10 K was checked at $t = 2\mu s$ and $t = 2ms$; within our experimental limits of determining E_d (± 0.02 eV), E_d does not show any shift, in agreement with Eq. (1).

Finally, inserting Eq. (1) in Eq. (2) and using $\nu \simeq 10^{13} sec^{-1}$ we can calculate the shape of the PA spectrum as a function of T and t. The smooth curves in Figs. 2 and 3 are the calculated spectra for the temperatures and delay times of our experiment; it is seen that the fit to the measured spectra is good. The effect of the shift of the spectrum with time on the decay of the integrated PA band was calculated and found that its influence on the power law reported in Refs. 5 and 6 was small (the error in the dispersion parameter α is smaller than 5% at 300 K and less at lower temperatures).

Conclusions.- We showed that the time and temperature dependence of the PA spectra in GD a-Si:H can be described with Eq. (1) and (2) with a single adjustable parameter ν whose value is $10^{13} sec^{-1}$. This material is a particularly simple case, apparently because the density of trap states decreases very sharply into the gap. In contrast, in sputtered a-Si:H[6] such a simple analysis does not appear to be applicable probably because the trap density is not so sharp. A difficulty occurs also with a-As$_2$Se$_3$ in which the PA spectrum cannot be described by Eq. (2) and the determination of E_d from the spectra has not been feasible.[8]

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