Amorphous silicon-based superlattices*

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Abstract. Synthesis and some interesting properties of amorphous silicon-based superlattices are reported. Both quantum-well type and doping modulated structures are studied. Quantum confinement, phonon folding and persistent photoconductivity are some of the fascinating effects which are described and their current interpretations discussed.

Keywords. Amorphous silicon-based superlattice; quantum-well type structure; doping modulated structure; multilayers; quantum confinement; phonon folding; persistent photoconductivity; acoustic phonons; differential absorption; electroabsorption.

1. Introduction

The first synthesis of the quantum-well type structures of amorphous silicon (a-Si:H)-based multilayers was reported by Munekata *et al* (1983) and Abeles and Tiedje (1983). These multilayers are the amorphous analogs of crystalline superlattices and offer the exciting possibility of studying, in disordered materials, the many special effects already found in crystalline superlattices (for a review of crystalline superlattices, see, for example, Dohler (1983a, b)). Unlike crystalline superlattices, which can be fabricated mainly from materials with sufficiently matching lattices to allow epitaxial growth of one on the top of the other, the amorphous superlattices can be made from materials without any such considerations and can have interfaces that are essentially defect-free and atomically sharp. One of these layers is usually chosen to be a-Si:H, whereas the other is a different gap amorphous semiconductor, e.g. a-SiN_x:H, a-SiC_x:H, a-Ge:H, etc. In addition to these quantum-well type structures, amorphous analogs of the crystalline doping modulated type superlattices have also been fabricated, in which the alternate layers of a-Si:H are doped differently (*nnp*..., *nipini*..., etc). Studies on both the quantum-well type and the doping modulated type multilayers have yielded a wealth of information which is both fascinating and intriguing.

Many phenomena in a-Si:H have been ascribed to the potential fluctuations and heterogeneities. In multilayers such theories can be tested by varying the potential fluctuations and heterogeneities in a controlled manner.

Let us first consider a-Si:H-based quantum-well type heterostructures. These consist of alternate layers of two different band gap semiconductors A and B. a-Si:H is chosen as the smaller gap material A, because of its high electronic quality. Alloys Si with C, N or O etc. serve as the insulating layer B. It has been possible to obtain remarkably flat and atomically sharp interfaces (see, for example, Abeles and Tiedje 1983). Considering that the layers themselves are amorphous, it is surprising that these structures can be made at all. This tells us something about the growth

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kinetics of these layers, which are made by glow discharge (also called plasma-enhanced chemical vapour deposition) of silane gas (SiH_4) and its mixtures with appropriate gases.

Why does one obtain flat and atomically sharp interfaces for the a-Si:H based heterostructures? No one knows yet for sure, but it may be that these flat surfaces are a result of the simultaneous action of chemical deposition and chemical etching during the growth process (Tsai 1989) which is done at a much lower temperature (250°C) than the temperatures during the epitaxial growth (typically 800°C) of crystalline superlattices. Sharpness is, in fact, favoured by low-temperature deposition, because of smaller interdiffusion.

Given the fact that chemically sharp and flat heterojunction multilayers can be made, one explores the question, whether quantum effects can be observed in quasi-two-dimensional layers of a-Si:H sandwiched between layers of an insulator like, a- $\text{SiC}_x\text{:H}$, a- $\text{SiN}_x\text{:H}$, or a- SiO_x . One expects to see a shift of the luminescence peak to higher energies, when a-Si:H layer thickness decreases. This is because the quantized energy levels move to higher energies, as the well width decreases and approaches the size of the localized wavefunctions.

The quantum confinement of the extended states might not be expected because of the smallness of the coherence length of electron and hole wavefunctions. Yet, quantum confinement effects have been reliably identified up to a-Si:H layer thickness of as high as $50 \text{ \AA}$ (Hattori *et al* 1988a, 1989a, b). This raises fundamental questions about our understanding.

Space charge doping has been observed in heterostructures. However, mobility enhancement observed in crystalline superlattices is not found in amorphous analogs. One understands this because the mobility in amorphous semiconductors is not limited by ionized impurity scattering as it is in crystalline semiconductors at low temperatures.

An indication of resonant tunnelling through quantum-wells has been observed in amorphous semiconductors (Miyazaki *et al* 1987), but the effect is weak compared to crystalline semiconductors, presumably because of localized states in the barrier regions.

Phonon-folding in mini-Brillouin zones, created by the periodicity of the layers, has been observed by Raman scattering (Santos *et al* 1985). This is possible because the observations are made in the region of long wavelength phonons. For continuum approximation at long wavelengths, it does not matter whether the material is amorphous or crystalline. Gaps in the folded phonon dispersion relations have also been observed in phonon transmission experiments. These are interesting experiments because they yield information about elastic constants and sound velocity in amorphous layers.

Some studies have recently been made on quasiperiodic and random structures (Chen *et al* 1989; Itoh *et al* 1989). X-ray diffraction and Raman spectra agree with expectations. However, no new electronic effects are observed, probably because the coherence length of the carriers is too small, so that they cannot probe more than one layer.

Let us now turn our attention to the modulation-doped a-Si:H multilayers. Here the sharpness of the interfaces is not of much concern, because most of the time the properties investigated depend on the modulation of the space charge. In comparing these structures with crystalline counterparts, one is struck by the fact that there are very few experimental results reported on the crystalline ones. This

may be because the preparation of the modulation-doped crystalline superlattices requires a control of very small quantities of dopant elements with a high precision. Moreover, many of the dopants are undesirable in ultra-high vacuum environments. In contrast, doping modulation in a-Si:H multilayers can be achieved simply by switching between the mixtures of $\text{SiH}_4 + \text{PH}_3$ and $\text{SiH}_4 + \text{B}_2\text{H}_6$, for the two layers in a glow discharge reactor.

Charge separation effects similar to crystalline doping modulated superlattices, as predicted by Doehler (1983a), have been observed in amorphous doping modulated multilayers (Hundhausen *et al* 1984). These include an increased lifetime of carriers at liquid helium temperatures. At higher temperatures, however, new and different effects, e.g. persistent photoconductivity (PPC) are observed in amorphous multilayers (Kakalios and Fritzsche 1984; Agarwal and Guha 1985b). This state of enhanced conductivity (PPC) can be obtained by a brief exposure to light or electric field, or by quenching from a high temperature in the presence of a high electric field, and persists for several hours or days, even at room temperature. Such long periods suggest interesting novel effects.

Some of the other unexplained phenomena include a blue shift of the photoluminescence peak in the doping modulated a-Si:H multilayers. The photoluminescence peak of individual *p* and *n* layers is red-shifted with respect to the undoped layers. It is, therefore, surprising that a superposition of *n* and *p* layers gives a blue shift (Kakalios *et al* 1984).

In this article some selected properties of amorphous silicon-based quantum-well type as well as modulation-doped type multilayers (superlattices) are described, and our present understanding of results is examined.

2. Quantum-well type structures

2.1 Synthesis

These amorphous heterostructures are usually deposited in thin film form using a conventional glow discharge system. In the a-Si:H/a-SiN_x:H structures, for example, silane gas is subjected to a RF glow discharge, to produce a good quality a-Si:H layer (Knights 1976). For the other (a-SiN_x:H) layer a suitable mixture of silane and ammonia is used. In their original study Munekata *et al* (1983) used the glow discharge decomposition method for fabricating their a-Si:H/a-SiC_x:H structures, using silane gas for the a-Si:H layers and tetramethylsilane for a-SiC_x:H layers. Thus a desired structure can be produced by switching alternately between two mixtures of gases.

Other methods have also been used. For example, recently, Hazma *et al* (1989) fabricated a-Si:H/a-SiN_x:H multilayers having atomically flat interfaces by controlled nitradition of a-Si:H in an NH₃ plasma. Because the two amorphous layers in these heterostructures (also called composition-modulated multilayers) have different gaps, one expects quantum-well type structures.

2.2 Structure and interfaces

Both X-ray diffraction and transmission electron microscopy have been used to obtain information about the structure and interfaces in the amorphous

multilayers. X-ray diffraction provides information averaged over the sample volume. Transmission electron microscopy, performed on a cross-section of the film can, on the other hand, reveal localized defects. The X-ray diffraction pattern of a-Si:H/a-SiN_x:H multilayers shows that the first peak is quite narrow (within a factor of 2 of the value for an ideal, non-absorbing, weakly scattering, layered sample), which is indicative of the good quality of the structure. The electron micrographs also show that the layers are remarkably flat and uniform (Abeles and Tiedje 1983). Concerns about one layer contaminating the other have been expressed, and workers making multilayers in separate chamber systems have been worried about the effects that the switching-off of plasma might have on the interface. It turns out, however, that with careful handling it is not difficult to obtain multilayers with excellent structural characteristics (Abeles *et al* 1984; Miyazaki *et al* 1989).

The interfaces have also been studied by Raman scattering and XAFS. In a-Si:H/a-Ge:H multilayers, these studies have concluded that there is approximately only one layer of Si-Ge heterobonds across the interface (Persans *et al* 1985; Sette *et al* 1987). This is unusual, as most other multilayers have more defects, and extra hydrogen incorporated at the interface because of these. In a-Si:H/a-Ge:H, such an abrupt interface with a low density of interface defects is possible because of the close match between a-Si:H and a-Ge:H networks, the difference in the bond lengths being less than 5%. Raman scattering and X-ray reflectivity measurements have also been used to obtain useful information about interdiffusion and relaxation in a-Si:H/a-Ge:H multilayers (Persans 1988; Persans *et al* 1989).

2.3 Properties

Since amorphous multilayers contain a large number of repeating interfaces, they provide an opportunity to study these interfaces in greater detail. The composition and bonding at the interface have thus been studied by several other methods as well. These include the Rutherford back-scattering, ¹⁵N nuclear reaction and infrared absorption (Abeles *et al* 1986; Liu *et al* 1989). In addition, optical absorption (Kakalios *et al* 1984b; Tiedje and Abeles 1984; Wang and Fritzsche 1988; Liu *et al* 1989), photoluminescence (Wang *et al* 1988; Liu *et al* 1989; Guang *et al* 1989), electroluminescence (Tiedje and Abeles 1984), electroabsorption (Roxlo *et al* 1984a; Roxlo and Abeles 1986) and photoemission (Evangelisti *et al* 1984; Abeles *et al* 1987), have also provided information about the density of electronic states and band discontinuity at the interfaces. Roxlo and Abeles (1986), for example, find that their a-Si:H/a-SiN_x:H multilayers have about 10¹¹–10¹² states per cm², at the interface. They also found in a-Si:H the existence of a strong electric field at the interface, and transfer doping effects. An increase in optical gap (Agarwal and Guha 1985a; Ibaraki and Fritzsche 1984) and a blue shift of photoluminescence with decreasing a-Si:H thickness below 50 Å are also found (Wang and Fritzsche 1987, 1988; Lecomber *et al* 1985). The results are usually interpreted as quantum size effects, arising from the decrease in the quantum-well width. More recently, Hattori *et al* (1989a, b) measured longitudinal transport in a-Si:H/a-SiN_x:H multilayers by the time-of-flight method, and explained their observations in terms of tunnel hopping between neighbouring wells and multiple trapping of photoexcited electrons temporarily confined in each well. Although questions were initially raised as to whether all or some of these effects are caused by cross-contamination of

layers (LeComber *et al* 1985), later investigations appear to discount such doubts (Wang and Fritzsche 1988). In the following we discuss a few selected properties of these heterostructures.

2.3a Phonon-folding and frequency gaps in acoustic phonons: It is well known (Colvard *et al* 1980) that the artificial periodicity of the layers of two crystalline materials produces a folding of the dispersion relation for acoustic phonons into a mini-Brillouin zone of dimension $2\pi/d$, where d is the repeat distance. This folding manifests itself as additional lines in the low-frequency Raman spectrum of these crystalline superlattices. Since the coherence length of low-frequency acoustic phonons can extend over many periods, these effects have also been observed in amorphous silicon-based multilayers, despite the disorder present in individual layers (Santos *et al* 1985, 1986; Santos and Ley 1988; Nemanich 1986; Santos and Trodhal 1989). High-frequency phonons, on the other hand, have coherence lengths comparable to interatomic distances and therefore are not affected by the periodicity and the phonon spectrum of the superlattice in the high-frequency region ($\omega > 100 \text{ cm}^{-1}$) is simply the superposition of the spectra of individual layers, with an additional contribution from the interface (Persans *et al* 1985; Santos and Ley 1987). With decreasing frequency, the coherence length increases and the dispersion relation $\omega = \mathbf{v} \cdot \mathbf{q}$ for an unlayered material, gets folded in the direction of periodicity z , within an artificial mini-Brillouin-zone, $-\pi/d < q_z < \pi/d$.

Figure 1a shows such a folded dispersion relation for LA (solid lines) and TA (dashed lines) phonons calculated for an a-Si:H/a-SiN_x:H multilayer, with a period of 85 Å. Notice also the presence of the frequency gaps, in which no mode is permitted in an infinite superlattice. The experimentally observed Raman spectrum in figure 1b is obtained at room temperature in the backscattering mode for a multilayered sample, in which the a-Si:H and a-SiN_x:H thicknesses were approximately 47 and 36 Å, respectively (Santos and Ley 1988; Santos and Trodhal 1989). In this configuration, the TA modes are not coupled. The two peaks, marked ω_{-1} and ω_1 correspond respectively to the excitation modes from the first and the second folded LA branches. By studying the Raman scattering experiment in the other scattering geometry in which the incident and scattered light travel in the same direction (forward scattering), and by using a weakly absorbed light, so that the resulting spectrum is a superposition of the forward and back-scattered signal it has been possible to study the gaps in the dispersion relations as well. In addition to the a-Si:H/a-SiN_x:H multilayers, a-Si:H/a-Ge:H multilayers have also been studied and similar results have been found (Santos and Ley 1988).

The phonon transmission experiments clearly show the presence of gaps in the dispersion relation. The phonons cannot propagate through these gaps, thus giving rise to a number of minima, indicated by arrows in figure 1c, in the phonon transmission spectrum measured at 1 K, on a sample with the same multilayer geometry. The substrate was 4 mm thick crystalline silicon, and the phonons were generated by a superconducting $1 \times 1 \text{ mm}^2$ Al tunnel junction evaporated on the back side of the substrate and detected by a Sn-tunnel junction of the same area deposited on the superlattice on the front (Eisenmenger 1979). The solid arrows and the horizontal bars indicate the positions and the widths respectively of the frequency gaps. In addition to the gaps at the mini-Brillouin zone boundary, the mixing of LA and TA modes at the crossing points of LA and TA dispersion curves

Figure 1. a. Dispersion relation for LA and TA phonons for a-Si:H/a-SiN_x:H superlattice with period 83 Å, b. Raman spectrum and c. phonon transmission spectra. The solid arrows and the horizontal bars in c indicate the positions of frequency gaps for LA and TA phonons. The dashed arrows mark the position of the internal gaps. The geometry of the sample used for phonon transmission experiment is also shown (after Santos and Trodhal 1989).

gives rise to additional internal gaps within the zone as well. These are indicated by dashed arrows in this figure. Frequency gaps for transverse phonons in a-Si/a-SiO_x have also been studied using this method (Koblinger *et al* 1986).

2.3b Differential absorption and electroabsorption spectroscopy: Hattori *et al* (1988a 1989a-d) studied the optical transitions between the levels quantized by confinement in the a-Si:H/a-SiC_x:H multilayers using modulation techniques. By measuring differential absorption, as they modulated temperature, wavelength, or electric field, they were able to see the transitions as identifiable structures in their spectra. Figure 2 shows a thermoabsorption spectrum measured on four samples with different well thicknesses (a-Si:H) $L_w = 20, 30, 50$ and 500 Å. The a-SiC_x:H

Figure 2. Thermoabsorption (TA) measured as a function of photon energy for a-Si:H/a-SiC:H quantum wells with well widths L_w as shown and barrier width $L_b = 100$ Å. Arrows show the calculated sub-band transition energies (after Hattori *et al* 1989a).

barrier thickness was fixed at 100 Å, to avoid tunnelling between layers. For this measurement, two light sources were used. A 1 W/cm^2 light from an argon ion laser (488 nm), chopped at 5 Hz, was used as pump light, and a 5 mW/cm^2 white light was used as the probe. The transmitted light was sent through a monochromator and the transmission and its modulated component were measured using a photomultiplier and a lock-in amplifier. For $L_w < 50$ Å the transition between the quantized levels is clearly visible as steps, as indicated by the arrows. The results can be understood if one assumes a quantization of levels in the quantum wells as shown in figure 3. Starting with the Tauc model for optical absorption, which assumes no k conservation, the optical absorption α is given by

$$\alpha \hbar \omega = \sum_n A_n (\hbar \omega - E_n) U(\hbar \omega - E_n), \quad (1)$$

where $\hbar \omega$ is the photon energy, E_n the energy difference between the two levels labelled n in the conduction and valence bands, A_n represents the squared overlap integral, and $U(E)$ is a step function. The derivative with respect to either E_n or $\hbar \omega$ is given by

$$d(\alpha \hbar \omega)/d(\hbar \omega) \text{ or } d(\alpha \hbar \omega)/d(E_n) \propto \sum_n A_n U(\hbar \omega - E_n). \quad (2)$$

A comparison of equation (1) with (2) shows that unlike $\alpha \hbar \omega$ the differential

Figure 3. a. Quantized energy levels and wavefunctions and allowed transitions for the quantum well structures as shown; b. density of states (after Hattori *et al* 1989a).

spectrum ($\Delta\alpha \propto \omega$) is expected to exhibit a staircase form with steps at photon energies $\hbar\omega = E_n$. In deriving these equations, no effect of disorder, except the relaxation of k conservation rule has been considered. In disordered solids the quantized density of states is expected to be broadened. Hattori *et al* (1989a) carried out a self-consistent calculation using Abe-Toyozawa site-disordered model within the framework of the coherent potential approximation, to see the effect of disorder on the absorption spectrum of the quantum-well (QW) structures. The QW system with infinite potential barriers is modelled on a thin board consisting of a simple cubic lattice. The thickness of the board corresponds to the well width L_w . They consider Gaussian site disorder in their calculation. The ratio of the width (W) of the Gaussian to the full band width (B), i.e. W/B represents the degree of disorder. Their calculation shows that for $W/B = 6\%$, a staircase-type differential spectrum is expected, even when the layers making the quantum wells are amorphous. They find reasonable fits to their data, if they assume the effective electron and hole masses to be $0.3 m_0$ and m_0 respectively, in both sublayer regions. Here m_0 is the mass of a free electron. They also studied the effect of barrier height on their wavelength differential absorption spectra by changing the barrier material from a-SiC_x:H to a-SiO_x:H and found results in qualitative agreement with their model.

To measure the field-modulated absorption (or electroabsorption, as it is commonly called), Hattori *et al* (1989b) used the following structure: glass/transparent conductive oxide (TCO)/*n*-type a-Si:H (400 Å)/undoped MQW of a-Si:H and a-SiC_x:H/*n*-type a-Si:H (200 Å)/Al. RF plasma chemical vapour deposition was used to prepare multilayers. A combination of a dc field and an ac field at 1 kHz was applied between the Al and TCO electrodes perpendicular to the layers. A white light of intensity 1 mW/cm² was shone on the semitransparent substrate, and the reflected light was dispersed through a monochromator. The reflectance R and its modulated component ΔR were measured. The major contribution to ΔR comes from the change in the intensity of the probe light as it passes through the multilayers twice, as it is reflected by the Al electrode. The $-\Delta R/R$ signal, which is proportional to the field-induced change in the optical absorption (EA), obtained on a sample with $L_w = 20$ Å, $L_B = 100$ Å, 150 layers, with $V_{dc} = 0$ and $V_{ac} = 10$ V

Figure 4. Thermoabsorption (TA) and electroabsorption (EA) signals for a-Si:H/a-SiC:H, MQW, with $L_w=20$ Å, $L_B=100$ Å. The sample geometry for the EA spectrum is also shown which was measured with $V_{dc}=0$ and $V_{ac}=10$ V. E_1 is the theoretically calculated transition energy (after Hattori *et al* 1989b).

bias is shown in figure 4. Here, the EA signal exhibits a triangular shape. This is quite different from the step-like spectrum obtained on an identical MQW structure on glass, by differential thermoabsorption (dashed line marked TA, figure 4). This difference can be understood by noting that the effect of the applied voltage across the layer gives a stronger perturbation to the lower-lying QW states. Therefore, the lower sub-band transitions contribute to EA more significantly than the higher ones. This is in contrast to the TA spectra where each step is directly related to the absorption from each sub-band. Assuming that only the ground state E contributes significantly to the EA spectrum, the quantities E_1 and A_1 can be estimated in the small field limit, using perturbation theory. One obtains:

$$E_1(\Delta F) = E_1(0) - C_E \Delta F^2, \quad C_E > 0, \quad (3)$$

$$\text{and} \quad A_1(\Delta F) = A_1(0) - C_A \Delta F^2, \quad C_A > 0, \quad (4)$$

where ΔF is the field across the well. Since the electric field has an odd parity, equation (3) does not have a first-order term. Also C_E is positive, since the energy of the ground sub-band transitions experiences a red shift, due to the applied field. The sign of C_A in (4) can be understood by noting that A_1 , which is the square of the overlap integral, decreases as the field increases and the electron and hole wavefunctions spatially separate. Using (1), (3) and (4), it is easy to obtain the following expression for $\Delta\alpha$, for the EA spectrum:

$$\hbar\omega\Delta\alpha = (A_1 C_E - C_A(\hbar\omega - E_1)) U(\hbar\omega - E_1) \Delta F^2. \quad (5)$$

This equation clearly shows that a triangular EA spectrum sharply rising at $\hbar\omega = E_1$ and then decreasing with photon energy is expected.

The interband optical transitions in a-Si:H-based QW structures have been studied by others, but there are no reports of such spectral behaviours, as reported by Hattori *et al* and which have features that can easily be distinguished from the unlayered films. In particular, Roxlo *et al* (1984b) observed only a broad bump-like shape, commonly observed for the unlayered films (Hamakawa 1984), in contrast to the results presented above. The disagreement may result from the difference in the sample structures used in the two experiments. Roxlo *et al* (1984b) used a-Si:H and a-SiC:H of optical gaps 1.77 and 2.22 eV respectively, whereas Hattori *et al* used a slightly lower optical gap a-Si:H (1.75 eV) and a higher optical gap a-SiC_x:H (2.8 eV), for their structures. A larger difference between the gaps of the two materials may give a larger discontinuity in the band edges. Hattori *et al* estimated the band discontinuities in their films to be 0.8 and 0.25 eV for the conduction and valence band respectively from photoelectron emission experiments. In the QW structures of Roxlo *et al* the discontinuity in the valence band is probably close to zero, so that the QW effect for holes is negligible. Further, Roxlo *et al* chose equal widths for their barriers and wells. This may give a broadening in the conduction band levels, with decreasing layer thickness, as the wells may start interacting with each other. In addition, the quality of the insulating layer is also required to be good. This may be responsible for the failure to observe similar effects using a-SiN_x:H of higher band gap.

Although this may explain some of the discrepancies in the observations regarding the quantized levels, it is puzzling that these effects can be seen in quantum wells, whose widths are much larger than the mean-free path of the carriers in these amorphous materials.

3. Doping modulated multilayers

3.1 Synthesis and structure

The amorphous doping modulated multilayers are also made using a glow discharge system, similar to the one used for preparing the amorphous heterostructures. Here, silane is mixed with small quantities of phosphine or diborane (typically, 10–1000 ppm vol fraction) to achieve phosphorous (for *n*-type) or boron (for *p*-type) doping, respectively. In contrast it is much more difficult to make crystalline *nnp*... superlattices. This is because very small amounts of impurities are required to be controlled precisely. One needs four or more sources in a molecular beam epitaxy apparatus which are computer-controlled. The thickness of the individual layers in doping modulated a-Si:H multilayers is kept typically between 10 and 1000 Å, and the total thickness of the multilayer is usually about 1 μm. They have been made in single chamber systems without extinguishing the plasma, as well as in separate chamber systems where the contamination between the differently doped layers is minimal, but the plasma has to be switched off between the layers. The results are similar (Agarwal and Guha 1985a). Secondary ion mass spectroscopy, nuclear reaction techniques, and the scanning electron microscopy after selective etching (Hundhausen *et al* 1984; Hundhausen

and Ley 1985) have been used to confirm the correct doping profiles, which can be *npnp... nipi... nini, pipi... etc*

3.2 Properties

In the studies reporting the synthesis of the doping modulated amorphous silicon multilayers (Kakalios and Fritzsche 1984; Kakalios *et al* 1984b), two interesting properties were described. These were (i) a very long-lived large excess conductivity parallel to the layers after a brief exposure to light at room temperature (called PPC), and (ii) a blue shift in the peak of the photoluminescence, with respect to the peak positions of the unlayered *p*- and *n*-doped films. It is interesting to note that despite considerable work on these multilayers, the physics of these phenomena has not yet been completely understood. Other experiments include the study of carrier recombination after excitation with light at helium temperatures (Hundhausen *et al* 1984), and thermally-stimulated currents (Hundhausen 1989). We discuss here salient features of some of these effects.

3.2a Carrier recombination at helium temperatures: Hundhausen *et al* (1984) found that their *nipi...* multilayers behaved at temperatures below 20 K in a manner which could be understood in terms of charge separation of carriers and their subsequent trapping in the localized states. The sample having a *nipi...* structure was first exposed to a white (higher than band gap) light and then to a sub-band gap (IR) light, after a delay t_D , as shown in figure 5, while it is kept at 5 K. During exposure to the white light a photocurrent is observed. The dark current during the time t_D , when the light is off, is too small to be seen. But when the IR light is put on, a transient current, presumably coming from the release of the field separated trapped charges, is observed. The initial trap current I_0 is a measure of the concentration of trapped carriers, N_{sep} . The variation of I_0 with the thickness d

Figure 5. The upper two graphs show the timing and duration of the band gap and IR light used. The lower part is the observed photocurrent in the *nipi...* structure (after Hundhausen *et al* 1984).

Figure 6. Initial magnitude of the photocurrent induced by shining IR light after a delay of 1 s plotted as a function of layer thickness d . Dopings used were 10^{-4} parts for PH_3 and B_2H_6 vs SiH_4 for the n and p layers respectively (after Hundhausen *et al* 1984).

of the individual layers is shown in figure 6. The maximum response is observed for $d = 105 \text{ \AA}$, and is about a factor of 20 larger than both the unstructured compensated and the undoped samples.

These results can be understood in terms of a charge separation model shown in figure 7. The photoexcited carriers separated by the built-in field are trapped in different layers thereby reducing the built-in potential ΔU . The carrier density necessary to reduce ΔU to zero follows from the Poisson's equation:

$$d^2 U / dz^2 = (1/\epsilon_r \epsilon_0) \rho(z). \quad (6)$$

Assuming ρ to be independent of z and equal and opposite in the n and p layers, but zero in the i layer, equation (6) gives:

$$\rho = -\Delta U (4/3) \epsilon_r \epsilon_0 / d^2, \quad (7)$$

where d is the individual layer thickness. This shows that the maximum number of field-separated trapped charges in this model is proportional to d^{-2} .

The plot of the square root of the excess photocurrent (i.e. after subtracting from the observed current the contribution from the unlayered compensated sample with $d = 0$ (shaded area in figure 6)), against d turns out to be a straight line (not shown). This is in agreement with the model above.

In addition an increase in the lifetime of the separate carriers is also observed (Hundhausen *et al* 1984); an effect which has not been found in the PPC experiments at room temperature.

3.2b Persistent photoconductivity (PPC) in doping modulated multilayers: This effect was first observed by Kakalios and Fritzsche (1984). Figure 8 shows the effect of light exposures of different durations on the conductivity of an nnp ... multilayer, measured parallel to the layers at room temperature, as reported by Agarwal and Guha (1985a). The sample which had $6n$ and $5p$ layers of equal thickness of $275 \text{ \AA}$ was exposed to heat-filtered light ($h\nu > 1.5 \text{ eV}$) from a tungsten-halogen lamp. The sample goes from the lower conductivity-annealed state (state A) to a higher conducting state (PPC state) after exposure. The conductivity in the

Figure 7. Band diagram of a $nipi\dots$ superlattices in real space Z ; a. At the beginning of excitation by light from the ground state. b. After maximum excitation, with spatially separated trapped charges (after Hundhausen *et al* 1984).

PPC state is about two orders of magnitude larger than the state A after the first exposure, and increases further by a smaller amount with each successive exposure. This PPC state is stable for several days at room temperature, and cannot be erased by exposure to sub-band gap light. Annealing to above 450 K brings back the annealed state A. Similar results were obtained by other workers as well (Hundhausen and Ley 1985; Su *et al* 1985).

Attempts to explain this effect in terms of a charge-separation model, similar to that of crystalline-doping superlattices, have not met with success. In this model, the built-in fields separate the photogenerated carriers. Thus the electrons go to the n layer and the holes to the p layer, where they get trapped in localized states. This brings the quasi-fermi levels in these layers closer to the band edges thereby enhancing the conductivity. The main difficulty with this explanation is that the PPC lasts much longer than is expected from this model. Also, from the position of the fermi level in the n and p layers the built-in field can be estimated. For the intensities of light used it should be possible to generate enough charges to neutralize this field in less than a millisecond, and reach a saturated PPC state. In contrast PPC continues to rise for exposures much longer than 1 second. Figure 9 shows the rise of PPC as a function of exposure time, for a few samples with varying doping concentrations of the p layer (Agarwal and Guha 1985b).

Figure 8. Persistent photoconductivity in a $6n/5p$ multilayered sample, with $\text{PH}_3/\text{SiH}_4 = 40$ ppm and $\text{B}_2\text{H}_6/\text{SiH}_4 = 200$ ppm measured at room temperature. i_d is the dark current in the annealed state A. Effect of successive light exposures of t_x seconds as shown results in a higher dark conductivity (PPC state) (after Agarwal and Guha 1985c).

Figure 9. Effect of variation of doping concentration in the p -layer on the PPC of a $6n/5p$ modulation-doped multilayer PPC is defined to be the ratio of the excess dark current and annealed dark current ($\Delta i/i_d$) measured after switching off the light (after Agarwal and Guha 1985b).

The internal fields can be varied by varying the doping concentrations or the individual layer thicknesses. Although the effect of thickness variation on PPC seems to agree with the charge-separation model (Kakalios and Fritzsche 1985), the

variation of doping does not (Agarwal and Guha 1985b). Table 1 shows the results for samples in which the fermi level in the n -layers is kept fixed at $\Delta E_{fn} = 0.18$ eV, and the fermi level in the p -layer is varied. As the difference of the fermi levels in the two layers (ΔE_{fn-p} , which is a measure of the internal field) increases, the PPC decreases. In addition, no PPC is observed in samples containing only one kind of dopant, viz. $nini...$ and $pipi...$ as first reported by Agarwal and Guha (1985b,c), and also found by others. This is contrary to expectation, if charge separation alone were responsible for PPC.

Kakalios and Fritzsche (1985) found that PPC at room temperature is higher for higher temperatures of exposure T_e (see figure 10, dashed curve). Similarly, Hundhausen and Ley (1985) reported that the amount of light necessary to generate a given level of PPC in their $nipi...$ multilayers, decreases exponentially with

Table 1. n -layer: $\text{PH}_3/\text{SiH}_4 = 80$ ppm, $\Delta E_{fn} = 0.18$ eV, p -layer doping is as shown (after Agarwal and Guha 1985a).

p -layer Boron (ppm)	ΔE_{fp} (eV)	$\Delta E_{fn-p} \approx 2.0 - (\Delta E_{fp} + \Delta E_{fn})$ (eV)	PPC = $\Delta i/i_d$
4	0.86 (n -type)	0.58	320
40	0.94	0.78	50
200	0.75	0.97	17
400	0.70	1.02	6
1000	0.60	1.12	< 1

Figure 10. Comparison of the plots of logarithms of excess conductivity vs reciprocal of exposure temperature for a compensated (full curve) and a multilayered (dashed curve) $a\text{-Si:H}$ (after Agarwal and Guha 1987).

increasing temperature of exposure. They point out that this thermally-activated generation of PPC is incompatible with the charge separation model.

It now appears that PPC in a-Si:H is not limited to the modulation-doped layered structures (Agarwal and Guha 1987). A similar effect was found by Mell and Beyer (1983) in unlayered compensated a-Si:H, even before PPC was discovered in the doping modulated multilayers. After some conflicting reports that this effect disappears in compensated films when their surface is etched, Agarwal and Guha (1985b, 1986) established that it is a bulk phenomenon, and that the *nnpn*... multilayers and the compensated films behave very similarly. Figure 11 compares PPC in a layered sample to a compensated sample, having the same amount of dopant gases during preparation (Agarwal and Guha 1985a). The similarity is quite striking. These authors also found that in both cases a large PPC is observed only in those samples, which are *n*-type (Agarwal and Guha 1985b). There are other similarities between the PPC in the compensated and modulation-doped multilayers. For instance, the annealing behaviour of PPC in the two cases is quite similar (Agarwal and Guha 1986) (figure 12). Further, as seen in figure 10 the effect of change of the exposure temperature on the PPC of a multilayered sample (dashed curve) is quite similar to that on the compensated sample (full curve).

Amongst these similarities, it is perhaps important to note a few differences between the compensated and the multilayered samples. For graphite electrodes the compensated samples are usually ohmic up to the highest measuring fields (10^3 V/cm), but the layered films are non-ohmic for electric fields as small as 10 V/cm (Agarwal and Guha 1985b). These non-ohmic effects, which are most severe with graphite electrodes, are found to be smaller, if nichrome electrodes are used. The best results are obtained for magnesium electrodes. Hamed (1989) was able to get ohmic behaviour up to 500 V/cm in multilayered films using evaporated Mg electrodes. Also, scratching the multilayers before depositing the electrodes does not seem to have much of an effect. Here, special care has to be taken to prevent Mg electrodes from getting oxidized and peeling-off, by covering them with another metal, immediately after deposition. Also, although figure 11 shows a case where

Figure 11. Comparison of PPC measured at room temperature for a compensated and a multilayered sample, both having identical dopant gas levels (after Agarwal and Guha 1985b).

Figure 12. Comparison of annealing of PPC at various temperatures between a layered (full curve) and a compensated sample (after Agarwal and Guha 1986).

PPC in the similarly doped layered and compensated samples is similar, this is not true in all cases. PPC effect is generally smaller in the compensated samples than in the layered ones for similar doping concentrations.

PPC in undoped and singly-doped a-Si:H layers (Choi *et al* 1986) is generally much smaller than in the doping modulated and compensated a-Si:H, and also lasts for a shorter time. There is a report of a large PPC in boron-doped a-Si:H, but it has been attributed to the presence of a surface oxide layer, as the effect is considerably reduced after etching (Aker and Fritzsche 1983).

In this connection, it is interesting to note that PPC has been observed in the past in a great variety of crystalline and amorphous materials (Sheinkman and Shik 1976). In most cases it can be attributed to the macroscopic potential fluctuations associated with inhomogeneities which give rise to charge separation. Indeed PPC in the unlayered films is larger if they are more heterogeneous. The short relaxation times observed allowed Choi *et al* (1986) to interpret their results in terms of charge separation in the potential fluctuations caused by inhomogeneities and subsequent trapping in traps, without any barriers.

The similarity of PPC effect in the modulation-doped and compensated a-Si:H suggests that it might have a common origin in the two cases. It has been proposed that it arises because of the presence of special centres in these films, situated in the upper half of the gap that acts as hole traps and not as recombination centres. Since the samples that show a large PPC are *n*-type, some or most of these centres may be filled in the annealed state. Upon shining light, they capture holes leaving the electrons behind to conduct. The slow rise of PPC with exposure and the observation that the creation of PPC is thermally activated requires one to assume a barrier for the capture of holes.

The absence of PPC in *nini*... and undoped and *p*-doped a-Si:H rules out the possibility of these centres arising from Si and P. No PPC is observed in *pipi*... or in B-doped a-Si:H. This, however, cannot be used to rule out the B-related defects, since these samples are *p*-type, and the centres, even if present, would be empty (Agarwal and Guha 1985b, 1986, 1987). Thus these centres may arise either from phosphorous complexes or from the presence of boron alone.

Recently, Hamed and Fritzsche (1989a, b) proposed a simple model to explain the PPC in terms of the well-known Staebler-Wronski effect (Staebler and Wronski 1977). In their model, the brief light exposure created metastable Staebler-Wronski defects in the *p*-type material shifting the fermi level in this layer towards the mid-gap, but does not have much effect on the *n*-layer. This is because longer light exposures are needed to create a Staebler-Wronski effect in the *n*-type material. This results in a net shift of the equilibrium fermi level of the whole multilayer towards conduction band. If the multilayer is *n*-type, this would give rise to an increased conductance or PPC, in agreement with the observations. The absence of PPC in *nini*... layer can be explained by noting that it does not contain boron whereas for the *pipi*... layers it can be argued that they do not show PPC, because they are *p*-type. This also explains the saturation of the PPC effect at longer exposures, since now the Staebler-Wronski effect in the *n*-layer, starts pushing the fermi level away from the conduction band edge. Indeed, a smaller PPC effect is observed at long enough exposures (Agarwal and Guha 1985a).

If this model is correct, the doping-modulated multilayers and the *p*-type layers should change in conductance at a similar rate, upon light exposure and upon annealing, the sign of the change being opposite for the two. This is indeed observed. One finds that the conductance of both the *p*-type film and the multilayer changes rapidly for small exposures, whereas that of the *n*-type film changes significantly only for exposures greater than 100s (figure 13, Hamed and Fritzsche 1989a). The annealing temperatures of the conductance of the *p*-layer and of the PPC of a multilayer are found to be the same in both cases.

This model can also explain the PPC in compensated films. Here the

Figure 13. PPC measured as the ratio of the conductance G_b , measured 5 min after exposure to G_a (conductance in the annealed state) for singly-doped and multilayered a-Si:H (after Hamed and Fritzsche 1989b).

compositional and structural inhomogeneities associated with boron-doping can produce the same effect as multilayers.

Thus this model ascribes the PPC effect to the creation of the 'Staebler-Wronski defects' associated with boron alone. Also, it does not require the presence of built-in fields or charge separation. If the defects were phosphorous boron complexes, it would imply that the PPC in compensated films should be higher than the modulation-doped multilayers, contrary to observations. Using this model it is easier to understand why PPC is smaller in compensated films as compared to the modulation-doped ones, because there are whole boron layers in the latter case, whereas one has to depend on the structural heterogeneities to get PPC in the former. However, one observation that this model has not explained so far is that an extremely small concentration of boron can give the highest PPC. Agarwal and Guha (1985b) reported that the highest PPC was obtained for a multilayer in which the boron-doped layer was doped so lightly that it was *n*-type with an activation energy of 0.86 eV (see table 1). This puts the fermi level of this B-doped layer in the middle or slightly above the mid-gap, thus leaving no room for the effect to take place in this model.

4. Summary and discussion

The method of plasma-enhanced chemical vapour deposition allows one to synthesize a-Si:H based quantum-well type structures with atomically sharp and abrupt interfaces. The deposition conditions, in particular the low substrate temperature, favour the sharp interfaces. Also, the amorphous nature of the layers allows one to choose a wide variety of materials without worrying about lattice matching. Thus the preparation of amorphous quantum-well type structures is relatively easier than their crystalline counterparts. Further, the doping-modulated type amorphous structures are also easier to make than the crystalline ones.

Among the several interesting properties of a-Si:H-based heterostructures discussed here is the phonon folding. This arises because of the introduction of artificial periodicity by the presence of two different amorphous layers at regular intervals. The phonon dispersion relations get folded into a mini Brillouin zone which result in peaks in Raman scattering and phonon transmission experiments. It should be noted that the measurements are done in the region of long wavelength phonons. This allows one to use continuum approximation even though the individual layers are amorphous. These experiments have also given information about the elastic constants and sound velocity in amorphous materials.

The situation is quite different with quantum confinement effects in amorphous heterostructures. These are expected when the size of the quantum wells becomes comparable to or smaller than the mean-free path of carriers. The fact that quantum confinement effects have been observed for well widths much larger than the mean-free path of carriers is quite puzzling. In order to observe the transitions between the quantized levels in the quantum wells by differential absorption and electroabsorption Hattori *et al* (1989b) had to use wide band gap good quality a-SiC_x:H as the barrier material. Thus the absence of the signature of quantum effects in other studies can be understood.

The interpretation of these results in terms of quantum confinement is however still open to question. This interpretation requires that the mean-free path of the

carriers (both electrons and holes) be larger than 50 Å, which is the well width below which such effects have been observed. The mean-free path for electrons is estimated to be no more than 10 Å. For holes it is expected to be even smaller perhaps by as much as a factor of 10. Although the Gaussian site disorder model used by Hattori *et al* (1989a) and described briefly in § 2.3b appears to provide a good fit to their data, they do not address the problem associated with the small mean-free paths. Raikh *et al* (1990) theoretically estimated the smallest mean-free path necessary to observe such effects. They have studied the effect of quantum confinement on the density of states in amorphous semiconductors. They treat disorder as a random potential (white noise) with zero correlation length. They find that peaks in differential spectra may be observed for well widths, which are one and a half times the mean-free path of the carriers. This somewhat reduces the disagreement between theory and experiment but does not completely explain it. This shows that further work is needed to fully understand the observed quantum confinement effects in these amorphous heterostructures.

Let us now consider the interesting effects in doping modulated multilayers. The carrier recombination at helium temperatures shows charge separation effects due to built-in fields. The persistent photoconductivity observed at room temperature has defied a satisfactory explanation so far. Clearly, the explanation in terms of charge separation model is not tenable. The similarity of the effect in the layered and compensated films and its absence in *nini...* and *pipi...* layers led Agarwal and Guha (1985a) to postulate that it is caused by special centres which are produced by the formation of phosphorous boron complexes. They performed an experiment in an attempt to resolve the question whether these special centres are phosphorous boron complexes or are related to the presence of boron alone (Agarwal and Guha 1986). A p^+pnn^+ sandwich structure was prepared in which the n and p layers were adjusted to give a large PPC. The p^+ and n^+ layers provided top and bottom ohmic contacts respectively. In another sandwich structure the n layers were replaced by Al to give a Schottky contact to the p -layer in the p^+p Al sandwich. In the second case the bending of the bands caused by the Schottky barrier was expected to fill the special centres, if any, in the p -layer. If the PPC is related to the formation of PB complexes, and not to B alone, the properties of the junction would change upon exposure to light only in the first sample. Unfortunately, no change was found upon measuring IV , V_{oc} , and $C(\omega)$ in the annealed and light exposed state in either case. The change in junction properties is perhaps too small to be detected by these experiments. More experiments are, therefore, needed to resolve this issue. One could, for example, change the doping of the n layer from phosphorus to arsenic, or see if the presence of an accumulation layer on a boron-doped film (by exposing it to moisture, for example) would give rise to PPC.

It has been proposed (Kakalios and Fritzsche 1984; Hundhausen and Ley 1985) that the barriers for the release of electrons from these special centres arise from a strong electron lattice coupling, in analogy to the DX centres in $Ga_{1-x}Al_xAs$. So far, however, no independent evidence for such a coupling exists. It is also possible that these centres are not present in the as-deposited films, but are created upon illumination. The creation kinetics have not been worked out so far. On the basis of available data, all one can say is that it must involve both the boron and phosphorous atoms or the boron atoms alone (Agarwal and Guha 1985b, 1987).

More recently, Hamed and Fritzsche (1989a) ascribed this effect as arising from

metastabilities similar to the Staebler-Wronski (S-W) effect. This model is supported by the fact that the doping modulated and p -type layers change their conductance at a similar rate but in opposite directions upon exposure to light and upon annealing, whereas the n -layers do not change much upon light exposure. Since the effect of light exposure is to shift the Fermi level towards the centre, this implies an increase in conductivity of the nnp ... multilayer for short exposures if the multilayer is n -type, thus explaining the observed results.

To conclude, we have discussed some interesting properties of the a-Si:H-based multilayers. Some of these can be understood as arising from the introduction of periodic placement of layers of two different materials, or charge separation effects, similar to those found in crystalline superlattices. Others will have to wait for further investigations before they are completely understood.

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