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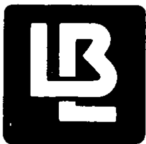

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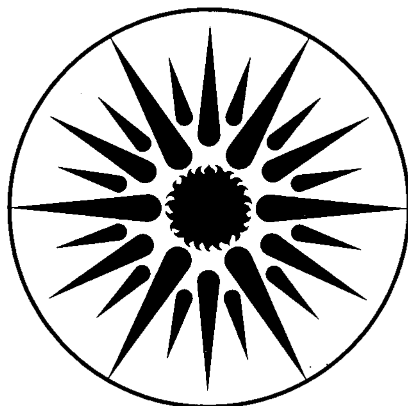

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INFLUENCE OF DANGLING BOND DEFECTS ON RECOMBINATION IN a-Si:H

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Time resolved photoinduced absorption (PIA) and steady state photoconductivity experiments (PC) are used to study recombination processes in electron irradiated hydrogenated amorphous silicon (a-Si:H). Defect densities measured by ESR range from $5 \times 10^{15}/cm^3$ to $10^{18}/cm^3$. It is found that while increasing the dangling bond density reduces the PC, the PIA decay rate increases. The results are discussed in the context of a recombination model where the defect density determines whether the carriers recombine through a fast bimolecular or a slower monomolecular process.

1. INTRODUCTION

Recombination processes in a-Si:H have been studied extensively using luminescence[1] and photoconductivity[2] experiments. These investigations have shown that the dangling bond is the major recombination center which determines the photoconductivity and luminescence in this material. Seemingly contradictory results were recently obtained by Wake and Amer[3] using time-resolved photo-induced absorption (PIA) measurements which probes the decay of photoexcited carriers. The results in the nanosecond time regime indicate that the PIA decay is slowest in samples with high defect densities rather than in high quality, low defect density material. It has been concluded that in the nanosecond time regime, the properties of the band tail determine the relaxation of excess carriers rather than the dangling bond defect density. In the following, we report on transient PIA and steady state PC measurements on electron-irradiated a-Si:H. The results indicate that two competing processes determine the mechanism by which the photoexcited carriers relax.

2. RESULTS AND DISCUSSION

Sample preparation, conductivity and ESR-measurements were done at the University of Marburg.[4] The ESR spin density N_s of the as-deposited samples is $5 \times 10^{15}/cm^3$. Electron irradiation was carried out as described in Ref. (4) and leads to an increase in N_s to more than $10^{18}/cm^3$ without changing the line shape and g-value of the ESR-signal. N_s is reduced by annealing the samples at increasingly higher temperatures, T_A, (annealing time 30 min) and is restored to its original value after a 220 °C anneal. The dark conductivity was measured between T_A and 0 °C and shows an activated behavior. Although a slight increase in both the activation energy, E_a, and the prefactor, σ_o, with decreasing N_s can be observed, the overall dependence of these quantities on the defect density is rather weak ($\sigma_o = 10^3 \ldots 10^4\ \Omega^{-1}\ cm^{-1}$, $E_a = 0.8 \ldots 0.9$ eV). This implies that the dark fermi level E_F remains almost constant over the entire defect density range which is an important premise for the present study, since it is known that the recombination properties of a-Si:H depend strongly on the position of E_F.[2] In contrast to σ_d, the photoconductivity changes by nearly three orders of magnitude, which is roughly the change in defect density. Since the

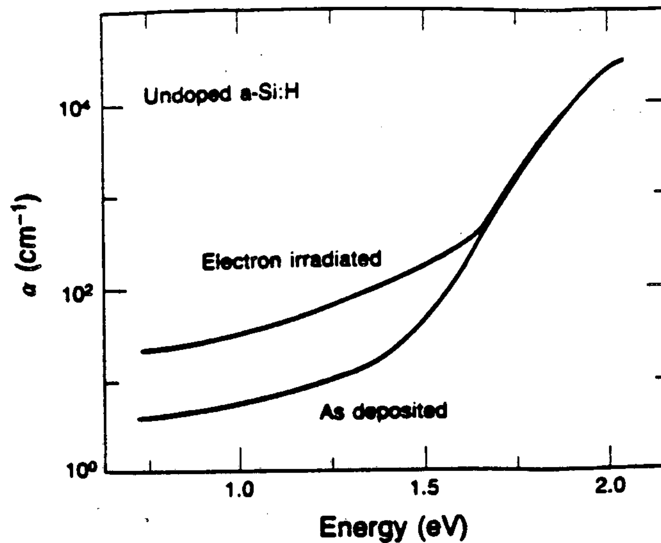

FIGURE 1
Optical absorption spectra of irradiated and unirradiated a-Si:H.

mobility, μ, enters both the prefactor σ_o of the dark conductivity and the photoconductivity we conclude that the much larger change in the case of σ_{ph} is mainly caused by a reduction of the carrier lifetime due to the increased defect density rather than by changes in μ. The intensity dependence of σ_{ph} was measured in the range 0.2 to 80 mW/cm^2 using neutral density filters. The exponent η in the relation $\sigma_{ph} \sim I^{\eta}$ indicates monomolecular recombination kinetics for all defect densities.

The optical absorption was measured using the photothermal deflection scheme described in Ref. (5). Fig. 1 compares absorption spectra from an unirradiated and irradiated sample. The spectrum consists of the typically reported features, namely an exponential edge ascribed to transitions involving tail states (Urbach edge) and a low energy band due to transitions from deep centers, predominantly dangling bonds.[5] Electron irradiation influences only the latter one, whereas the Urbach edge (slope 65 meV) remains unaffected. The defect absorption band increases upon irradiation without changing its shape. Therefore, we can conclude, that all changes introduced by electron irradiation are confined to deep gap states, while the band edges and tails remain unaffected.

Time-resolved photoinduced absorption was measured in the time regime of 10 nsec - 1 μsec using a 10 Hz Nd-YAG-pumped dye laser with 4 nsec pulses of 2.2 eV photons, and a cw broad-band (0.9 - 1.45 eV) IR probe beam. Details of the setup have been described elsewhere,[3] and the temperature range at which data were taken was 120 K - 200 K. At low excitation densities ($n_{exc} < 5 \times 10^{17}/cm^3$) the data can be approximated by a power law decay [$\Delta T/T \propto t^{-\alpha}$] with $\alpha = 0.15$. In both samples the decay becomes faster at higher intensities and shows deviations from the power law behavior. In contrast to what one might expect the fast decay component is most prominent in the unirradiated, low defect density sample. This can be clearly seen in Fig. 2, where the slope α of the transients at 20 ns and 2 μs is plotted as a function of n_{exc}. At long times and low excitation densities α approaches a constant value of 0.15 which is roughly the same for both defect densities. Multiple trapping theory[6] (MT) predicts a value given by the temperature and the exponential slope, E_o, of the trap distribution, $\alpha = kT/E_o$. Assuming that the slope of the Urbach edge (Fig. 1)

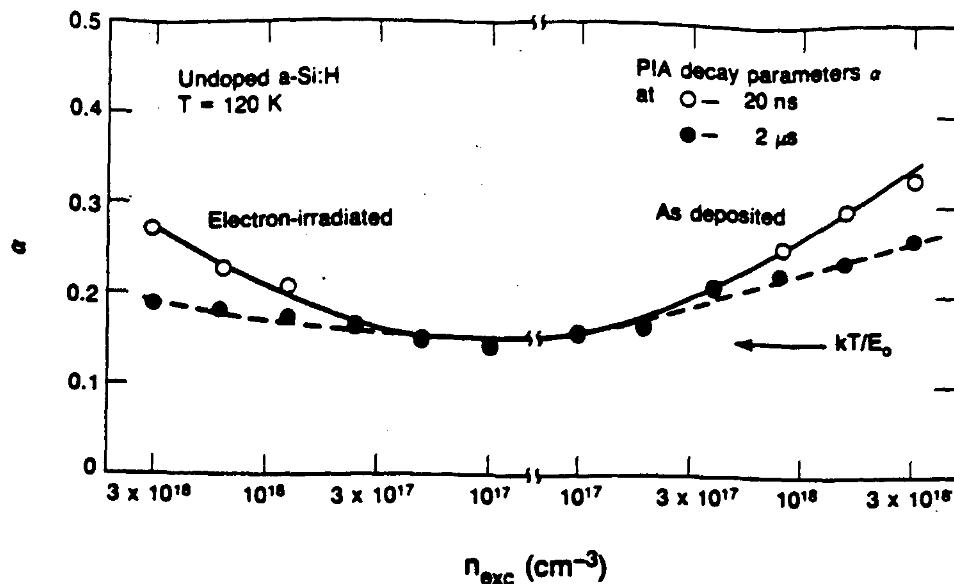

FIGURE 2
Slope of the Transients of the photoinduced absorption at 20 ns and 2 μs as a function of the excitation density.

reflects the trap density of states, we obtain $\alpha = 0.15$ in agreement with the low excitation density data. Also, the temperature dependence given by the equation above was verified in the range 120-200K, which supports the multiple trapping model as being adequate to describe the transients for low n_{exc}.

With increasing intensity α becomes larger, reaching 0.33 in the as-deposited and 0.27 in the irradiated sample. The intensity dependent decay constant requires a second bimolecular or higher order process to be present. The transition between the monomolecular and bimolecular regions occurs at roughly $5 \times 10^{17}/cm^3$. Similar transitions at the same excitation density have been observed in luminescence decay experiments. It has been suggested that the fast process is due to Auger recombination[1,7] or nongeminate, bimolecular recombination[1] of bandtail carriers. The present results clearly show that the fast component decreases as the defect density is enhanced, whereas the monomolecular decay at low excitation densities remains almost unaffected.

From the photoconductivity results we conclude that the steady state recombination in the present samples is dominated by a monomolecular process involving the dangling bond defects. It is therefore most likely that the same process governs the monomolecular PIA-decay at low excitation densities. Tauc[8] concluded from the spectral dependence of the photoinduced absorption band that it is mainly due to holes trapped in the exponential valence bandtail. The process responsible for the PIA decay is therefore a transition between a trapped hole and the defect. The decay at high excitation densities is ascribed to recombination between bandtail carriers which may occur through a direct bimolecular process or, involving a third carrier, through Auger recombination. The unexpected inverse dependence of this process on the defect density can be understood within a recombination model recently derived from spin dependent photoconductivity measurements.[4] According to that model, recombination at dangling bonds occurs by tunneling of trapped electrons towards neutral dangling bonds thereby forming a doubly occupied,

negatively charged defect state. Recombination is completed by subsequent transitions between trapped holes and the negatively charged dangling bond. This latter process is responsible for the decay of the PIA at low intensities and long times, and can be accounted for by the multiple trapping model. At higher carrier densities, a second recombination mechanism becomes operative which depends on both hole and electron densities. Due to the low defect density, more electrons will remain in the conduction band tail of the unirradiated sample than in the case of the irradiated one, where a larger fraction of electrons has been captured by the defects. Therefore, the bimolecular recombination channel will be stronger in the unirradiated sample as is clearly observed. In the irradiated sample the fast capture of electrons by dangling bonds forces more carriers into the slower defect-related recombination channel and makes them unavailable for the bimolecular process.

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