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DUAL LIGHT BEAM MODULATION OF PHOTOCARRIER LIFETIME IN INTRINSIC a-Si:H

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ABSTRACT - We present dual beam photoconductivity measurements on intrinsic glow discharge a-Si:H. Several recombination effects have been isolated by varying the wavelength and chopping frequency of a secondary photon beam while pumping the sample with a primary beam of band gap photons and detecting the changes in the photoconductivity. For T < 200K infrared quenching of photoconductivity with a low energy threshold of \(-0.6\) eV is found. Above 250K a different infrared quenching process occurs with a low energy threshold of \(-0.9\) eV. At intermediate temperatures (200K < T < 250K) a new positive modulation signal is observed. Quenching results are interpreted in terms of optical excitation of electrons from the valence band into recombination centers with small electron capture coefficients. The positive modulation signal is attributed to an optical transition between localized gap states.

INTRODUCTION - The dual beam photoconductivity modulation technique involves the use of a steady pump light beam of interband photons and a chopped modulation beam which scans the energies within the gap. The pump beam changes the occupation of gap states through which recombination takes place. The relationship between gap state occupation and recombination of photogenerated carriers is investigated by modulating the occupation of specific sets of gap states using the modulation beam and measuring the resultant changes in the photoconductance. In the following we describe and interpret some features observed in the dual beam spectra of intrinsic a-Si:H. Further data and a more complete analysis are presented elsewhere.

RESULTS AND DISCUSSION - Amorphous Si:H films were prepared by the rf plasma decomposition of SiH\(_4\) mixed with Ar buffer gas in a capacitively coupled system under conditions described earlier.

The temperature dependence of the dc photoconductance \(G_\text{p}\) of a 2\(\mu\)m thick undoped a-Si:H sample excited with a pump beam flux of \(\approx 2 \times 10^{16}\) photons cm\(^{-2}\) sec\(^{-1}\) is shown in Figure 1. Three temperature regimes are denoted in the figure corresponding to characteristic temperature and intensity dependence of \(G_\text{p}\). In region I \(G_\text{p}\) is nearly independent of temperature and depends on photon flux intensity \(\Phi\) as \(G_\text{p} \propto \Phi^\gamma\) with \(0.9 < \gamma < 1.0\). In region II \(G_\text{p}\) decreases with increasing \(T\) (thermal quenching) and has a supralinear intensity dependence with \(1.0 < \gamma < 1.1\). In region III \(G_\text{p}\) increases with increasing \(T\) with an activation energy of \(-0.2\) eV and \(\gamma \approx 1.8\).

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...which is related to the number of effective recombination centers. The thermal quenching and the supralinear intensity dependence of $G_P$ in region II as well as the constancy of $G_P$ in region I indicate the presence of at least two distinct classes of recombination centers.\textsuperscript{4,5}

In order to explain these observations as well as infrared quenching discussed later, we use the following simple model. We assume two kinds of recombination centers: (i) $r$-centers which have a large electron capture coefficient and lie primarily above the dark Fermi level $E_F$ (above midgap) and (ii) $s$-centers with a small electron capture coefficient which lie predominantly below $E_F$. The hole capture coefficients are assumed for simplicity to be the same for $r$- and $s$-centers. The majority photocarriers are assumed to be electrons.

In the low $T$ region I under the high pump conditions of Figure 1, most $r$-centers are occupied and most $s$-centers are empty due to the difference in electron capture coefficients. $G_P$ is relatively large because the $r$-centers, which are the most effective recombination centers, contain a relatively small number of holes compared to $s$-centers. As $T$ is raised (region II) an increasing number of $s$-centers come into thermal equilibrium with the valence band raising the number of free holes available for capture into the more effective $r$-centers and decreasing $G_P$ (thermal quenching). Thermal quenching of photoconductivity has recently been observed in intrinsic a-$Si:H$ samples prepared under a variety of conditions\textsuperscript{6} and has been correlated with infrared quenching of photoconductivity.\textsuperscript{1,2} Infrared quenching is the result of optical excitations of electrons from the valence band to empty $s$-centers and the subsequent capture of some of the free holes into $r$-centers.

The dual beam modulation spectra $P_m(h \nu)$ in Figure 2 clearly demonstrate infrared quenching of photoconductivity. Solid lines in the figure indicate negative modulation photoconductance. Thus at $T=100K$ the total photoconductance of the sample is decreased upon the addition of the beam for $0.6 \text{ eV} < h \nu < 1.5 \text{ eV}$. Note that $P_m$ is the rms change in $G_P$ normalized linearly to the modulation beam photon flux and that the change in $G_P$ is a small perturbation upon the total photoconductance.

In an earlier paper\textsuperscript{1} it was shown that the low energy optical threshold for infrared quenching of $G_P$ is $0.58 \pm 0.05 \text{ eV}$ and is temperature independent and the same for many samples. This indicates that the $s$-centers are distributed in energy above $0.6 \text{ eV}$ above the valence band. From the saturation of the magnitude of the infrared quenching effect with increasing modulation beam intensity we deduce an electron capture coefficient for $s$-centers of $4 \times 10^{-13} \text{ cm}^3 \text{ sec}^{-1}$ at $133K$. This value suggests charged centers because typical capture coefficients for neutral centers in crystals are about $10^{-8} \text{ cm}^3 \text{ sec}^{-1}$. The low $T$ quenching effect disappears when $T$ is increased above $210K$ as the trap quasi-Fermi level for holes passes through the $s$-center distribution.

In temperature region III a different infrared quenching signal is observed with a threshold energy of $\sim 0.9 \text{ eV}$, as shown by the curve labeled $300K$ in Figure 3. A negative modulation effect will occur whenever there is an optical exchange of charge from a center with a small capture coefficient to one with a large one as long as both levels are enclosed by their respective trap quasi-Fermi levels. Supralinearity will only be
Figure 1
Photoconductance \( G_p \) of an intrinsic a-Si:H sample as a function of temperature. Band gap photon flux is \( 2 \times 10^{16} \text{ cm}^{-2} \text{ sec}^{-1} \).

Figure 2
Normalized modulation photoconductance \( P_m (h\nu m) \) for several \( T \) measured in phase with the modulation beam chopped at 5 Hz. Solid lines denote negative and dashed lines denote positive modulation signal.

Figure 3
Same as in Fig. 2 except \( T=200 \text{ K} \) and \( 300 \text{ K} \).

Figure 4
Normalized modulation photoconductance \( P_m (h\nu m) \) at 220 K measured 90° out of phase with the modulation beam chopped at 4 Hz.
observed when the small coefficient center lies below and the large coefficient center above \( E_F \). We designate these small electron capture coefficient centers as X-centers. Since suprilinear behavior is not observed, either (i) the number of X-centers is small or (ii) the X-centers lie above the dark Fermi level. The high threshold energy of 0.9 eV supports the latter suggestion. If the ratio of electron capture coefficients of r- and X-centers were constant in T one would expect to observe the high threshold quenching process not only for \( T > 250 \) but also at lower T. The absence of this effect at lower T suggests that the X-center capture coefficient decreases with respect to the r-center coefficient as T increases. One possible mechanism is the capture of electrons into X-centers through an intermediate trap or excited state. We note here that modulation spectra above 200K are sample dependent, however, only two classes of behavior are observed in intrinsic samples.\(^8\)

We turn now to an interesting and as yet unexplained modulation feature which is most apparent at 220K for a pump flux of \( 2 \times 10^{16} \) cm\(^{-2}\) sec\(^{-1}\). The Pm(hwm) spectrum measured in phase with the 5 Hz chopped modulation beam is labeled 220K in Figure 3. The shape of the spectrum suggests a positive bump at 1.0 eV superposed upon the normal positive background due to excitation from the band tail. The spectral shape of the bump can be isolated by noting that the spectrum in this region is composed of a slow (\( \tau = 32 \) msec) and a fast response time (\( \tau < 1.4 \) msec) component; removal of the fast background is accomplished by tuning to the signal which is 90° out of phase with the modulation excitation at a low chopping speed of 4 Hz. The resulting spectrum in Figure 4 clearly shows the peaked nature of this new feature. Peaked behavior is indicative of a direct localized-to-localized state transition between two bands of states which affects the recombination kinetics. Further work is necessary to clarify the origin of this effect.

Dual beam photoconductivity modulation is a valuable tool for elucidating the nature of gap states in amorphous semiconductors. We have established the existence of at least three distinct types of gap state in a-Si:H which are distributed in well-defined energy ranges and which control the lifetime of free carriers. We thank E. A. Schiff for helpful discussions. This work was supported in part by the NSF-MRL program at The University of Chicago and by NSF Grant DMR-8009225.

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