# SPIN DEPENDENT PHOTOINDUCED ABSORPTION IN a-Si:H

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

## ABSTRACT

We have studied photoexcitation dynamics in undoped a-Si:H from 80 K to 300 K by the techniques of photoluminescence (PL), photoinduced absorption (PA) and their respective versions of optically detected magnetic resonance, namely PLDMR and PADMR. Both PL and PA spectra are composed of low and high energy bands, respectively. Using their respective temperature dependences we correlate the low energy PA band (~ 0.4 eV) to the high energy PL band (~ 1.3 eV) and the high energy PA band (~ 1 eV) to the low energy PL band (~ 0.8 eV). We also found that the PADMR spectrum is composed of three main contributions: a narrow resonance at g ~ 2 (FWHM = 15 G), a broad resonance at g ~ 2 (160 G) and a narrow asymmetric resonance at g ~ 4 (20 G). By measuring their photon energy spectrum we found that the g ~ 4 and the broad g ~ 2 resonances are correlated with each other and therefore are due to trapped electron-hole pairs in the triplet spin configuration.

## INTRODUCTION

Photoluminescence (PL) and photoinduced absorption are frequently employed techniques for examining the below-gap density of electronic states. PL gives information on the distribution of levels from which radiative recombination takes place. PA, on the other hand, measures photoinduced changes in transmission of an external probe light and, therefore, compared to PL has the advantage that it probes all levels of excited carriers. Employing these techniques, both band-tail states and deep-defect states (dangling bonds) can be probed.

In the existing models the high energy PL band at 1.3 eV[1] and the low energy PA onset at 0.4 eV[2] have been ascribed to the same photoexcited carriers, namely electrons and holes trapped in the band tails. Similarly, the low energy PL at 0.8 eV[3] and the high energy PA[4] have been ascribed to carriers trapped in the midgap states. To test this correlation between PL and PA we examine their respective temperature dependences. We have also measured photoinduced- absorption-detected magnetic resonance (PADMR). This technique links electron spin resonance (ESR) and PA by monitoring changes in the transmission of an external probe light due to resonant changes in the recombination rates of the photogenerated carriers. In a simple model, resonance occurs when $h\nu = g\beta H_0$, where $\nu$ is the applied microwave frequency, h is Planck's constant, g the Lande`-factor, $\beta$ the Bohr magneton and $H_0$ the external dc-magnetic field. Two different kinds of PADMR measurements were done: First, the magnetic field was scanned at a fixed probe energy (H-PADMR) to identify various spin resonances and, second, the probe energy was scanned at the fixed resonant magnetic field (E-PADMR).

## EXPERIMENTAL

The sample used in our measurements was an intrinsic, 7μm thick a-Si:H film deposited on a sapphire substrate. The ESR spin density was $3 \times 10^{16}$ $cm^{-3}$ before optical excitation. The PL measurements were taken from Ref. 5. For the PA measurements the sample was mounted in a nitrogen-cooled cryostat and excited from the film side with approximately 400 mW of an $Ar^+$ laser



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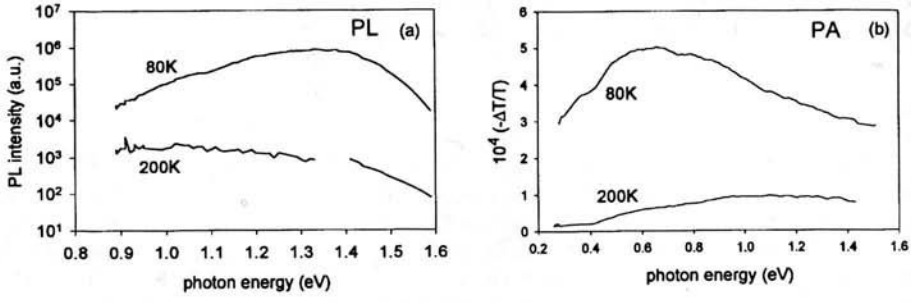


Fig. 1. (a) Photoluminescence (PL) and (b) photoinduced absorption (PA) spectra of a-Si:H at 80 K and 200 K.

(514.5 nm line). The probe light was obtained from halogen tungsten source or an ir-glow bar, and was dispersed by a single-grating monochromator and detected with Si, Ge and InSb solid state detectors. For the magnetic resonance measurements the sample was mounted in a 3 GHz microwave cavity installed in a cryostat cooled with liquid helium. The details of the experimental setup are described elsewhere.[6] The PL data where corrected for system response which is inherently not necessary for PA and PADMR, since the data are normalized with respect to the transmission.

## RESULTS

Figure 1 shows the PA and PL spectra at 80 K and 200 K. The PL-spectrum at 80 K shows the commonly observed dominant high-energy band at approximately 1.3 eV, attributed to radiative recombination of charge carriers trapped in band-tail states. The PL spectrum also contains a much weaker PL band at 0.8 eV, attributed to radiative recombination involving midgap states. At 200 K the band-tail PL at 1.3 eV is strongly reduced and the PL is dominated by the low-energy band. Note that Fig. 1a uses a semi-logarithmic scale. The PA spectrum (Fig. 1b, linear scale) at 80 K shows a band around 0.6 eV and at 200 K a broad PA around 1.0 eV, confirming that at least two different photoexcitations contribute to PA. The low-energy PA is attributed to optical transitions from photogenerated carriers trapped in the band tail to the adjacent band. This contribution to the PA shows a stronger temperature dependence than the high-energy PA ascribed to transitions from occupied midgap states (dangling bonds).

Both PA and PL show an exponential temperature dependence, $\exp(-T/T_0)$, above 80 K in the measured spectral range. Figure 2a shows a typical example of the PL and PA decays at 0.75 eV and 1.4 eV, respectively. The dependence of the temperature constant $T_0$ for PL and PA is shown in Fig. 2b, where a smaller $T_0$ corresponds to a stronger temperature dependence. The temperature dependences of PL and PA exhibit approximately opposite behaviors over the examined spectral range. The strongest temperature dependences are found for the peak energy of PL near 1.3 eV and the lowest PA energies near 0.3 eV where the temperature constants are $T_0 = 25$ K and $T_0 = 40$ K, respectively. On the other hand, the low energy PL and high energy PA both exhibit the weakest temperature dependences with $T_0 = 80$ K. The temperature dependence of PA seems to level off at both ends of the measured energy range. For PL it is known that the temperature constant levels off

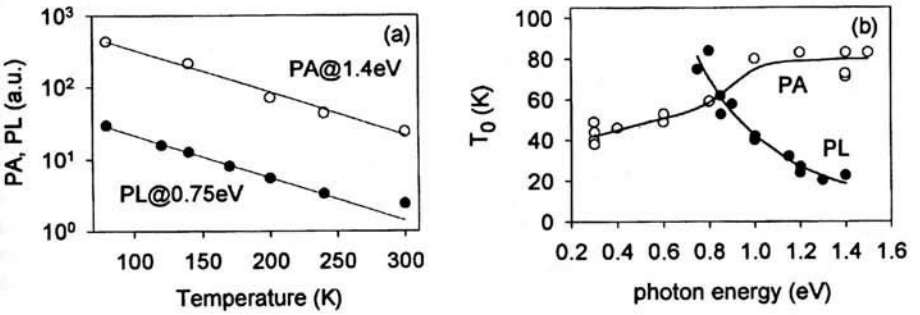


Fig. 2. (a) Typical example of the decrease of PA and PL with temperature. (b) The dependence of the temperature constant $T_0$ on photon energy of PL and PA in the spectral range 0.3 to 1.5 eV at 80 K. The PL data were taken from Ref. 5.

at the lowest energies and that it is only very weakly temperature dependent at the highest PL energies of around 1.7 eV.[5]

To study the nature of the photoexcited carriers that are responsible for the PA signal we examined their spin dependence over the entire probe-energy range. Figure 3 shows the H-PADMR spectrum at a probe energy of 0.9 eV at 10 K. As previously observed[6] we find three resonances: an asymmetric (half-field) resonance at $g \sim 4$ (signal C) and two symmetric resonances at $g \sim 2$ (narrow and broad signals labeled A and B, respectively). Figure 3 also shows the decomposition of the narrow signal into two Lorentzians (A and B) of widths 15 G (FWHM) and 160 G, respectively.

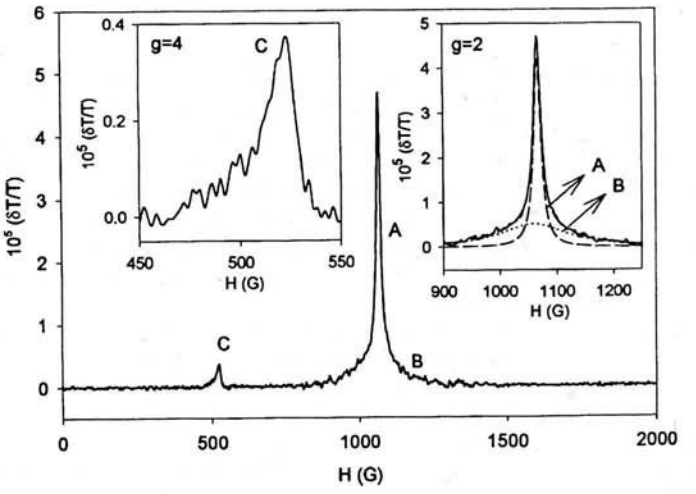


Fig. 3. H-PADMR of a-Si:H at 0.9 eV. The right-hand inset shows the decomposition of the $g \sim 2$ resonances into two Lorentzians. The left-hand inset shows the $g \sim 4$ signal in more detail. Data were taken at 10 K.

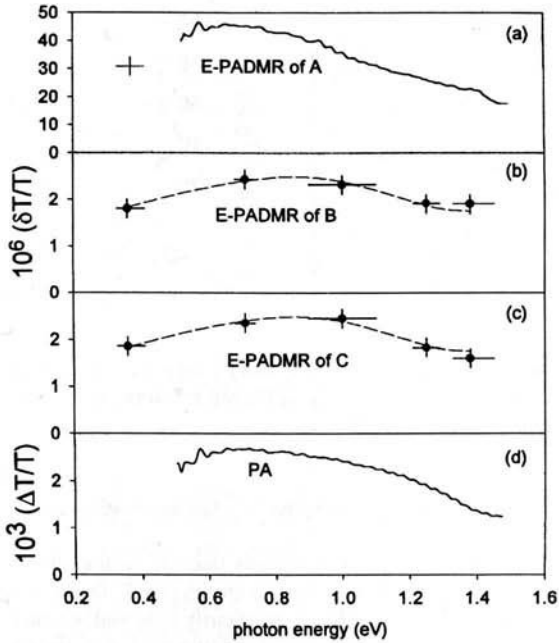


Fig. 4. E-PADMR spectra of the three observed resonances A, B and C compared to PA at 10 K.

We also measured the E-PADMR at the three resonances, as shown in Fig. 4. The energy spectrum taken at the peak of resonance A is shown in Fig. 4a. Neglecting the small contribution of the underlying resonance B, (~ 10%) this spectrum essentially represents the spectral response of resonance A. To get the energy dependence of resonance B (Fig. 4b), we decomposed the H-PADMR resonances (Fig. 3) at various probe energies into two Lorentzians and scaled the height ratios to the energy dependent spectrum of resonance A. Lorentzian lineshapes were chosen because they provided a much better fit to the data than Gaussian lineshapes. The energy dependence of the $g = 4$ resonance is also shown in Fig. 4c. The data in Fig. 4c were taken only at five selected energies because of signal-to-noise restrictions.

The $g \sim 2$ resonance A probably originates from spin $S = 1/2$ excitations because there is no correspondence with the energy dependence of the $g \sim 4$ resonance. Resonance A peaks at around 0.7 eV, falls off rapidly to lower energies and drops gradually by about a factor of 2 to higher energies. This spectrum resembles the PA spectrum at 10 K (Fig. 4d), with a maximum at 0.7 eV. The spectra of the broad resonance B and the resonance at $g \sim 4$ strongly resemble each other, and in contrast to resonance A, show a much weaker energy dependence. From these two spectra a broad maximum appears around 0.8 eV. We therefore conclude that these two spectra are correlated. In fact we propose that they are the half-field and full-field resonances of photo-excited electrons and holes that are closely coupled so as to produce an effective spin 1 system.

## DISCUSSION

As shown in Fig. 2, there exists a similar temperature dependence for the high energy PA band and the low energy PL band. In addition, the temperature dependences for the low energy PA and the high energy PL are very similar. This behavior confirms the models[2,4] that ascribe these signals to the same underlying photoexcitations, although the physical interpretation of the exponential behavior remains unclear. If the rate for nonradiative recombination is determined by tunneling of carriers between wells due to potential fluctuations of the bands, it can be shown that an exponential temperature decay follows.[7] PA calculations based on the multiple trapping model also lead to an exponential temperature dependence.[4] This model may be similarly applied to the PL temperature dependence.

The E-PADMR measurements show similar spectra for the asymmetric $g \sim 4$ signal and the broad $g \sim 2$ resonance that link them to a common origin, namely spin $S = 1$ photoexcitations. From deconvolutions of the $g \sim 2$ resonance into two Lorentzians at various probe energies we find equal resonance strengths at a probe energy of 1.3 eV. At lower energies the E-PADMR spectrum of the narrow signal rises faster than the broad E-PADMR that shows only very little energy dependence. We conclude that at 1.3 eV there exists an equal contribution of $S = 1/2$ and $S = 1$ photoexcitations to the PA. At lower energies the fractional contribution of the $S = 1/2$ excitations increases with respect to the $S = 1$ excitations.

## ACKNOWLEDGMENTS

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