EXCITATION ENERGY DEPENDENCE OF PHOTOINDUCED ABSORPTION IN INTRINSIC a-Si:H

N. SCHULTZ, Z.V. VARDENY, P.C. TAYLOR
Department of Physics, University of Utah, Salt Lake City, Utah 84112

ABSTRACT

We have studied subgap absorption of intrinsic a-Si:H induced by below- and above-gap photoexcitation. We find very similar photoinduced subgap absorption spectra when excited with 2.4 eV or 1.2 eV light. Both spectra exhibit a power-law dependence on laser intensity $\Delta T \sim 1^{\alpha}$, where α is 0.5 and 0.7 for 2.4 and 1.2 eV excitation energy, respectively. This behavior indicates a change in the recombination mechanism as a function of excitation energy. The PA spectrum excited at 1.2 eV shows a strong dependence on bias illumination. Bias illumination bleaches the absorption according to a power-law as $\Delta T = c(E_{bias}) \cdot I^{\beta}$, where β is approximately 0.85 and independent of probe energy and bias energy. The parameter $c(E_{bias})$ increases superlinearly with bias illumination energy for $E_{bias} > 1.7$ eV.

INTRODUCTION

Photomodulation (PM) is a powerful technique to investigate electronic states within the gap of amorphous semiconductors. In this technique the sample is photoexcited with a (pump) laser and induced changes are monitored in the absorption of (probe) light from an external light source. When the subgap absorption is enhanced or decreased under photoexcitation the process is called either photoinduced absorption (PA) or photoinduced bleaching (PB), respectively. In previous studies excitation energies, E_{ex}, well above the bandgap energy were employed, and photoinduced absorption was found over the whole below-gap energy range. Though featureless, the spectrum was attributed to at least two contributions,[1,2] and correlations to the photoluminescence spectrum were found.[3] In the studies presented here we examine subgap absorption induced by below-gap photoexcitation. In photoluminescence, below-gap excitation has yielded much information on the distribution of states within the gap, and evidence has been found[4] for two-step excitation processes of charge carriers through the deep defects.

EXPERIMENTAL

The intrinsic sample used in theses studies was prepared by glow discharge deposition on a sapphire substrate. The sample, whose thickness was ~7 μm, was mounted inside a cryostat and illuminated from the film side. For above- and below-gap excitation the unfocused laser lines of an argon laser (514.5 nm, 30 mW/cm^2) and a Nd:YAG laser (1064 nm, 2.5 W/cm^2), respectively, were used. The modulation frequency of the exciting light was in all cases around 130 Hz. The photoinduced changes in absorption that were in phase with the laser excitation were probed with a focused tungsten halogen light or an ir-glowbar. The probe light was dispersed by a single grating monochromator and detected with Si, Ge and InSb solid-state detectors covering the spectral range from 0.3 to 1.7 eV. The measured spectra contained interference fringes due to internal reflections that were spectrally averaged. For the measurements under bias illumination, the light of a second tungsten halogen lamp was focused onto the sample. In all cases the low energy part of the tungsten spectrum was blocked with a heat absorbing filter (shortpass at approximately 800 nm), and bias

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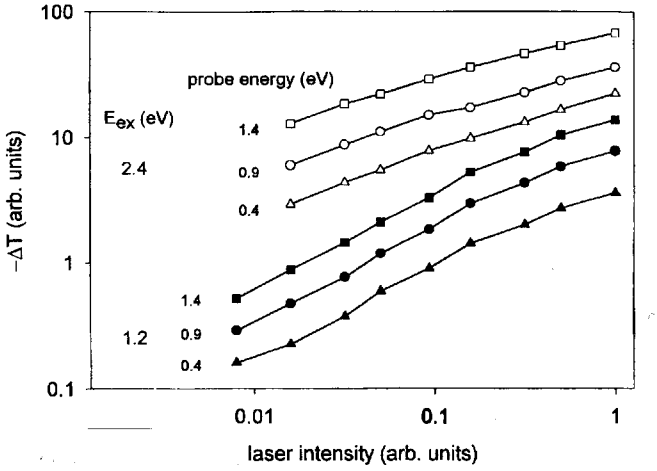

Fig. 1. Photoinduced absorption of a-Si:H as a function of laser excitation intensity. The open and filled symbols refer to excitation energies of 2.4 eV and 1.2 eV, respectively.

light of different energies and intensities was achieved by employing band-pass and neutral density filters. Without neutral density and band-pass filters the power of the bias illumination was about 80 mW/cm^2. All measurements were performed at 80 K.

RESULTS

Figure 1 shows the photoinduced changes in absorption ΔT as a function of the incident laser intensity. ΔT was measured at three representative below-gap probe energies of 0.4 eV, 0.9 eV, and 1.4 eV as the sample was excited with 2.4 eV (upper curves) and 1.2 eV (lower curves) light. At both excitation energies and all probe energies we find a power-law dependence of ΔT on laser intensity: $\Delta T \sim I_{ex}^{\gamma}$, where I_{ex} is the laser intensity and γ is approximately 0. 5 and 0.7 for 2.4 eV and 1.2 eV excitation energies, respectively.

Figure 2 shows the PM spectra collected at excitation energies of 2.4 eV and 1.2 eV. Breaks in the data occur near the excitation energy of 1.2 eV due to filters used in the experimental setup to block the Nd:YAG laser light from entering the detectors. Since both spectra were taken one after another, the gap appears also in the spectrum excited at 2.4 eV. Without bias illumination (Fig. 2a) the PA spectra excited at above- and below-gap energies show strong similarities. Both spectra exhibit a broad maximum around 0.5-0.6 eV. The weak dependence of the induced absorption on photoexcitation energy is also seen in photoluminescence (PL), even when the PL excitation energy lies below the detected luminescence energy.[4] Figure 2a also shows the spectra under white-light bias illumination of approximately 80 mW/cm^2. Whereas the induced subgap absorption is only slightly reduced under bias when excited at 2.4 eV, we find photoinduced bleaching when the material is excited at 1.2 eV. If a linear superposition of the bleached transitions under bias illumination and photoinduced absorption without bias illumination is assumed, the bleaching component can be extracted by subtraction of the PA spectrum without illumination from the

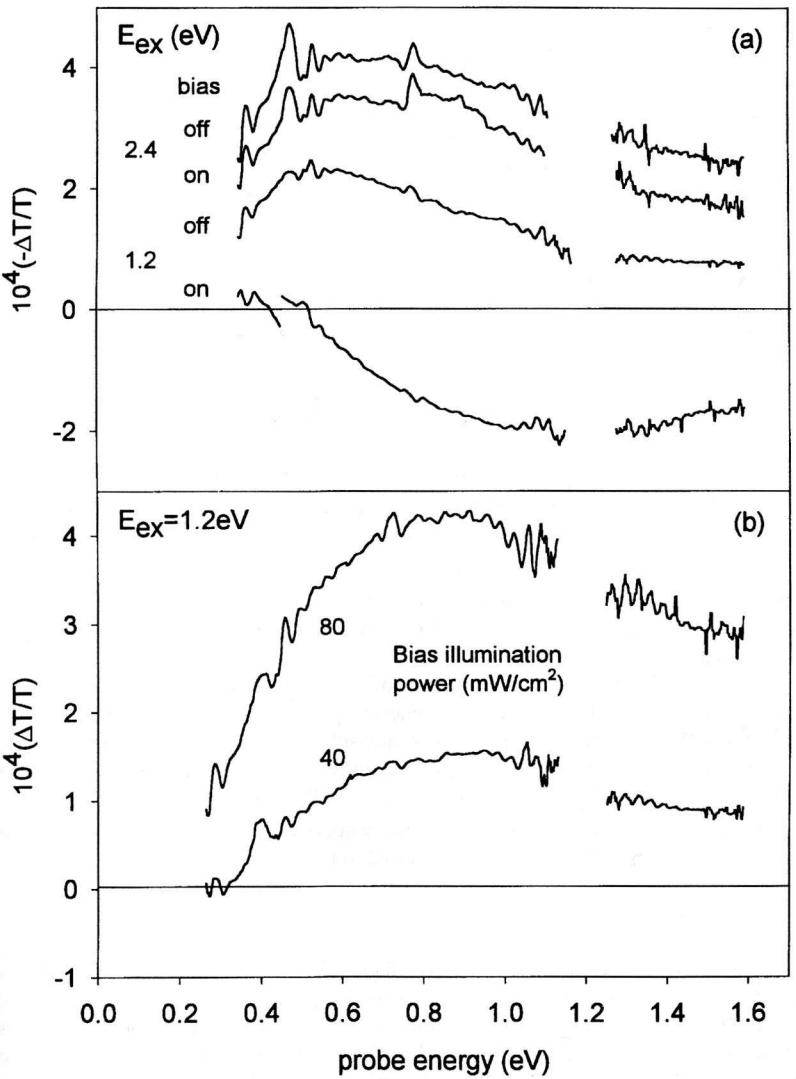

Fig. 2. (a) Photomodulation of a-Si:H excited at 2.4 eV and 1.2 eV with and without bias illumination of 80 mW/cm^2. Under 1.2 eV photoexcitation energy the induced absorption spectrum changes with bias illumination to an induced bleaching spectrum. (b) Bleaching component of the photomodulation spectra excited at 1.2 eV. The sharp spectral features that occur in all spectra are experimental artifacts.

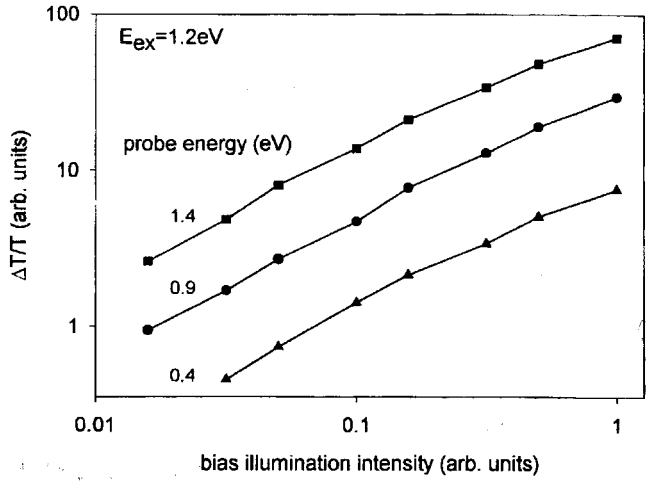

Fig. 3. Magnitude of the observed photobleaching at E_{ex} = 1.2 eV as a function of bias illumination intensity.

spectrum with illumination. These subtractions are shown in Fig. 2b for two illumination intensities. Note the change in sign of the ordinate in Fig. 2b. We find a broad featureless bleaching signal with a broad maximum at around 0.9 eV.

We have examined in more detail the bleaching of the subgap absorption under bias illumination when photoexcited with light of below-gap energy. Figure 3 shows the magnitude of the photoinduced bleaching as a function of bias light intensity at three representative probe energies of 0.4 eV, 0.9 eV, and 1.4 eV. At all probe energies we find a power-law dependence of the bleaching process, ΔT_{bias}, on the bias-light intensity, I_{bias}. Specifically, $\Delta T_{bias} = c(E_{bias}) \cdot I_{bias}^{\beta}$, where $c(E_{bias})$ increases rapidly with the energy of the bias light above approximately $E_{bias} \simeq 1.7$ eV. The exponent β is independent of the bias-light energy and is found to be 0.85 ± 0.10.

DISCUSSION OF RESULTS

The power-law dependence of the induced subgap absorption on excitation intensity has an exponent that changes from 0.5 to 0.7 when exciting the sample with 2.4 eV and 1.2 eV, respectively. A square-root dependence indicates bimolecular recombination kinetics under 2.4 eV excitation. The larger exponent that is observed in the case of below-gap excitation may be interpreted as a mixture of bimolecular recombination, as occurs for above-gap light, and monomolecular recombination when both carriers are generated close to each other in localized states. For excitation at 1.2 eV it is conceivable that charge carriers are photoexcited either from the extended states of the valence and conduction bands into the deep defects or from tail states to tail states deeper in the gap. In the former transition, one mobile carrier is generated in extended states and one localized carrier at a deep defect. Two-step excitations through the deep defects (a process whose existence is known from below-gap excitation of photoluminescence[4]) lead to an increase of mobile carriers in both bands. In the latter tail-to-tail transition both excited carriers are localized, presumably near each

other. Therefore, from the mobile carriers excited at 1.2 eV one expects recombination kinetics that are bimolecular because they are similar to those observed for excitation at 2.4 eV, and when both carriers are created in localized states monomolecular (geminate) recombination is probable. The sum of the two processes in the limited excitation range could yield an approximate power law whose exponent is between 0.5 and 1.

Bias illumination produces a bleaching effect on the photoinduced absorption, especially under low-energy excitation. The power-law behavior of the bleaching with the intensity of the bias illumination is independent of the probe and bias light energies. In particular, bleaching increases rapidly when the photon energy of the bias light exceeds 1.75 eV. Since this energy coincides approximately with the band-gap energy, it is most likely that the bleaching process involves a redistribution of carriers in the band-tail states. From temperature dependent measurements it has been concluded that the photoinduced absorption spectrum consists of at least two contributions.[3] Bias illumination, however, appears to affect the entire induced absorption spectrum, therefore a fundamental process, that affects all carriers, such as that mentioned above, is necessary to explain the bleaching.

SUMMARY

In a-Si:H photomodulation excited with both above- and below-gap energies exhibits a photoinduced subgap absorption spectrum $\Delta\alpha$ whose spectral dependence is independent of excitation energy and has a power-law dependence on the excitation intensity, I_{ex}. Specifically, $\Delta\alpha \sim I_{ex}^{\gamma}$, where γ is approximately 0.5 and 0.7 for 2.4 eV and 1.2 eV excitation energies, respectively. This change in recombination kinetics is ascribed to a change from bimolecular recombination when the material is excited with above-gap-energy light, to a mixture of bimolecular and monomolecular recombination when excited with below-gap-energy light. When the sample is excited with below-gap energy light, bias illumination of the sample, in addition to the standard pump and probe light sources, leads to photoinduced bleaching. The bleaching spectrum peaks at around 0.9 eV and is attributed to a redistribution of band-tail carriers.

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