

Midgap injection-induced absorption in amorphous silicon

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Carrier injection in a forward-biased amorphous silicon Schottky diode is used to modulate the sample absorption coefficient. The measured modulation is interpreted as predominantly due to the elimination of transitions of holes from midgap states to the valence band. The method and conclusions are compared with other techniques sensitive to midgap optical transitions: photoinduced absorption, photocapacitance spectroscopy, and steady-state photoconductivity.

I. INTRODUCTION

The investigation of the optical transitions involving midgap states in amorphous hydrogenated silicon (a -Si:H) has made use of many experimental techniques. Most of these techniques measure the optical absorption transitions of electrons whose initial or final states lie within the band gap. The direct measurement of the optical transmission of thin films is not sufficiently sensitive for the reliable deduction of midgap optical absorption coefficients and thus is an unsuitable probe for midgap states.¹ Thus the experiments used include more indirect methods, such as photoconductivity,² photoacoustic spectroscopy,^{3,4} photothermal deflection spectroscopy,⁵ calorimetry,⁶ and guided-wave spectroscopy.⁷

The disadvantage of all these techniques is the ambiguity of assignment of the transitions involved. There are two transitions which contribute in principle to the midgap absorption coefficient, depicted as transitions 1 and 2 in Fig. 1. Transition 1 involves the transfer of an electron from a midgap state below the Fermi level to the conduction band, and transition 2 the promotion of a hole from a midgap state above the Fermi level to the valence band.⁸ The position of the Fermi level will determine which states can contribute to each of these transitions. The separation of the contributions of each of these transitions is a subject of recent work, with varying conclusions. Several authors have assumed or concluded that the dominant midgap absorption transition is transition 1,^{2,9-11} while others have deduced that transition 2 is dominant,¹² or that one cannot separate the two.^{4,13}

This disagreement in the type of transition responsible for midgap absorption, and the credibility of deducing absorption spectra from indirect measurements such as photoconductivity, is responsible in part for the difficulties associated with the attempts to deduce gap density-of-states (GDOS) distributions from absorption spectra, however derived.² Thus for a greater understanding of the GDOS, one must attempt to resolve the contributions of the various midgap optical transitions.

In addition to the absorption methods just discussed, defect photoluminescence can be used to obtain information about optical transitions involving midgap states.¹⁴ In this discussion, however, we will concentrate on the methods of absorption rather than emission spectroscopy.

Modulation spectroscopy is a class of methods¹⁵ which also may be used in principle to investigate the optical properties of midgap states in amorphous silicon. Previous work in this area has concentrated on electric field modulated reflection¹⁶⁻¹⁹ and absorption,^{20,21} photoinduced absorption,²²⁻²⁴ and photocapacitance spectroscopy.^{25,26} Of these techniques, only photoinduced absorption and photocapacitance spectroscopy have been able to provide information concerning midgap states. The electroabsorption and reflection experiments normally give signals only at spectral energies near the band gap.

In photoinduced absorption, the absorption coefficient of a steady-state population of photoexcited electrons and holes is measured with a modulation technique. Unfortunately, because the photoexcitation creates both electrons and holes, photoinduced absorption entails the presence of four different induced transitions. These four transitions, shown in Fig. 1, can in principle all contribute to the total photoinduced absorption signal. The first two transitions are identical in nature to those for simple absorption discussed above. In the photoinduced absorption

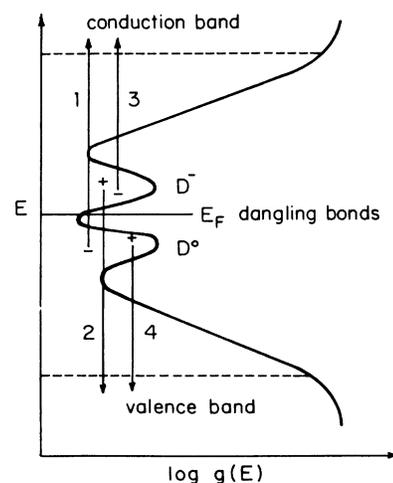


FIG. 1. Schematic of midgap optical transitions in amorphous silicon.

experiment, these two transitions would contribute a negative signal (induced transparency) because the number of carriers in the initial state is reduced and the number of carriers in the final state is increased by the photoexcitation. Transitions 3 and 4 represent the new transitions available by virtue of the presence of the photoexcited carriers, and thus contribute a positive signal (induced absorption). Transition 3 represents the transition of a photoexcited electron from a gap state above the equilibrium Fermi level to the conduction band. Transition 4 represents the promotion of a photoexcited hole from a gap state below the equilibrium Fermi level to the valence band. Assuming steady-state statistics, the quasi-Fermi levels for electrons and holes determine where the photoexcited carriers are, and thus which states can contribute to each of these four types of transitions. It may be possible to use the details of the spectral dependence of the photoinduced absorption to help sort out the contributions of these transitions.²⁷

A second, more complicated modulation technique is that of photocapacitance spectroscopy. In this method, a reverse-biased diode is brought briefly into flat-band condition, thereby injecting electrons into trapping states in the semiconductor. These electrons detrapp upon absorption of incident photons, and are measured electrically when they drift across the sample now under reverse bias again. This method has the advantage over photoinduced absorption that only one sign of carrier is injected. Because it is sensitive only to electrons, however, it does not greatly aid in the separation of the optical absorption transitions.

An alternative method of investigating the optical properties of electrically injected carriers is to measure the optical absorption directly. The experimental procedure for such a measurement is similar to a normal electroabsorption measurement. Here, however, the modulation is due not to the field itself, but to the injection of carriers. Because this measurement method enjoys the noise-rejection advantages of modulation techniques, the modulated absorption coefficient can be measured directly, without requiring the more indirect majority-carrier transport methods used in photocapacitance spectroscopy. In this way, the electrical injection can produce carriers just as the photocreation in the photoinduced absorption experiment.

Injection through an electrical contact differs, however, from photoinjection in that, by the choice of a Schottky structure, one can choose to inject just one sign of carrier. This will simplify the interpretation of the experimental results by reducing the number of transitions induced by the modulation. The direct measurement of the optical absorption allows the detection of transitions which do not produce free electrons as well as those which do, making both majority and minority carrier transitions detectable, and discriminable by the sign of the signal. In this paper, we report the use of injection of electrons in a Schottky diode structure to produce this form of electroabsorption. Because we know that the injected carriers are electrons, the transitions which may contribute to the modulation signal are transitions 2 and 3 in Fig. 1.

This measurement technique can be compared with a

similar experiment performed recently by Mescheder and Weiser on symmetric coplanar electrode α -Si:H devices.^{21,28} In their experiment, carrier injection exerts an influence on the absorption coefficient, in that the spatially inhomogeneous distribution of injected carriers gives a contribution to the internal field, producing near-band-gap electroabsorption. This method thus gives much the same information as earlier work on electric-field-modulated absorption.

II. APPARATUS AND PROCEDURES

A sensitive ratio electroabsorption apparatus was constructed to perform the measurements presented in this paper. A schematic diagram of the apparatus is given in Fig. 2. An Ealing Stabilarc 100 lamp house with an Osram XBO 75-W high-pressure short arc xenon lamp powered by an Ealing Universal Arc Lamp Supply is used as a light source. The light is focused onto the entrance slit of a Model 82-415 Jarrell Ash 0.25-m grating monochromator. Normally 500- μ m slits are used. A mechanical chopper is placed before the entrance slit and used during alignment of the optical path. The monochromated light is passed through a Corning CS2-58 red long-pass filter to remove higher-order diffracted wavelengths. The light is then split by a glass plate placed at a 45° angle with respect to the beam. The reflected portion of the light is focused onto an EG&G Model UV-215BG silicon photodiode operated at zero bias (photovoltaic mode) for maximum signal-to-noise ratio. This beam is used to monitor the source intensity and its fluctuations. The main portion of the light is focused onto the sample, and the transmitted light is collected and focused onto a matching silicon photodiode.

A major consideration of the electronic portion of the apparatus is the elimination of noise. After elimination of effects such as ground loops, vibration, pickup, etc., one of the important remaining sources of noise is the fluctuation in the received light intensity, caused primarily by the 120-Hz current ripple in the arc lamp power supply on the short time scale, and by arc wandering on the long time scale. An electronic subtraction scheme was devised

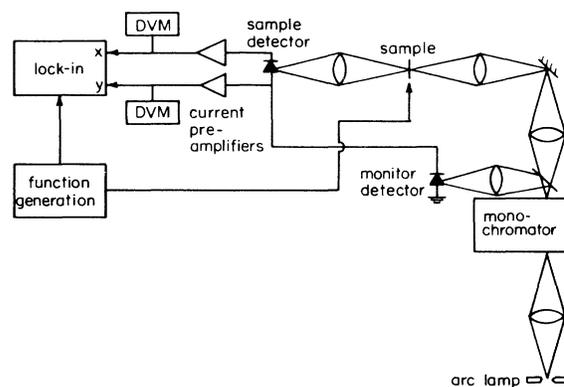


FIG. 2. Electroabsorption system apparatus.

to eliminate some of the noise due to lamp fluctuation. The dc components of the current outputs of the two photodiodes are set to be equal by manually adjusting a variable neutral density filter in the sample beam or a variable aperture in the monitor beam. These currents are subtracted at a Kirchhoff node before being sent to a current-sensitive preamplifier, an Ithaco Model 1211 preamp. This preamplifier thus sees only the difference between the two detector photocurrents, whose dc component is zero, and thus whose lamp fluctuation component should cancel to first order. The current preamplifier can then be set to a relatively high gain, because the input current is very small. The output of this amplifier is introduced to the input of an EG&G Model 5208 dual phase lock-in amplifier, which provides as its output the ΔT signal. The digital output of the lock-in amplifier was transferred over a GPIB (general purpose interface bus) to the computer. A second current-sensitive preamplifier, an EG&G Model 181 preamp, is used to monitor the dc component of the monitor photodiode current, producing the T signal.

The signals are averaged digitally over periods of up to one minute per point to provide greater signal-to-noise ratio. For most of the experiments, simultaneous measurements of both the in-phase (ΔT_x) and out-of-phase (ΔT_y) components of the modulation signal were performed. The reasons are twofold: Measuring both phase components obviates the need for setting the phase correctly before the data are taken, which in the presence of very long time constants can be difficult. Also, frequency-dependent electronic phase shifts can occur which are appreciable, necessitating the measurement of both phase components. The two phase components ΔT_x and ΔT_y are converted into polar form ΔT and ϕ . The ratio $\Delta T/T$ is computed, and it is this value which is called the electroabsorption (EA) signal. With this apparatus, EA magnitudes of the order 10^{-7} are routinely measurable, and with care it is possible to get down to 10^{-8} . This compares favorably with the magnitude of the signals seen here and elsewhere.¹⁵

For these experiments, the sample is a Schottky diode based on a device quality amorphous hydrogenated silicon intrinsic layer, with semitransparent top and bottom contacts. The sample geometry consists of a Corning 7059 glass substrate which has been coated by Corning with a SnO_2 transparent conductive layer. On this layer is deposited a thin (~ 30 Å) layer of chromium, in an attempt to isolate the tin in the SnO_2 bottom layer, and prevent it from diffusing into the amorphous silicon layer. Then a thin (~ 700 Å) n^+ -type $a\text{-Si:H}$ layer is deposited by glow discharge on the Cr back contact to act as a source of electrons. On this is deposited the thick ($5 \mu\text{m}$) intrinsic $a\text{-Si:H}$ film, and then one or more relatively thin Cr dots (~ 50 Å), forming a Schottky barrier on top. The sample devices are tested by measuring the current-voltage characteristic, and only those with rectification ratios of about 10^4 or higher at ± 1 V are used. All measurements of electroabsorption and current-voltage characteristics were performed at room temperature. The Fermi level is near midgap in the intrinsic layer, as evidenced by a conductivity activation energy E_σ of about 0.7 eV. Other

characterizational measurements normally performed at this laboratory, such as photoluminescence, optical and infrared absorption, and photoconductivity, were carried out on codeposited samples, and indicate that the intrinsic layer in these devices is of high electronic quality.

The samples are inserted into the electroabsorption apparatus, and a mask is installed to prevent stray light from passing around the devices or around the entire sample substrate. This mask is especially important for measurements in the spectral region above the semiconductor band gap, where the transmitted light intensity is very low. Data are taken as a function of modulation voltage, modulation frequency, and spectral energy.

III. RESULTS AND DISCUSSION

In Fig. 3 are presented two representative curves showing the dependence of the EA magnitude on the modulation voltage at forward bias, and one curve at reverse bias. At low modulation voltages, the detected signal is found under forward bias only, indicating that the detected signal is not due to merely the application of an electric field. The two forward bias curves differ only in that the wavelength setting of the monochromator, set to an energy in the midgap region, was changed slightly. In both of these curves, the EA magnitude rises roughly linearly for small modulation voltage, levels off, and then behaves in a more unpredictable fashion at large modulation voltages. The behavior at large modulation voltage is due to Ohmic heating of the sample, which causes a modulation of the product of the index of refraction and the sample thickness (nd). A modulation of the nd product modulates the interference fringes, which dominate the transmission coefficient in this spectral region of low absorption. The sign of the apparent EA spectrum at large modulation voltages depends on the side of the transmission interference fringe, and therefore alternates in sign.²⁹ This behavior is manifested in the unpredictability in the large voltage regime in Fig. 3. For the rest of the measurements presented here, the modulation voltage was restricted to the linear region at small voltages.

In all cases, the Ea signal disappeared completely when the incident light was blocked, indicating that neither electroluminescence³⁰ nor incandescence contributed to the

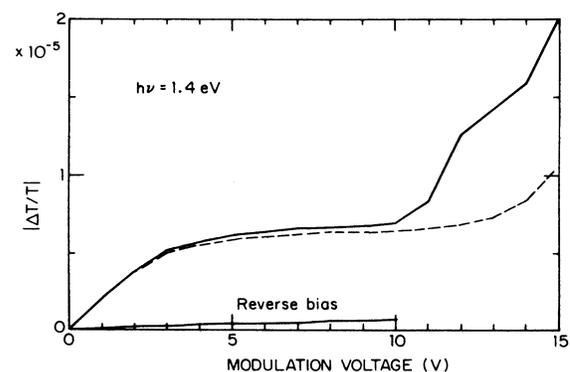


FIG. 3. Voltage dependence of the electroabsorption signal.

signal. To test for photoenhanced electroluminescence (electroluminescence enhanced by the illumination of the device³¹), we used off-axis illumination, where the transmitted illumination was not focused onto the detector, but the device was still imaged onto the detector. In this case, no modulation signal was seen, indicating that photoenhanced electroluminescence was also unimportant.

The modulation frequency dependences of the midgap and near gap EA signals are shown in Fig. 4. The nearly flat dependence of the midgap EA magnitude on the modulation frequency indicates that the signal is not thermally induced.¹⁸ The rolloff at large frequency is due to the RC time constant of the measurement circuit and sample. The near gap EA signal falls off rapidly with increasing modulation frequency. For the rest of the measurements presented here, the modulation frequency was fixed at 1 kHz.

We have assumed that the injected carriers are located near and above the equilibrium Fermi level. We now give a justification for this assumption. The dielectric relaxation time τ_{rel} is given by

$$\tau_{rel} = \epsilon\epsilon_0 / \sigma, \quad (1)$$

where the dielectric coefficient ϵ is typically about 13 for amorphous silicon, and the conductivity σ is about $2 \times 10^{-9} (\Omega \text{ cm})^{-1}$ for this series of undoped glow discharge samples. As the devices used in this study made use of n^+ -type layers, there may be some contamination of the intrinsic layer, implying the possibility of a somewhat higher conductivity than this value. Thus we estimate that the dielectric relaxation time is about 0.5 msec or lower for these devices. As this value is of the same magnitude as the duration of the voltage pulses used in the EA measurement, we conclude that the injected carriers are relatively well thermalized, and thus do lie near and above the equilibrium Fermi level. This conclusion, together with the magnitude and spectral dependence of the observed signal, also allows us to exclude the possible influence of free-carrier absorption on our results.

In Fig. 5 are shown the magnitude and relative phase of a typical EA spectrum taken in the low voltage regime.

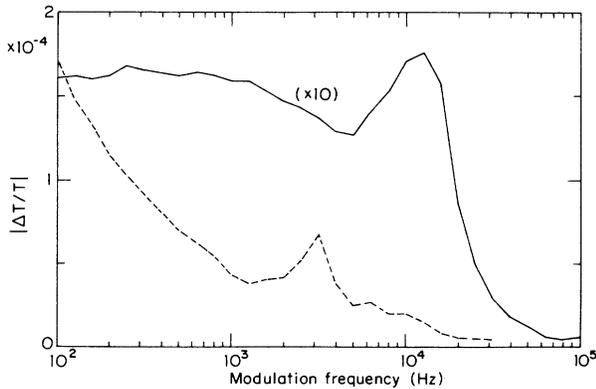


FIG. 4. Modulation frequency dependence of the midgap (solid) and near-gap (dashed) electroabsorption signal.

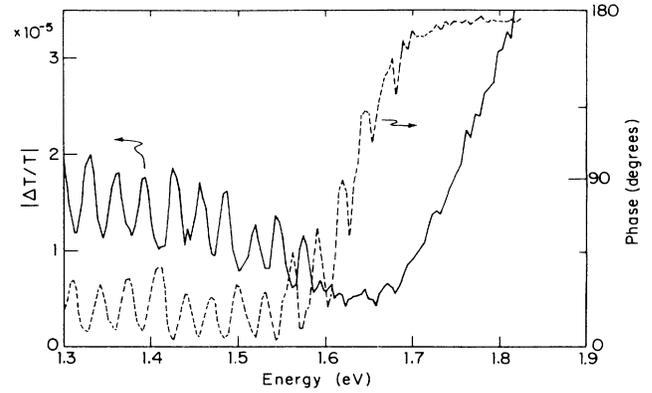


FIG. 5. Spectral dependence of the electroabsorption magnitude (solid) and phase (dashed).

Near 1.8 eV is the equivalent of the well-known band-edge EA peak,¹⁸ which will not be discussed further here. A minimum in the EA signal magnitude is observed near 1.6 eV, and the signal changes phase by nearly 180°. The midgap electroabsorption is found as a broad spectrum below 1.6 eV, and exhibits interference fringes as expected for this range of low absorption.³² The observed interference fringe spacing corresponds to the known sample thickness. We interpret these results as due to the injection of a space charge into the sample, which causes a modification of the absorption coefficient directly by changing the occupancy of the gap states near and above the equilibrium Fermi level E_F .

The modulation of the absorption coefficient $\Delta\alpha$ is determined from the experimental data using the relation

$$d\Delta\alpha = -\Delta T/T, \quad (2)$$

where d is the film thickness. If the above interpretation of the midgap EA is correct, then $\Delta\alpha$ may be related to the injected carrier density ρ by

$$\Delta\alpha = \frac{(2\pi e)^2 E}{3\rho_A n \hbar c} \rho [R_c^2 N_c (E_F + E) - R_v^2 N_v (E_F - E)], \quad (3)$$

where E is the photon energy (eV), the $R_{c,v}^2$ are dipole matrix elements squared (\AA^2), $N_{c,v}(E)$ is the density of states ($\text{eV}^{-1} \text{cm}^{-3}$), n is the index of refraction, e is the charge of the electron, and ρ_A is the density of a -Si:H. The matrix elements and densities of states are averaged over the range of the energies of the injected carriers, which in the case of quasisteady state can be assumed to be near and above the equilibrium Fermi level E_F . There are two contributions to $\Delta\alpha$ in Eq. (3). The positive term corresponds to new transitions of type 3 in Fig. 1. The negative term corresponds to the elimination of transitions of type 2 in Fig. 1. The average matrix elements of these two types of transitions need not be equal, and are not necessarily equal to matrix elements averaged in a different manner.

Because the measured midgap EA signal is negative, the second term of Eq. (3) dominates. Thus for this experiment, the dominant transition is a hole transition from just above the equilibrium Fermi level to the valence band. This result contradicts the assumption of several

authors that the dominant transition in a midgap optical absorption experiment is from the midgap to the conduction band. There is evidence from the phase dependence of photoconductivity,¹² and from the time dependence of photoinduced absorption,²³ demonstrating that the dominant absorption process at midgap energies is due to holes. A direct comparison with our results indicates an agreement with the conclusions of Okamoto *et al.* and Tauc.

We can attempt to use the magnitude of the EA to estimate the value of the matrix element difference in Eq. (3). The following assumptions are made: We use the geometrical capacitance of the samples to estimate the average injected carrier density

$$\rho = \kappa \epsilon V / d^2, \quad (4)$$

where κ is a numerical factor $1 \leq \kappa \leq 2$. A typical injected carrier density in these results is on the order of $5 \times 10^{13} \text{ cm}^{-3}$, which is small compared with the total number of available midgap states. We take an average value of 10^{22} cm^{-3} for the band density of states. We measure the EA signal to be about 2×10^{-5} . With these assumptions, we obtain an average dipole matrix element difference of approximately 10^3 \AA^2 . This value corresponds to an optical cross sectional difference σ_{opt} of about 10^{-15} cm^2 , where σ_{opt} is defined by

$$\sigma_{\text{opt}} = \Delta \alpha / \rho. \quad (5)$$

Within this method, the value for the matrix element is a lower bound to the individual matrix element R_v^2 . Note that this value does not depend on the density of midgap states in the sample.

These results are 2 orders of magnitude larger than an estimate of the near- and sub-band-gap dipole matrix element of about 10 \AA^2 obtained by a combination of measurements of photoemission, bremsstrahlung isochromat spectroscopy, and photothermal deflection spectroscopy (Jackson *et al.*³³) and 1 order of magnitude larger than an estimate of the optical cross section of about 10^{-16} cm^2 obtained by measurements of photocapacitance and deep-level transient spectroscopy (Gelatos *et al.*²⁶). This discrepancy must be considered with the following facts in mind.

First, the experiments of Jackson *et al.* and Gelatos *et al.* measure a different set of transitions averaged over a different energy range. Optical absorption experiments such as those performed by Jackson measure transitions 1 and 2, shown in Fig. 1, and photocapacitance measurements measure only transition 3. In contrast, this electroabsorption experiment looks at transitions 2 and 3, with transition 2 dominating. The energy range over which the matrix elements are averaged in the optical absorption experiments is the whole range allowed by energy conservation and Fermi factor filling. Indeed, this implies that the Fermi level position could influence the comparison of the deduced matrix elements by determining which states are available to the transitions being studied. The corresponding energy range in this experiment and in the pho-

tocapacitance experiment is determined by the energy range occupied by the injected carriers, which we have argued is restricted to the states just above the equilibrium Fermi level. The values of the averaged matrix elements will in general depend on the energy range and the nature of the transitions probed by the experiment.

A second explanation for the discrepancy lies in the estimation of the injected carrier density. The time dependence of the space-charge-limited current through a semiconductor containing traps is somewhat complicated (cf. the Many-Rakavy cusp³⁴). In particular, the ac capacitance $C(\omega)$ of a Schottky device under forward bias is enhanced over the geometrical capacitance C_0 by a value of

$$C(\omega \rightarrow 0) \approx C_0 / \omega_c t_T, \quad (6)$$

where ω_c is a typical free-carrier capture frequency and t_T is the carrier transit time.³⁵ The $(\omega_c t_T)^{-1}$ product can have a value exceeding 10^2 for amorphous silicon devices similar to the ones used for this study.³⁶ This capacitance enhancement, not accounted for in the analysis of the EA data, could increase the magnitude of the injected charge over the amount calculated from the geometric capacitance by a sufficient degree to account for the difference between the optical matrix element deduced above and the values obtained by Jackson *et al.* and Gelatos *et al.*

IV. SUMMARY

Electroabsorption by carrier injection in a forward-biased Schottky diode provides a new method for obtaining information on optical transitions involving localized states in amorphous silicon. It has the advantage over photoinduced absorption in that only one type of carrier is injected into the sample, simplifying the analysis. The midgap signal is found to be negative (induced transparency), and is shown to correspond to the elimination of transitions from the valence band to just above the Fermi level. The dominance of the valence band transitions in this experiment agrees with the deductions from photoinduced absorption and photoconductivity, and disagrees with the assumption of several authors. The transition matrix element deduced from the electroabsorption results disagrees with results from photocapacitance spectroscopy and other methods. The cause of this disagreement may lie in the differences in the transitions probed by the different techniques, or in the details of the estimate of the injected carrier densities.

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